Ensemble Prediction of Air Quality Using the WRF/CMAQ Modeling System for Health Effects Studies in China

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Abstract

Accurate exposure estimates are required for health effects analyses of severe air pollution in China. Chemical transport models (CTMs) are widely used to provide spatial distribution, chemical composition, particle size fractions, and source origins of air pollutants. The accuracy of air quality predictions in China is largely affected by the uncertainties of public available emission inventories. The Community Multiscale Air Quality (CMAQ) model with meteorological inputs from the Weather Research and Forecasting (WRF) model were used in this study to simulate air pollutants in China in 2013. Four sets of simulations were conducted with four different anthropogenic emission inventories, including the Multi-resolution Emission Inventory for China (MEIC), the Emission Inventory for China by School of Environment at Tsinghua University (SOE), the Emissions Database for Global Atmospheric Research (EDGAR), and the Regional Emission inventory in Asia version 2 (REAS2). Model performance was evaluated against available observation data from 422 sites in 60 cities across China. Model predictions of O_3 and $PM_{2.5}$ generally meet the model performance criteria, but performance difference exists in different pollutants and regions among inventories. Ensemble predictions were calculated by linearly combining the results from different inventories to minimize the sum of the squared errors between the ensemble results and the observations from all the cities. The ensemble concentrations show improved agreement with observations in most cities. The mean fractional bias (MFB) and mean fractional errors (MFE) of the ensemble annual $PM_{2.5}$ at the 60 cities are -0.11 and 0.24, respectively, which are better than the MFB (-0.25 – -0.16) and MFE (0.26 – 0.31) of individual simulations. The ensemble annual daily maximum 1-hour O_3 (O_3-1h) concentrations are also improved, with mean normalized bias (MNB) of 0.03 and mean normalized errors (MNE) of 0.14, compared to MNB of 0.06 – 0.19 and MNE of 0.16 – 0.22 of the individual predictions. The ensemble predictions agree better with observations with daily, monthly, and annual averaging times in all regions of China for both $PM_{2.5}$ and O_3-1h. The study demonstrates that ensemble predictions by combining predictions from individual emission inventories can improve the accuracy of predicted temporal and spatial distributions of air pollutants. This study is the first ensemble model study in China using multiple emission inventories and the results are publicly available for future health effects studies.

Key words: chemical transport model; emission inventory; ensemble; China; $PM_{2.5}$

1. Introduction

A significant portion of the population in China has been exposed to severe air pollution in recent decades as the consequence of intensive energy use without efficient control measures. Based on ambient air pollution data published by the China National Environmental Monitoring Center (CNEMC), most of the major cities are in violation of the Chinese Ambient Air Quality Standards grade II standard (35 μg m^{-3}) for annual average particulate matter with diameter of 2.5 μm or less ($PM_{2.5}$) (Zhang and Cao, 2015;Wang et al., 2014b), with a mean population weighted $PM_{2.5}$ concentration of over 60 μg m^{-3} during 2013-2014. Long-term exposure to such high levels of $PM_{2.5}$ greatly threatens public health in China. Recent studies have suggested that approximately more than one million premature deaths can be attributed to outdoor air pollution each year in China (Lelieveld et al., 2015a;Liu et al., 2016;Hu et al., 2017a).

Accurate exposure estimates are required in health effects studies. Ambient air quality is usually measured at monitoring sites and used to represent the exposure of population in the surrounding areas. However, a routine central monitoring network in China has just been established since 2013, and is still limited in spatial coverage and lack of detailed information of the chemical composition, PM size fractions, and source origins of air pollutants. Chemical transport models (CTMs) have been widely used in health effects studies to overcome the limitations in central monitor measurements for exposure estimates (Philip et al., 2014;Lelieveld et al., 2015b;Liu et al., 2016;Laurent et al., 2016a;Laurent et al., 2016b;Ostro et al., 2015). However, the accuracy of the predictions from CTMs is largely affected by the accuracies of the emission inventories (Wang et al., 2010), meteorological fields (Hu et al., 2010), and numerical solutions to the equations that describe various atmospheric processes (Hu et al., 2006;Yu et al., 2005). Emission inventories are indispensable tools for a wide range of environmental activities from management of chemicals to the prevention of air pollution. Several emission inventories have been created to cover China. Different emission inventories focus on specific geographical regions in the urban, regional (Zhao et al., 2012;Zhang et al., 2008), national or continental (Zhang et al., 2009;Kurokawa et al., 2013) scales; and/or focus on pollutants from individual (Su et al., 2011;Ou et al., 2015) and specific sectors (Zhao et al., 2008;Xu et al., 2017).

Despite the great efforts in improving the accuracy of emission inventories in China, large uncertainties still remain. Generally, the emissions of pollutants are estimated as the product of activity levels (such as industrial production or energy consumption), unabated emission factors (i.e. mass of emitted pollutant per unit activity level), and the efficiency of emission controls. Large uncertainties are associated with activity levels, emission source fractions, and emission factors (Akimoto et al., 2006;Lei et al., 2011a). The uncertainties are especially significant for some pollutants, such as ammonia (NH_3) and volatile organic compounds (VOCs). For example, for a Pearl River Delta (PRD) inventory in 2006, SO_2 emission has low uncertainties of -16%~21% from power plant sources quantified by Monte Carlo simulations, while NO_x has medium to high uncertainties of -55%~150% and VOC, CO, and PM have even higher uncertainties (Zheng et al., 2009). For an inventory for the Yangtze River Delta (YRD) region, the overall uncertainties for CO, SO_2, NOx, PM_{10}, $PM_{2.5}$, VOCs, and NH_3 emissions are ±47.1 %, ±19.1 %, ±27.7 %, ±117.4 %, ±167.6 %, ±133.4 %, and ±112.8 %, respectively (Huang et al., 2011). A comprehensive quantification study by Zhao et al. (2011) using Monte Carlo simulations showed that the uncertainties of Chinese emissions of SO_2, NOx, $PM_{2.5}$, BC, and OC

in 2005 are −14%~13%, −13%~37%, −17%~54%, −25%~136%, and −40%~121%, respectively.

The uncertainties in emission inventories are carried into CTMs simulations, leading to biases in air quality predictions, which need to be carefully evaluated to identify the useful information for health effects studies (Hu et al., 2016b;Hu et al., 2014c;Hu et al., 2014b;Hu et al., 2015b;Tao et al., 2014). An evaluation of one-year air pollutants predictions using the Weather Research and Forecasting (WRF) / Community Multi-scale Air Quality (CMAQ) modeling system with the Multi-resolution Emission Inventory for China (MEIC) has been reported (Hu et al., 2016a). The model predictions of O_3 and $PM_{2.5}$ generally agree with ambient measured concentrations, but the model performance varies in different regions and seasons. In some regions, such as the northwest of China, the model significantly under-predicted $PM_{2.5}$ concentrations.

The technique of ensemble is often used to reduce uncertainties in model predictions by combining multiple sets of predictions. This technique has been widely used in the climate predictions (Murphy et al., 2004;Tebaldi and Knutti, 2007), and recently adopted in air quality predictions (Delle Monache et al., 2006;Huijnen et al., 2010). A recent study has compared a few anthropogenic emission inventories in China during 2000-2008 (Saikawa et al., 2016), but detailed evaluation of model results based on these inventories have not been performed. The methods to utilize the strength of different emission inventories to get improved air quality predictions for China have not been reported in the literature. The aim of this study is to create an improved set of air quality predictions in China by using an ensemble technique. First, four sets of one-year air quality predictions were conducted with the WRF/CMAQ modeling system with four different anthropogenic emission inventories for China in 2013. In addition to MEIC, the three other emission inventories are the Emissions Database for Global Atmospheric Research (EDGAR), Regional Emission inventory in Asia version 2 (REAS2), and Emission Inventory for China developed by School of Environment at Tsinghua University (SOE). The model performance on $PM_{2.5}$ and O_3 concentrations in 2013 with different emission inventories was then evaluated against available observation data in 60 cities in China. The differences among air quality predictions were also compared and identified. Finally, an ensemble technique was developed to minimize the bias of model predictions and to create improved exposure predictions. To the authors' best knowledge, this is the first ensemble model study in China using multiple emission inventories. The ensemble predictions of this study are available for public health effects analyses.

This paper is organized as follows. The CMAQ model, emissions and other inputs for the model, observational datasets used for model performance evaluation, and the method for ensemble calculation are described in Section 2. Section 3 discusses the model performance on gaseous and particulate pollutants simulated with four emission inventories, as well as the performance of the ensemble predictions in different regions/cities and with different averaging times. At last, the major findings are summarized in the Conclusion section.

2. Method

2.1 Model description

In this study, the applied CMAQ model is based on CMAQ v5.0.1 with changes to improve the model's performance in predicting secondary organic and inorganic aerosols. The details of these changes could been found in previous studies (Hu et al., 2016a;Hu et al., 2017b) and the references therein, therefore only a brief description is summarized here. The gas phase photochemical mechanism SARPC-11 was modified to better treat isoprene oxidation chemistry (Ying et al., 2015;Hu et al., 2017b). Formation of secondary organic aerosol (SOA) from reactive uptake of dicarbonyls, methacrylic acid epoxide, and isoprene epoxydiol through surface pathway (Li et al., 2015;Ying et al., 2015) was added. Corrected SOA yields due to vapor wall-loss (Zhang et al., 2014) were adopted. Formation of secondary nitrate and sulfate through heterogeneous reactions of NO_2 and SO_2 on particle surface (Ying et al., 2014) was also incorporated. It has been shown that these modifications improved the model performance on secondary inorganic and organic $PM_{2.5}$ components.

2.2 Anthropogenic emissions

The CMAQ model was applied to China and surrounding countries in East Asia using the horizontal resolution of 36-km. The modeling domain is shown in Figure 1. The anthropogenic emissions are from four inventories: MEIC, SOE, EDGAR, and REAS2. MEIC was developed by a research group in Tsinghua University (http://www.meicmodel.org). Compared with other inventories for China, e.g. INTEX-B (Zhang et al., 2009) or TRACE-P (Streets et al., 2003), the major improvements include a unit-based inventory for power plants (Wang et al., 2012) and cement plants (Lei et al., 2011b), a county-level high-resolution vehicle inventory (Zheng et al., 2014), and a novel NMVOC speciation approach (Li et al., 2014). The VOCs were speciated to the SAPRC-07 mechanism. As the detailed species to model species mapping of the SAPRC-11 mechanism is essentially the same as the SAPRC-07 mechanism (Carter and Heo, 2012), the speciated VOC emissions in the MEIC inventory were directly used in the simulation.

The SOE emission inventory was developed using an emission factor method (Wang et al., 2011;Zhao et al., 2013). The sectorial emissions in different provinces were calculated based on activity data, technology-based and uncontrolled emissions factors, and penetrations (fractions of pollutants not collected) of control technologies. Elemental carbon (EC) and organic carbon (OC) emissions were calculated based on $PM_{2.5}$ emissions and their ratios to $PM_{2.5}$. The sectorial activity data and technology distribution were obtained using an energy demand modeling approach with various Chinese statistics and technology reports. More details, including the spatio-temporal distributions and speciation of NMVOC emissions, can be found in previous publications (Zhao et al., 2013;Wang et al., 2011;Bin et al., 2013). Since MEIC and SOE emission inventories only cover China, emissions from outside China countries and regions were based on REAS2 (Kurokawa et al., 2013).

The version 4.2 of EDGAR emission (http://edgar.jrc.ec.europa.eu/overview.php?v=42) has a spatial resolution of $0.1^o \times 0.1^o$. The EDGAR inventory contains annual emissions from different sectors based on IPCC designations. REAS2 has a spatial resolution of $0.25^\circ \times 0.25^\circ$ for the entire Asia. The inventory contains monthly emissions of pollutants from different source categories. Saikawa et al. (2016) compared the major features of different anthropogenic emission inventories for China. Detailed information regarding these inventories can be found in the publications presenting them. Table S1 shows the total emissions of major pollutants within

China in a typical workday of each season. In general, large differences exist among different inventories for China. MEIC has the highest CO emissions in winter while REAS2 has the highest in other seasons. MEIC has the highest NO_x emissions while REAS2 has the highest emissions of VOCs in all seasons. EDGAR predicts the highest SO_2 emissions, which are approximately a factor of two higher than those estimated by SOE. SOE has highest NH_3 emissions while EDGAR has much lower NH_3 emissions than the other three. EDGAR also has lowest EC and OC emissions, but the total $PM_{2.5}$ emissions are the highest. Standard deviations indicate that winter has the largest uncertainties for all species except SO_2 and NH_3. Winter has the smallest SO_2 uncertainties while summer has the largest NH_3 uncertainties.

All the emissions inventories were processed with an in-house program and re-gridded into the 36-km resolution CMAQ domain when necessary. Representative speciation profiles based on the SPECIATE 4.3 database maintained by U.S. EPA were applied to split NMVOC of EDGAR and REAS2 into SAPRC-11 mechanism and $PM_{2.5}$ of all inventories was split into AERO6 species. Monthly emissions were temporally allocated into hourly files using temporal allocation profiles from previous studies (Chinkin et al., 2003; Olivier et al., 2003; Wang et al., 2010a). More details regarding EDGAR can be found in Wang et al. (2014a), while those for REAS2 can be found in Qiao et al. (2015).

2.3 Other inputs

The Model for Emissions of Gases and Aerosols from Nature (MEGAN) v2.1 was used to generated biogenic emissions (Guenther et al., 2012). The 8-day Moderate Resolution Imaging Spectroradiometer (MODIS) leaf area index (LAI) product (MOD15A2) and the plant function type (PFT) files used in the Global Community Land Model (CLM 3.0) were applied to generate inputs to MEGAN. The readers are referred to Qiao et al. (2015) for more information. Open biomass burning emissions were generated using a satellite observation based fire inventory developed by NCAR (Wiedinmyer et al., 2011). The dust emission module was updated to be compatible with the 20-category MODIS land use data (Hu et al., 2015a) for in−line dust emission processing and sea salt emissions were also generated during CMAQ simulations.

The meteorological inputs were generated using WRF v3.6.1 (Skamarock et al., 2008). The initial and boundary conditions to WRF were downloaded from the NCEP FNL Operational Model Global Tropospheric Analyses dataset. WRF configurations details can be found in Zhang et al. (2012). WRF performance has been evaluated by comparing predicted 2m above surface temperature and relative humidity, and 10m wind speed and wind direction with all available observational data at ~1200 stations from the National Climate Data Center (NCDC). The model performance is generally acceptable and detailed evaluation results can be found in a previous study (Hu et al., 2016a).

The initial and boundary conditions representing relatively clean tropospheric concentrations were generated using CMAQ default profiles.

2.4 Model evaluation

Model predictions with the four emission inventories were evaluated against available observation data in China. Hourly observations of $PM_{2.5}$, PM_{10}, O_3, CO, SO_2, and NO_2 from March to December 2013 at 422 stations in 60 cities were obtained from CNEMC (http://113.108.142.147:20035/emcpublish/) but no observations were available for January and February. Observations at multiple sites in one city were averaged to calculate the average concentrations of the city. Detailed quality control of the data can be found in previous studies (Hu et al., 2016a;Hu et al., 2014a;Wang et al., 2014b). Statistical matrix of mean normalized bias (MNB), mean normalized error (MNE), mean fractional bias (MFB) and mean fractional error (MFE) were calculated using the Equations (E1)-(E4):

$$MNB = \frac{1}{N}\sum_{i=1}^{N}\left(\frac{C_m - C_o}{C_o}\right) \quad \text{(E1)}$$

$$MNE = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{C_m - C_o}{C_o}\right| \quad \text{(E2)}$$

$$MFB = \frac{1}{N}\sum_{i=1}^{N}\frac{\left(C_m - C_o\right)}{\left(\frac{C_o + C_m}{2}\right)} \quad \text{(E3)}$$

$$MFE = \frac{1}{N}\sum_{i=1}^{N}\frac{\left|C_m - C_o\right|}{\left(\frac{C_o + C_m}{2}\right)} \quad \text{(E4)}$$

where C_m and C_o are the predicted and observed city average concentrations, respectively, and N is the total number of observation data. MNB and MNE are commonly used in evaluation of model performance of O_3, and MFB and MFE are commonly used in evaluation of model performance of $PM_{2.5}$ (Tao et al., 2014). The U.S. EPA previously recommended O_3 model performance criteria of within ± 0.15 for MNB and less than 0.30 for MNE (as shown in Figure 1) and PM model performance criteria of within ± 0.60 for MFB and less than 0.75 for MFE (EPA, 2001b). Figure 2 includes the criteria and goals for PM as a function of PM concentration, as suggested by Boylan and Russell (2006), which have been widely used in model evaluation.

2.5 Ensemble predictions

The four sets of predictions with different inventories were combined linearly to calculate the ensemble predictions, as shown in Equation (E5):

$$C^{pred,ens} = \sum_{m=1}^{N_m} w_m C^{pred,m} \quad \text{(E5)}$$

where $C^{pred,ens}$ is the ensemble prediction, $C^{pred,m}$ is the predicted concentration from the m^{th} simulation, N_m is the number of simulations in the ensemble (N_m=4), and w_m is the weighting factor of the m^{th} simulation. The weighting factor for each set of predictions was determined by minimizing the objective function Q in Equation (6):

$$Q = \sum_{i}^{N_{city}} \left[C_i^{obs} - \sum_{m=1}^{N_m} w_m C_i^{pred,m} \right]^2 \quad \text{(E6)}$$

where C_i^{obs} is the observed $PM_{2.5}$ or O_3 concentration at the i^{th} city, N_{city} is the total number of cities with observation (N=60), $C_i^{pred,m}$ is the predicted concentration at the i^{th} city from the m^{th} simulation, and N_m is the number of simulations in the ensemble (N_m=4). The weight factor w_m of the m_{th} simulation to be determined is within the range of [0, 1], with w=0 represents no influence of the individual simulation on the ensemble prediction, and w=1 indicates that concentrations of the individual simulation are fully accounted in the ensemble prediction. The observations data were the same as used in the model evaluation. Ensemble predictions were performed for $PM_{2.5}$ and O_3 in this study. A MATLAB program was developed to solve above equation and determine the weighting factors using the linear least square solver "lsqlin".

3. Results

3.1 Model performance on gaseous and particulate pollutants

Table 1 summarizes the overall model performance on O_3, CO, NO_2, SO_2, $PM_{2.5}$, and PM_{10} with different inventories using the averaged observations in 60 cities in 2013. Model performance meets the O_3 criteria for all inventories. O_3 from SOE are 7.2 parts per billion (ppb) lower than the mean observed concentration while the under-predictions of the other three inventories are less than 2 ppb. CO, NO_2, and SO_2 are under-predicted by all inventories, indicating potential uncertainties in the inventories. CO predictions from three inventories (SOE inventory does not include CO) are substantially lower than observations, with the best performance (lowest MNB and MNE) from REAS2. NO_2 overall performance is similar to CO. However, MEIC and SOE yield the lowest MNB, and EDGAR yields the highest. SO_2 performance is better than CO and NO_2, and MEIC and SOE yield the lowest MNB, while MNE values of the four inventories are very similar. $PM_{2.5}$ and PM_{10} predictions using all inventories meet the performance criteria with similar MFB and MFE values. REAS2 generally yields slightly better $PM_{2.5}$ and PM_{10} performance, but all inventories under-predict the concentrations generally.

The difference in model performance with the four inventories also varies seasonally and spatially. Figure 2 shows the comparison of model performance for hourly gaseous species (O_3, CO, NO_2, and SO_2) in each month from March to December 2013. The MNB values of O_3 in most months are within the criteria for all inventories except for SOE, which under-predicts O_3 concentrations. March has the worst performance for all inventories with MNE values larger than 0.4 for MEIC, SOE, and EDGAR. No significant performance difference among different inventories in different months is found, but large difference exists in various regions of China (see the definition of regions of China in Figure 1). O_3 predicted using MEIC, EDGAR, and REAS2 meets the performance criteria in most regions except for YRD by MEIC and PRD by EDGAR. O_3 predicted using SOE only meets the criteria in Northwest (NW) and other region (Other) of China. CO and NO_2 are under-predicted in all regions, with the largest under-predictions in NW and Other. This pattern is similar among the results with all inventories. SO_2 is generally under-predicted in all regions except, but over-predicted in the Sichuan Basin (SCB) by all inventories. SO_2 is also over-predicted by EDGAR in the PRD region. SO_2 in Northeast (NE) is substantially under-predicted by MEIC and REAS2. In general, model performance in

the more developed regions such as YRD, NCP, and PRD are relatively better, compared to NW and Other regions.

Figure 3 illustrates the $PM_{2.5}$ and PM_{10} performance statistics of MFB and MFE as a function of absolute concentrations in different months of 2013 and in different regions. $PM_{2.5}$ predictions based on each inventory are within the performance goal of MFB and between the goal and criteria of MFE in all months. There is no significant difference among inventories. Half of monthly averaged PM_{10} MFB values fall within the goal while the rest are between the goal and criteria. MFE values of PM_{10} are all between the goal and criteria. From the regional perspective, $PM_{2.5}$ performance in NE by SOE is out of the MFB criteria, while that in SCB by MEIC, SOE, and REAS2 are out of the MFE criteria. MFB values of PM_{10} at all regions meet the criteria except NW, due to under-estimation of windblown dust emissions in NW.

3.2 Spatial variations in predicted gaseous and particulate pollutants

Figure 4 shows the spatial distribution of annual averaged gas species, daily maximum 1-hour O_3 (O_3-1h), and 8-hour mean O_3 (O_3-8h), NO_2, and SO_2 predicted by MEIC and differences between SOE, EDGAR, and REAS2 to MEIC. MEIC predicts annual O_3-1h concentrations are ~60ppb in most parts of China with the highest values of ~70ppb in SCB. SOE predicts lower O_3-1h values than MEIC, with about 5 ppb differences in the SCB, central China (CNT), and North China Plain (NCP) regions and 2-3 ppb differences in other regions. EDGAR also predicts 2-3 ppb lower O_3-1h in most regions than MEIC but its O_3-1h predictions in the Tibet Plateau, NCP and ocean regions are 2-3 ppb higher than MEIC predictions. REAS2 predicted O_3-1h values are lower than MEIC for scattered areas in the NE, NW, and CNT regions and other regions experience slightly higher O_3-1h. MEIC, SOE, and REAS2 have similar results out of China (the difference is generally less than 1 ppb) since the simulations used same emissions for those regions. O_3-8h shows similar spatial distributions as O_3-1h among inventories with slightly less differences. NO_2 concentrations are 10-15ppb in developed areas of the NCP and YRD regions, and greater than 5 ppb at other urban areas as predicted by MEIC. SOE predicts 2-3 ppb lower NO_2 concentrations in most areas except the vast NW region. EDGAR predicts lower NO_2 (more than 5 ppb difference) in urban areas of the NCP and YRD areas but higher concentrations in the entire west part of China by approximately 1-2 ppb. REAS2 has the closest NO_2 with MEIC as the 1-2 ppb underestimation and overestimation are almost evenly distributed in the whole country. SO_2 concentrations are up to 20ppb in the NCP, CNT, and SCB regions while are less than 5 ppb in other regions. SOE mostly predicts 2-3 ppb lower SO_2 in the east half of China with the largest difference of -10 ppb in the CNT region. EDGAR and REAS2 had very similar difference with MEIC, i.e., more than 5 ppb higher concentrations in the NCP and YRD, ~2 ppb higher concentrations in the PRD, 2-3 ppb lower concentrations in the NE and up to 5 ppb lower concentrations in the CEN and SCB.

Figure 5 shows the seasonal distribution of $PM_{2.5}$ total mass predicted by MEIC and differences between SOE, EDGAR, and REAS2 to MEIC. In spring, MEIC predicted $PM_{2.5}$ concentrations are ~50 $\mu g\ m^{-3}$ in east and south parts of China, and Southeast Asia has the highest value of ~100 $\mu g\ m^{-3}$. SOE predicts 5-10 $\mu g\ m^{-3}$ lower $PM_{2.5}$ in north China and < 5 $\mu g\ m^{-3}$ higher values in south China and along the coastline. EDGAR predicts >20 $\mu g\ m^{-3}$ lower values in NCP and ~10 $\mu g\ m^{-3}$ lower values in NE, CNT, and SCB, but up to 20 $\mu g\ m^{-3}$ higher values in PRD. REAS2

predicts higher $PM_{2.5}$ values in most parts of China except under-predictions in NE and SCB. The over-predictions in YRD and NCP are up to 20-30 μg m^{-3}. In summer, the high $PM_{2.5}$ regions are much smaller compared to spring with ~50 μg m^{-3} ppb concentrations in NCP, north part of YRD and SCB and 20-30 μg m^{-3} in other parts. Generally, SOE predicts <10 μg m^{-3} lower values in most regions. EDGAR predicts lower values in NCP and SCB and 5-10 μg m^{-3} higher values in south part. REAS2 almost predicts higher values in all the regions except some scattered areas in NCP, YRD, and SCB.

In fall, $PM_{2.5}$ concentrations are larger than 50 μg m^{-3} in most regions except NW and are ~100 μg m^{-3} in part of NCP, CNT, and SCB. SOE predicted values are lower in north part and higher in south part. EDGAR predicts up to 30 μg m^{-3} lower values in NCP and SCB while up to 20 μg m^{-3} higher values in YRD. REAS2 again estimates close values to MEIC with less than 5 μg m^{-3} differences in most regions and up to 20 μg m^{-3} higher values in scattered areas in YRD and SCB. In winter, MEIC predicted $PM_{2.5}$ concentrations are up to 200 μg m^{-3} in NCP, CNT, YRD, and SCB, while PRD has concentrations of ~50 μg m^{-3}. SOE severely underestimates by 30 μg m^{-3} in all regions with high $PM_{2.5}$ concentrations and only coast areas experience <10 μg m^{-3} higher values. EDGAR also predicts 30 μg m^{-3} lower $PM_{2.5}$ concentrations in NE, NCP, CNT, and SCB, but the YRD region has 20 μg m^{-3} higher values. The regions with lower values by REAS2 compared to MEIC are at the regions of NE, NCP, CNT and SCB, similar to EDGAR but with much smaller areas. SOE predicts higher $PM_{2.5}$ in the south parts of YRD and NCP than MEIC.

Figure 6 shows the annual averaged concentrations of $PM_{2.5}$ components predicted by MEIC and the differences between other inventories with MEIC. Annual averaged particulate sulfate (SO_4^{2-}) concentrations are 20-25 μg m^{-3} in NCP, CNT, and SCB, and about 10 μg m^{-3} in other regions in the southeast China. SOE predicts ~10 μg m^{-3} lower values in high concentration areas and 2-3 μg m^{-3} lower in other areas. EDGAR predicts ~5 μg m^{-3} higher SO_4^{2-} in southeast China and 2-3 μg m^{-3} lower values in SCB. REAS2 predicted SO_4^{2-} are generally 2-3 μg m^{-3} lower than that of MEIC in areas except the coastal areas. MEIC predicts the highest particulate nitrate (NO_3^-) concentrations of up to 30 μg m^{-3} in NCP and YRD and values in other regions are 5-10 μg m^{-3} except the northwest China. SOE predicts <5 μg m^{-3} lower values in the high concentrations areas and ~2 μg m^{-3} higher values in coastal areas. EDGAR uniformly predicts lower NO_3^- values than MEIC with the largest different of 10 μg m^{-3}. REAS2 has similar results to SOE. Particulate ammonium (NH_4^+) concentrations predicted by MEIC have a peak of 15 μg m^{-3} and are mostly less than 10 μg m^{-3} in the east and south parts of China. SOE predicts slightly lower values except for coastal areas in PRD, where 1-2 μg m^{-3} higher values are observed.

EC concentrations are generally low compared to other components as predicted by MEIC. The highest values are less than 10 μg m^{-3} in NCP, CNT and SCB. All other three inventories predict 1-2 μg m^{-3} lower EC values throughout the country. Primary organic aerosol (POA) predicted by MEIC are 20-30 μg m^{-3} in NCP, CNT and SCB, and are ~10 μg m^{-3} in other areas in east and south parts of China. SOE predicts up to 5 μg m^{-3} higher values in most areas with scattered places with ~2 μg m^{-3} lower values compared to MEIC. EDGAR and REAS2 predict up to ~10 μg m^{-3} lower values except for coastal areas. SOA concentrations are low in north part of China and up to 10 μg m^{-3} in the whole east and south parts. All three other inventories predict ~2 μg m^{-3} lower SOA values compared to MEIC. For other implicit components (OTHER), the highest concentrations are ~15 μg m^{-3} in NW and NCP, while other regions have lower than 5 μg m^{-3}

concentrations. In NW, the major sources of OTHER are windblown dust online generated by CMAQ simulations, thus almost no differences are observed among inventories. SOE and EDGAR predict lower OTHER vales in north part (~2 μg m^{-3}) and slightly higher values in south and east parts (~5 μg m^{-3}). REAS2 predicts higher OTHER values in the whole east part uniformly with up to 10 μg m^{-3} differences in NCP, YRD, and SCB regions.

Additional comparisons of model predictions in different regions and some major cities in China are shown in Figures S1-S4 in the Supplemental Material.

3.3 Ensemble predictions

Above analyses indicate that model performance with different inventories varies on different pollutants and in different regions. Table S2 shows the observed annual average concentrations of $PM_{2.5}$ in the 60 cities and the predictions from the four inventories as well as the weighted ensemble predictions. The weighting factors for predictions using MEIC, REAS2, SOE and EDGAR are 0.31, 0.36, 0.24 and 0.20, respectively (Table 2). The ensemble predictions greatly reduce MFB with a value of -0.11, compared to the MFB values of -0.25 – -0.16 using the annual average concentrations in the individual simulations. Also, the ensemble predictions have an MFE value of 0.24, lower than any MFE values of 0.26 – 0.31 in individual simulations (Figure 7). The ensemble predictions of annual O_3-1h have the MNB and MNE of 0.03 and 0.14, improved from MNB of 0.06 – 0.19 and MNE of 0.16 – 0.22 in the individual predictions, respectively.

To further evaluate the ability of the ensemble method in improving predictions at locations where observational data are not available, ensemble predictions were made using a data withholding method. For each city, the observations at the other 59 cities were used to determine the weighting factors in E6 and the ensemble prediction at the city was calculated. Performance of the ensemble predictions at the city was calculated using the withheld observations to evaluate the performance. The evaluation process was repeated for each of the 60 cities and the performance was compared to that with individual inventories (shown in Table S3). The results show that the ensemble predictions are better than those with EDGAR, MEIC, REAS2 and SOE at 36, 37, 32 and 40 cities for $PM_{2.5}$, and 39, 39, 43, and 38 cities for O_3-1h, respectively. The ensemble predictions are better than ≥ 2 of the individual predictions at 45 and 41 cities for $PM_{2.5}$ and O_3-1h, respectively. Out of the 15 cities that the ensemble $PM_{2.5}$ is only better than one or none of the individual predictions, 10 cities have MFB within ±0.25 and MFE less than 0.25. Out of the 19 cities that the ensemble O_3-1h is only better than one or none of the individual predictions, 14 cities still have MNB within ±0.2 and MNE less than 0.2. The results demonstrate that the ensemble can improve the predictions even at locations with no observational data available.

Previous studies have revealed that CTMs predictions agree more when averaging over longer times (i.e., annual vs. monthly vs. daily averages) (Hu et al., 2014b;Hu et al., 2015b). Ensemble predictions were also calculated with daily and monthly averages for $PM_{2.5}$, in addition to the calculation with annual averages discussed above. The weighting factors and the performance of ensemble predictions are shown in Table 2 and Figure 7, respectively. The weighting factors vary largely with the averaging times, suggesting that the prediction optimization need to be

conducted separately when using different time averages. The ensemble predictions improve the agreement with observations in all averaging time cases, with lower MNB and MNE than any of the individual predictions. In general, EDGAR and REAS have large weights for daily and monthly ensemble calculations, and MEIC and SOE have large weights for annual ensemble calculations. This result indicates that the annual total emission rates of MEIC and SOE are likely accurate but the temporal profiles to allocate the annual total emissions rates to specific day/hours need to be improved.

Table 3 shows the ensemble prediction performance on $PM_{2.5}$ and O_3-1h in different regions of China using the daily average observations and daily average predictions with individual inventories. The weighting factors vary greatly among regions, reflecting that substantial difference in the spatial distributions of $PM_{2.5}$ and O_3 when using different inventories. The MNB and MNE values of ensemble predictions are reduced in all regions for both pollutants, suggesting the ensemble predictions improve the accuracy and can be better used in further health effects studies. The similar findings are also found with the monthly average observations and predictions (shown in Table S4).

Figure 8 shows spatial distributions of $PM_{2.5}$ and its components from the ensemble predictions using the weighting factors of annual averages. The ensemble of $PM_{2.5}$ components were calculated using the same weighting factors for $PM_{2.5}$. Over 80 μg m^{-3} annual average $PM_{2.5}$ concentrations are estimated in NCP, CNT, YRD and SCB regions in 2013. Secondary inorganic aerosols (SO_4^{2-}, NO_3^-, and NH_4^+) account for approximately half of $PM_{2.5}$, and exhibit similar spatial patterns. Carbonaceous aerosols (EC, POA, and SOA) account for about 30%, but POA and SOA have quite different spatial distributions. High POA concentrations are mainly distributed in NCP, CNT and SCB, while high SOA concentrations are found in the south part of China. By considering the spatial distributions of population and ensemble $PM_{2.5}$, the population-weighted annual averaged $PM_{2.5}$ concentration in China in 2013 is 59.5 μg m^{-3}, which is higher than the estimated value of 54.8 μg m^{-3} by Brauer et al. (2016).

The products of the current study can be further applied in health effects studies. The first such analysis used the annual $PM_{2.5}$ ensemble predictions to assess the spatial distribution of excess mortality due to adult (> 30 years old) ischemic heart disease (IHD), cerebrovascular disease (CEV), chronic obstructive pulmonary disease (COPD) and lung cancer (LC) in China caused by $PM_{2.5}$ exposure (Hu et al., 2017a). Any health studies requiring human exposure information to different pollutants would benefit from this study. Even though the weighted factors vary depending on the regions, averaging times and different study years, the ensemble method proposed in this study is to minimize the difference between predictions and observations and can be applied in different studies. The way to calculate the weighting factors depends on the objectives of specific studies. But in general, more observation data used in the calculation, more accurate the ensemble prediction would be.

4. Conclusion

In this study, air quality predictions in China in 2013 were conducted using the WRF/CMAQ modeling system with anthropogenic emissions from four inventories including MEIC, SOE, EDGAR, and REAS2. Model performance with the four inventories was evaluated by comparing

with available observation data from 422 sites in 60 cities in China. Model predictions of hourly O_3 and $PM_{2.5}$ with the four inventories generally meet the model performance criteria, but that model performance with different inventories varies on different pollutants and in different regions. To improve the overall agreement of the predicted concentrations with observations, ensemble predictions were calculated by linearly combining the predictions from different inventories. The ensemble annual concentrations show improved agreement with observations for both $PM_{2.5}$ and O_3-1h. The MFB and MFE of the ensemble predictions of $PM_{2.5}$ at the 60 cities are -0.11 and 0.24, respectively, which are better than the MFB (-0.25 – -0.16) and MFE (0.26 – 0.31) of any individual simulations. The ensemble predictions of annual O_3-1h have the MNB and MNE of 0.03 and 0.14, improved from MNB (0.06 – 0.19) and MNE (0.16 – 0.22) in individual predictions. The ensemble predictions with data withholding method at each city show better performance than the predictions with individual inventories at most cities, demonstrating the ability of the ensemble in improving the predictions at locations where observational data are not available. The ensemble predictions agree better with observations with daily, monthly, and annual averaging times in all regions of China. The study demonstrates that ensemble predictions by combining predictions from individual emission inventories can improve the accuracy in the concentration estimation and the spatial distributions of air pollutants. The data presented in the paper is available for downloading via requests.

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Table 1. Overall model performance of gas and PM species in 2013 using different inventories. Obs is observation, MFB is mean fractional bias, MFE is mean fractional error, MNB is mean normalized bias, and MNE is mean normalized error. The indices were calculated with hourly observations and predictions.

		Prediction	MFB	MFE	MNB	MNE
	Mean Obs: 51.70 ppb					
	MEIC	49.83	-0.08	0.35	0.02	0.33
O_3	SOE	44.51	-0.2	0.38	-0.09	0.32
	EDGAR	49.82	-0.04	0.28	0.03	0.28
	REAS2	51.17	-0.04	0.33	0.05	0.33
	Mean Obs: 0.96 ppm					
	MEIC	0.31	-0.92	0.96	-0.57	0.63
CO	SOE	/	/	/	/	/
	EDGAR	0.23	-1.12	1.16	-0.66	0.73
	REAS2	0.42	-0.72	0.82	-0.41	0.59
	Mean Obs: 21.45 ppb					
	MEIC	10.12	-0.79	0.93	-0.41	0.66
NO_2	SOE	11.59	-0.65	0.81	-0.33	0.61
	EDGAR	6.82	-1.02	1.07	-0.6	0.67
	REAS2	9.3	-0.81	0.92	-0.46	0.63
	Mean Obs: 17.21 ppb					
	MEIC	12.5	-0.51	0.87	0.01	0.87
SO_2	SOE	12.76	-0.44	0.83	0.06	0.86
	EDGAR	15.86	-0.16	0.73	0.31	0.88
	REAS2	15.15	-0.23	0.74	0.23	0.86
	Mean Obs: 70.01 μg m^{-3}					
	MEIC	56.39	-0.32	0.64	-0.02	0.63
$PM_{2.5}$	SOE	59.77	-0.24	0.61	0.09	0.67
	EDGAR	52.59	-0.3	0.59	-0.05	0.56
	REAS2	60.35	-0.21	0.59	0.08	0.63
	Mean Obs: 118.61 μg m^{-3}					
	MEIC	62.7	-0.63	0.79	-0.32	0.61
PM_{10}	SOE	63.32	-0.6	0.76	-0.3	0.6
	EDGAR	55.76	-0.67	0.78	-0.38	0.58
	REAS2	71.41	-0.49	0.7	-0.21	0.59

Table 2. The weighting factors (w) of each inventory in the ensemble predictions of $PM_{2.5}$ when using daily, monthly, or annual averages in the objective function (E5).

	Daily	Monthly	Annual
MEIC	0.07	0.13	0.31
SOE	0.14	0.16	0.24
EDGAR	0.38	0.23	0.20
REAS2	0.49	0.63	0.36

Table 3. Performance of daily $PM_{2.5}$ (MFB and MFE) and O_3-1h (MNB and MNE) in different regions of China based on individual inventories and the ensemble. The weighting factors (w) used to calculate the ensemble of each region are also included.

	Region	MEIC			SOE			EDGAR			REAS2			ENSEMBLE	
	(# of Cities)	w	MFB	MFE	w	MFB	MFE	w	MFB	MFE	w	MFB	MFE	MFB	MFE
$PM_{2.5}$	NE (4)	0.16	-0.23	0.44	0.21	0.38	0.68	0.20	-0.30	0.43	0.43	-0.12	0.43	-0.08	0.42
	NCP (14)	0.00	-0.30	0.47	0.52	-0.34	0.46	0.14	-0.40	0.51	0.56	-0.20	0.41	-0.12	0.40
	NW (6)	0.00	-0.87	0.90	0.20	-0.80	0.84	0.59	-0.85	0.87	1.00	-0.81	0.83	-0.49	0.66
	YRD (20)	0.05	-0.29	0.45	0.00	-0.27	0.43	0.61	-0.23	0.40	0.35	-0.13	0.40	-0.18	0.38
	CNT (5)	0.09	-0.10	0.46	0.18	-0.05	0.41	0.50	-0.27	0.40	0.22	0.09	0.44	-0.14	0.37
	SCB (2)	0.00	0.10	0.48	0.64	0.23	0.48	0.00	-0.10	0.39	0.08	0.07	0.43	-0.15	0.40
	SOUTH (9)	0.10	-0.35	0.51	0.00	-0.18	0.41	0.59	-0.07	0.45	0.30	-0.25	0.44	-0.16	0.41
	CHINA (60)	0.07	-0.34	0.52	0.14	-0.26	0.50	0.38	-0.33	0.49	0.49	-0.22	0.46	-0.20	0.45
		w	MNB	MNE	w	MNB	MNE	w	MNB	MNE	w	MNB	MNE	MNB	MNE
O_3-1h	NE	0.09	0.44	0.50	0.00	0.16	0.34	0.45	0.41	0.47	0.27	0.42	0.48	0.14	0.31
	NCP	0.29	0.33	0.47	0.12	0.23	0.44	0.06	0.46	0.59	0.42	0.47	0.56	0.25	0.43
	NW	0.00	0.65	0.72	0.82	0.54	0.62	0.00	0.70	0.77	0.00	0.68	0.74	0.25	0.46
	YRD	0.00	0.20	0.41	0.53	0.14	0.38	0.00	0.25	0.45	0.45	0.27	0.44	0.17	0.39
	CNT	0.27	0.27	0.47	0.18	0.16	0.43	0.10	0.35	0.53	0.36	0.35	0.52	0.18	0.42
	SCB	0.44	0.59	0.68	0.14	0.42	0.58	0.28	0.59	0.70	0.00	0.60	0.72	0.33	0.53
	SOUTH	0.84	0.39	0.50	0.00	0.29	0.46	0.00	0.38	0.51	0.00	0.42	0.53	0.16	0.37
	CHINA	0.19	0.34	0.49	0.20	0.23	0.44	0.00	0.39	0.54	0.51	0.41	0.53	0.21	0.42

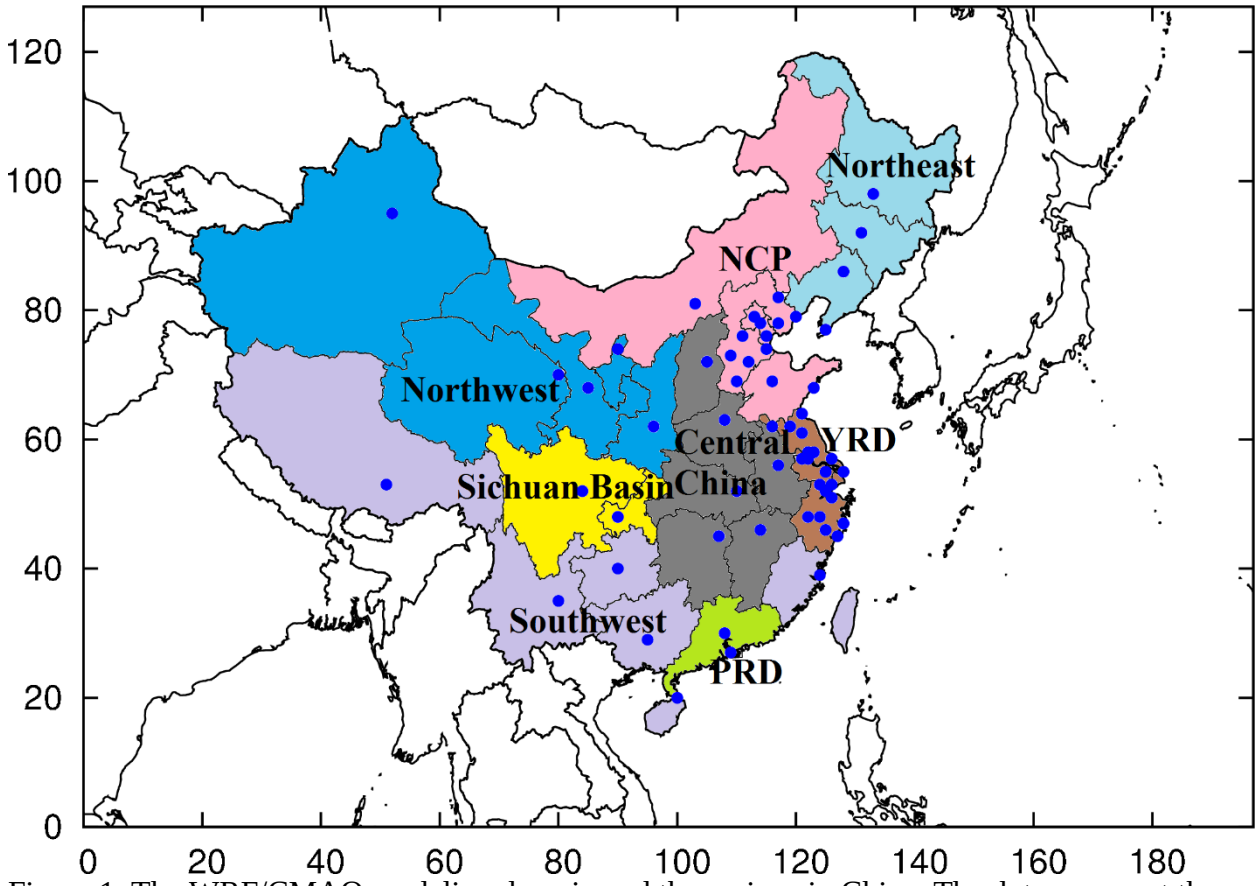

Figure 1. The WRF/CMAQ modeling domain and the regions in China. The dots represent the 60 cities where observational data are available for ensemble analysis.

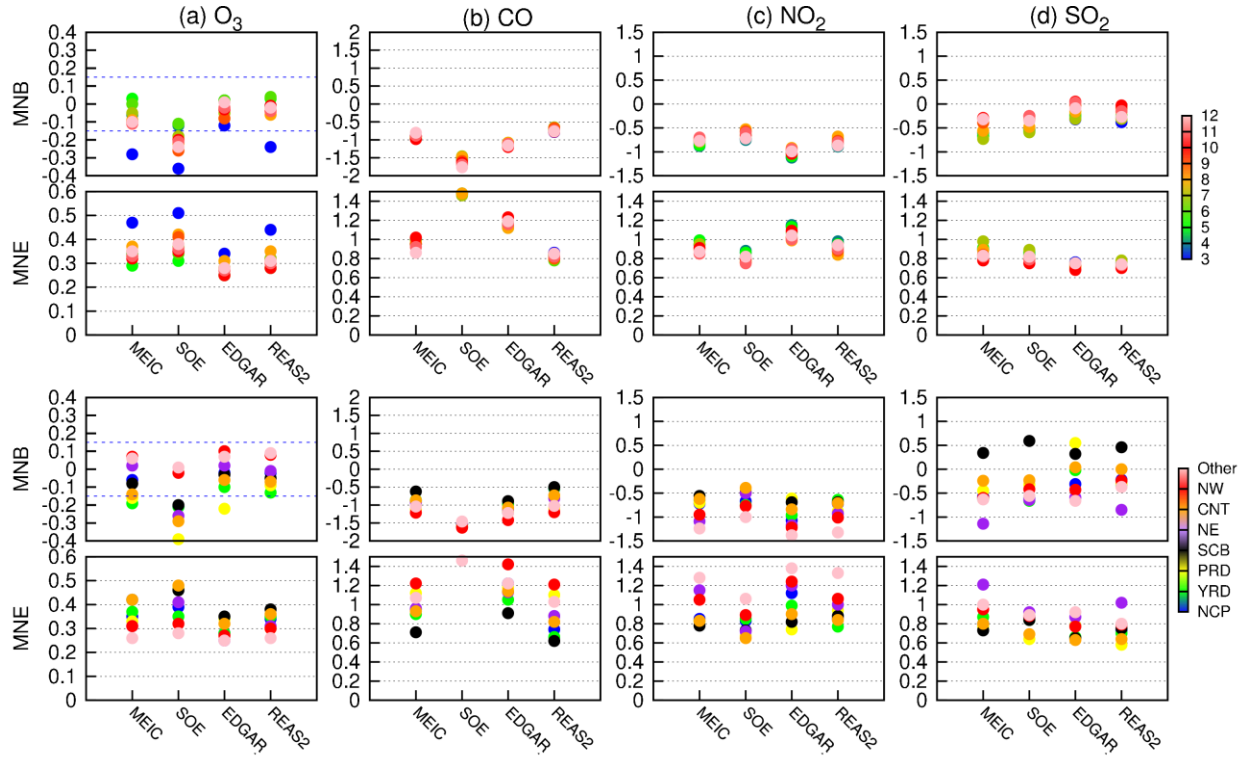

Figure 2. Performance of predicted O_3, CO, NO_2, and SO_2 for different months (top two rows) and regions based on simulations with individual inventories. The blue dashed lines on the O_3 plots are +/-0.15 for MNB and 0.3 for MNE as suggested by U. S. EPA (2001a). Changes of colors show the months from March to December in top two rows, while show regions from NCP to Other in the bottom two rows. The same for Figure 2.

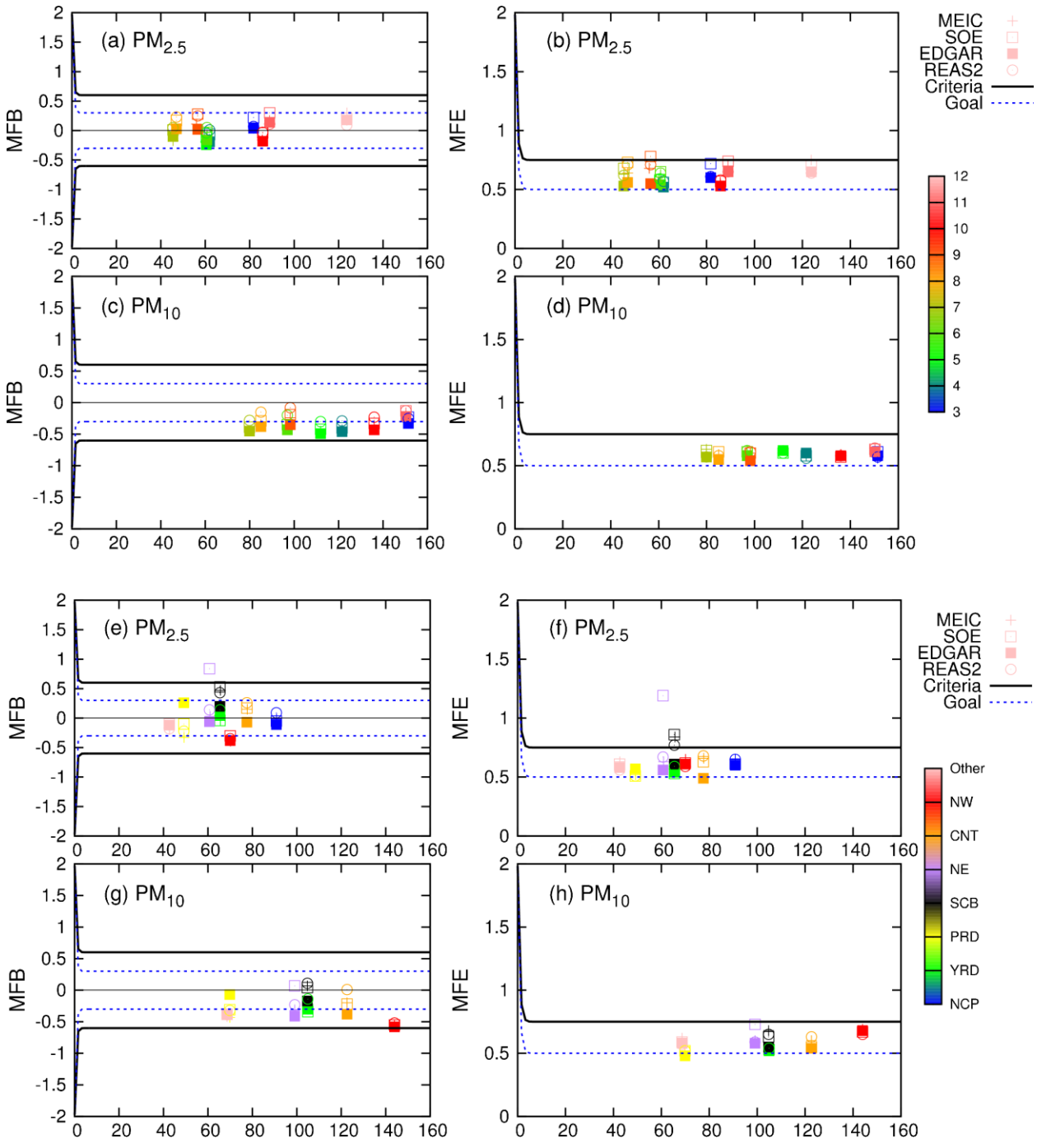

Figure 3. Performance of predicted $PM_{2.5}$ and PM_{10} for different months (a-d) and regions (e-h) based on simulations with individual inventories. The x-axis is the observed concentrations. The model performance criteria (solid black lines) and goals (dash blue lines) are suggested by Byun and Russell (2006). The model performance goals represent the level of accuracy that is considered to be close to the best a model can be expected to achieve, and the model performance criteria represent the level of accuracy that is considered to be acceptable for modeling applications.

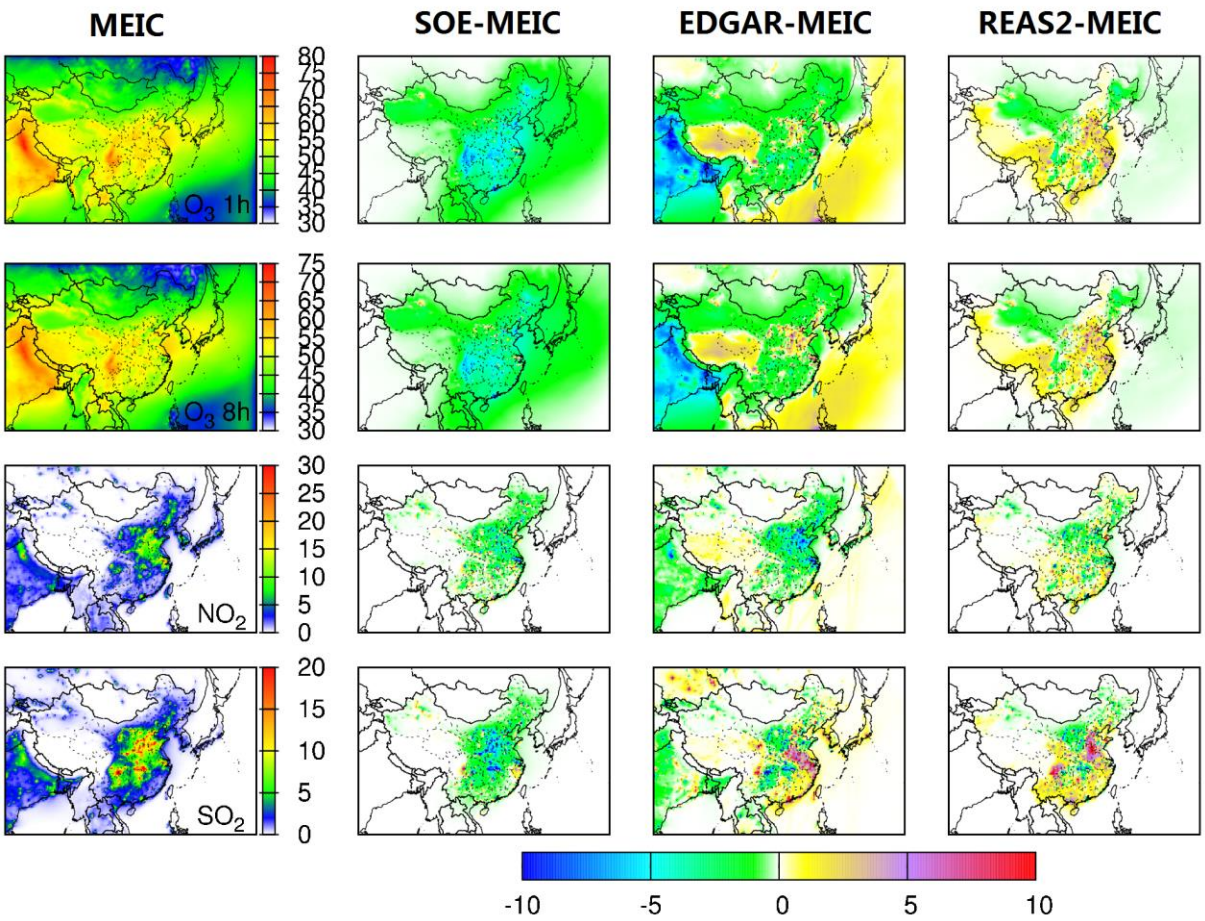

Figure 4. Spatial difference of model predicted annual averaged gas species concentrations (in the horizontal panels) with different inventories (in the vertical panels). Units are ppb. The color bars of the first column are different to better show the spatial distribution of different species. White indicates zero while blue, green, yellow and red means concentrations from low to high. The color bar for the other three columns are same, white indicates zero, blue and green mean values less than zero while yellow, purple and red mean values larger than zero. O_3-1h represents daily maximum 1h O_3 and O_3-8h represents daily maximum 8h mean O_3.

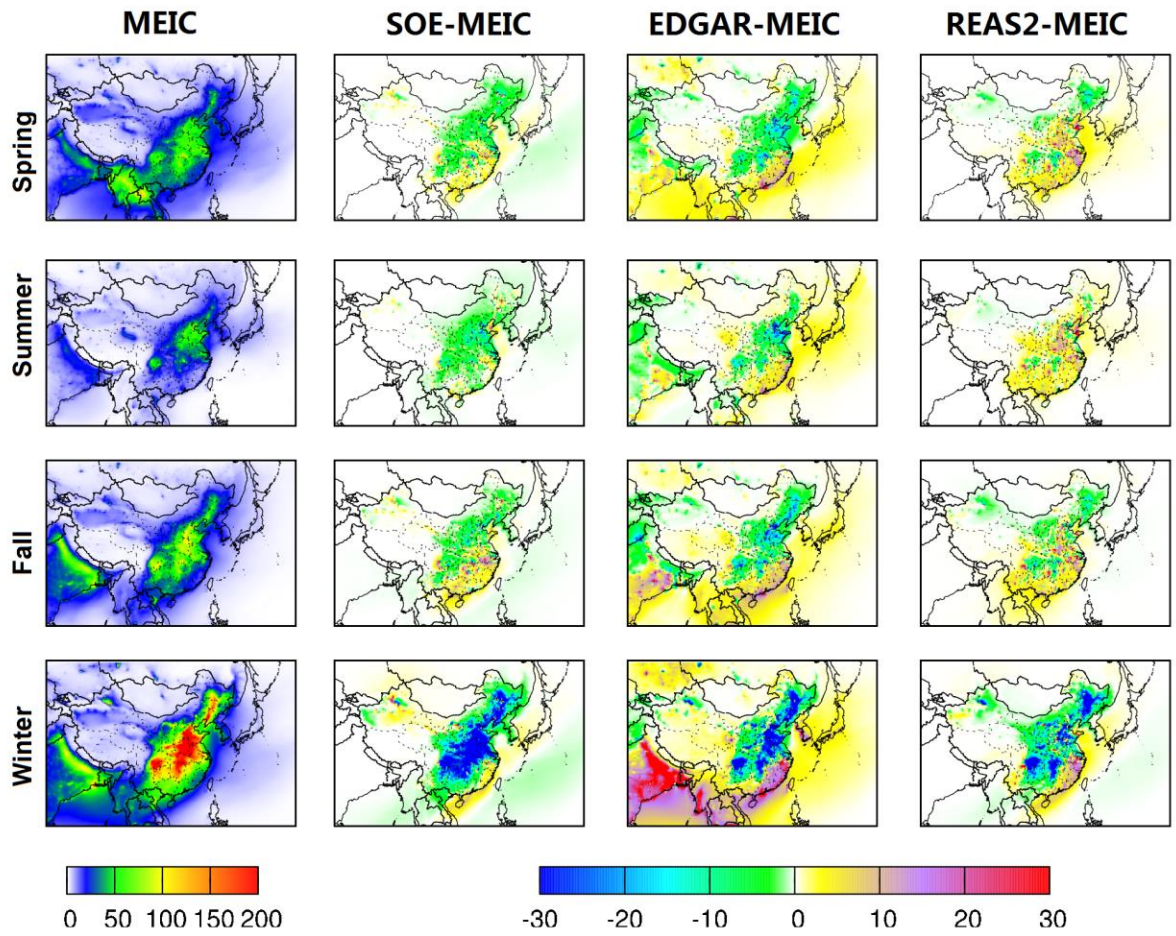

Figure 5. Spatial difference of model predicted seasonal averaged $PM_{2.5}$ concentrations (in the horizontal panels) with different inventories (in the vertical panels). Units are μg m^{-3}. In the first column, white indicates zero while blue, green, yellow and red means concentrations from low to high. The color bar for the other three columns are same, white indicates zero, blue and green mean values less than zero while yellow, purple and red mean values larger than zero. The same for Figure 6.

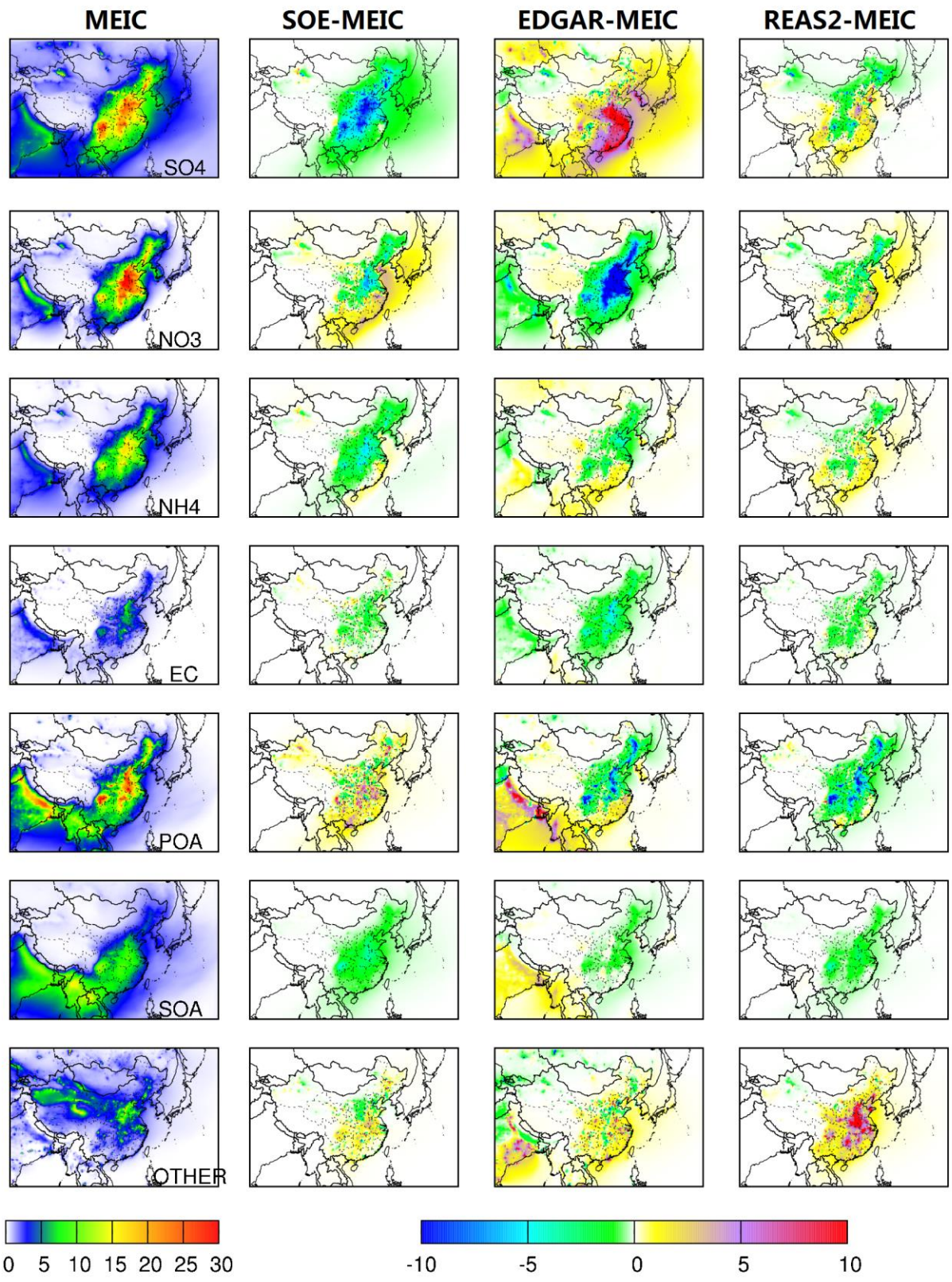

Figure 6. Spatial difference of model predicted annual $PM_{2.5}$ components (in the horizontal panels) with different inventories (in the vertical panels). Units are μg m^{-3}.

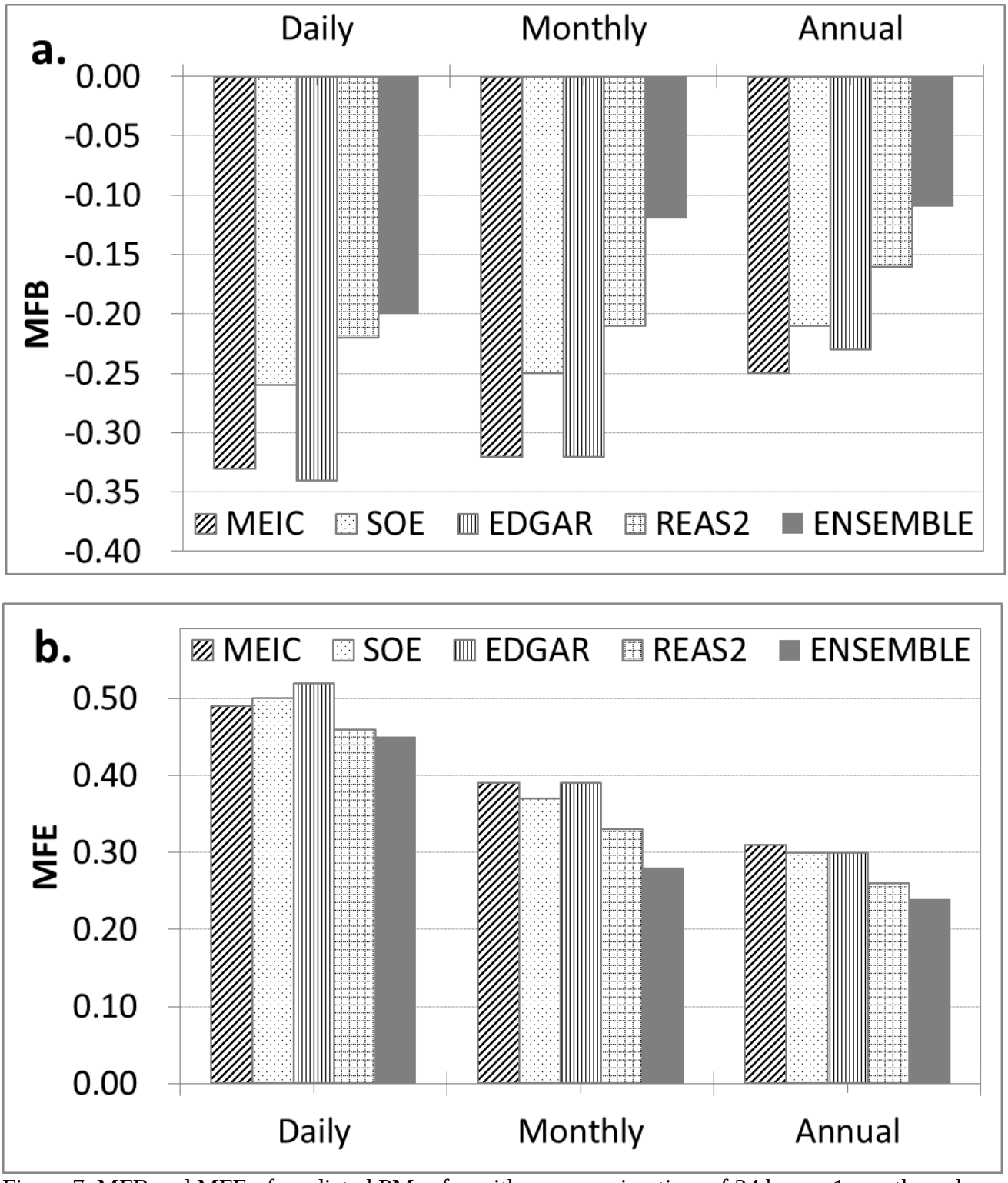

Figure 7. MFB and MFE of predicted $PM_{2.5}$ for with an averaging time of 24 hours, 1 month, and 1 year based on the individual inventories and the ensemble.

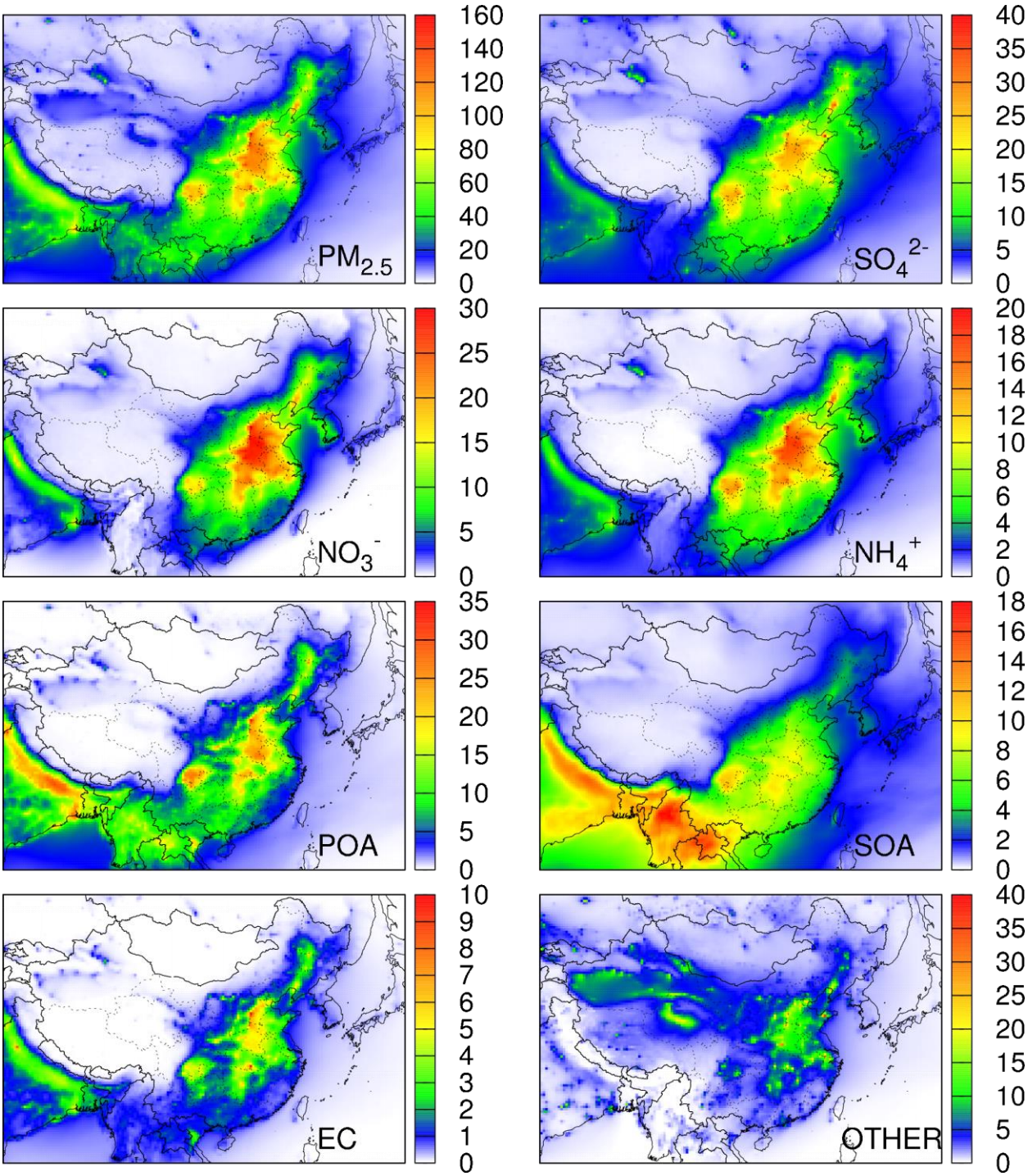

Figure 8. Spatial distributions of $PM_{2.5}$ and its components in the ensemble predictions. Units are μg m^{-3}. The scales of the panels are different. White indicates zero while blue, green, yellow and red means concentrations from low to high.