

1 **A six-year-long (2013–2018) high-resolution air quality reanalysis** 2 **dataset in China based on the assimilation of surface observations** 3 **from CNEMC**

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24 **Abstract**

25 A six-year-long high-resolution Chinese air quality reanalysis (CAQRA) dataset is presented in this study obtained from the
26 assimilation of surface observations from China National Environmental Monitoring Centre (CNEMC) using the ensemble
27 Kalman filter (EnKF) and Nested Air Quality Prediction Modeling System (NAQPMS). This dataset contains surface fields
28 of six conventional air pollutants in China (i.e., PM_{2.5}, PM₁₀, SO₂, NO₂, CO and O₃) for period 2013–2018 at high spatial (15
29 km×15 km) and temporal (1 hour) resolutions. This paper aims to document this dataset by providing detailed descriptions of
30 the assimilation system and the first validation results for the above reanalysis dataset. The fivefold cross-validation (CV)
31 method is adopted to demonstrate the quality of the reanalysis. The CV results show that the CAQRA yields an excellent
32 performance in reproducing the magnitude and variability of surface air pollutants in China from 2013 to 2018 (CV R² = 0.52–
33 0.81, CV root mean square error (RMSE) = 0.54 mg/m³ for CO and CV RMSE = 16.4–39.3 µg/m³ for the other pollutants
34 at the hourly scale). Through comparison to the Copernicus Atmosphere Monitoring Service reanalysis (CAMSR) dataset
35 produced by the European Centre for Medium-Range Weather Forecasts (ECWMF), we show that CAQRA attains a high

36 accuracy in representing surface gaseous air pollutants in China due to the assimilation of surface observations. The fine
37 horizontal resolution of CAQRA also makes it more suitable for air quality studies at the regional scale. The PM_{2.5} reanalysis
38 dataset is further validated against the independent datasets from the U.S. Department State Air Quality Monitoring Program
39 over China, which exhibits a good agreement with the independent observations ($R^2 = 0.74\text{--}0.86$ and RMSE =16.8–33.6
40 $\mu\text{g}/\text{m}^3$ in different cities). Besides, through the comparison to satellite-estimated PM_{2.5} concentrations, we show that the
41 accuracy of the PM_{2.5} reanalysis is higher than that of most satellite estimates. The CAQRA is the first high-resolution air
42 quality reanalysis dataset in China that simultaneously provides the surface concentrations of six conventional air pollutants,
43 which is of great value for many studies, such as health impact assessment of air pollution, investigation of air quality changes
44 in China, model evaluation and satellite calibration, optimization of monitoring sites and provision of training data for
45 statistical or artificial intelligence (AI)-based forecasting. All datasets are freely available at
46 <https://doi.org/10.11922/sciencedb.00053> (Tang et al., 2020a), and a prototype product containing the monthly and annual
47 means of the CAQRA dataset has also been released at <https://doi.org/10.11922/sciencedb.00092> (Tang et al., 2020b) to
48 facilitate the evaluation of the CAQRA dataset by potential users.

49 **1 Introduction**

50 Air pollution is a critical environmental issue that adversely affects human health and is closely connected to climate
51 change (von Schneidemesser et al., 2015). Exposure to ambient air pollution has been confirmed by many epidemiological
52 studies to be a leading contributor to the global disease burden, which increases both morbidity and mortality (Cohen et al.,
53 2017). China, as the largest developing country, has achieved great economic development since the 1980s. This large-scale
54 economic expansion, however, is accompanied by a dramatic increase in air pollutant emissions, leading to severe air pollution
55 in China (Kan et al., 2012). Since 2012, the Chinese government has established a nationwide ground-based air quality
56 monitoring network (Fig. 1) to monitor the surface concentrations of six conventional air pollutants in China, i.e., particles
57 with an aerodynamic diameter of 2.5 μm or smaller (PM_{2.5}), particles with an aerodynamic diameter of 10 μm or smaller (PM₁₀),
58 sulfur dioxide (SO₂), nitrogen dioxide (NO₂), carbon monoxide (CO) and Ozone (O₃), which plays an irreplaceable role in
59 understanding the air pollution in China. In addition, since the implementation of Action Plan for the Prevention and Control
60 of Air Pollution in 2013, a series of aggressive control measures has been applied in China to reduce the emissions of air
61 pollutants. According to the estimates of Zheng et al. (2018b), the Chinese anthropogenic emissions has decreased by 59% for
62 SO₂, 21% for NO_x, 23% for CO, 36% for PM₁₀ and 35% for PM_{2.5} from 2013 to 2017. Concurrently, the air quality in China
63 has changed dramatically over the past six years (Silver et al., 2018; Zheng et al., 2017). Such large changes in the Chinese air
64 quality and their effects on human health and the environment have become an increasingly hot topic in many scientific fields
65 (e.g., Xue et al., 2019; Zheng et al., 2017) which requires a long-term air quality dataset in China with high accuracy and
66 spatiotemporal resolutions.

67 Ground-based observations can provide accurate information on the spatial and temporal distributions of air pollutants in
68 China, but they are sparsely and unevenly distributed in space. Satellite observations exhibit the advantages of a high spatial
69 coverage and have widely been applied in air pollution monitoring over large domains. A series of satellite retrievals related
70 to air quality has been developed over the past two decades, such as the observations of NO₂, SO₂ and O₃ columns from the
71 Ozone Monitoring Instrument (OMI; Levelt et al., 2006), CO column observations from the Measurement of Pollution in the
72 Troposphere (MOPITT; Deeter et al., 2003) and aerosol optical depth (AOD) observations from the Moderate Resolution
73 Imaging Spectroradiometer (MODIS; Barnes et al., 1998). These satellite column measurements have also been used to
74 estimate surface concentrations based on different methods, such as chemical transport models (CTMs) (e.g., van Donkelaar
75 et al., 2016; van Donkelaar et al., 2010), advanced statistical methods (e.g., Ma et al., 2014; Ma et al., 2016; Xue et al., 2019;
76 Zou et al., 2017) and semi-empirical models (e.g., Lin et al., 2015; Lin et al., 2018), which have been proven to be an effective
77 way to acquire wide-coverage distributions of surface air pollutant with a good accuracy (Chu et al., 2016; Shin et al., 2019).
78 However, challenges remain in satellite-based estimates due to missing values related to cloud contamination, uncertainties in
79 satellite measurements, and difficulties in modelling the complex relationship between surface concentrations and column
80 measurements (Shin et al., 2019; van Donkelaar et al., 2016; Xue et al., 2019). In addition, most satellite-based estimates of
81 surface concentrations exhibit low temporal resolutions (daily or even longer), which limits their application in fine-scale
82 studies, such as the assessment of the acute health effects of the air quality. To our knowledge, a nationwide long-term estimate
83 of the surface concentrations of all conventional air pollutants in China at the hourly scale have not yet been reported in
84 previous satellite estimates.

85 A long-term air quality reanalysis dataset of critical air pollutants can provide constrained estimates of their concentrations
86 at all locations and times, which optimally combines the accuracy of observations and the physical information and spatial
87 continuity of CTMs through advanced data assimilation techniques. Reanalysis datasets are uniform, continuous and state-of-
88 science best-estimate data products that have been adopted by a vast number of research communities. For example, several
89 long-term meteorological reanalysis datasets have been developed by various weather centres in different regions/countries,
90 such as the ERA-Interim reanalysis developed by the European Centre for Medium-Range Weather Forecasts (ECMWF; Dee
91 et al., 2011), the National Center for Atmospheric Research (NCAR)/National Centers for Environmental Protection (NCEP)
92 reanalysis developed by the NCEP (Saha et al., 2010), the Modern-Era Retrospective Analysis for Research and Applications
93 (MERRA) developed by the NASA Global Modeling and Assimilation Office (NASA-GMAO; Rienecker et al., 2011), the
94 Japanese 55-year Reanalysis (JRA-55) developed by the Japan Meteorological Agency (Kobayashi et al., 2015) and the China
95 Meteorological Administration's global atmospheric Reanalysis (CRA-40) developed by the China Meteorological
96 Administration (CMA). The use of data assimilation in atmospheric chemistry reanalysis is more recent, and certain reanalysis
97 datasets for the atmospheric composition have been produced over the past decades, for example the Monitoring Atmospheric
98 Composition and Climate (MACC), Copernicus Atmosphere Monitoring Service (CAMS) interim reanalysis (CIRA), and
99 CAMS reanalysis (CAMSRA) produced by the ECWMF (Flemming et al., 2017; Inness et al., 2019; Inness et al., 2013), the
100 MERRA-2 aerosol reanalysis produced by the NASA-GMAO (Randles et al., 2017), the tropospheric chemistry reanalysis

101 (TCR) from 2005–2012 produced by Miyazaki et al. (2015) and its latest version TCR-2 (Miyazaki et al., 2020), the global
102 reanalysis of carbon monoxide produced by Gaubert et al. (2016), the multi-sensor total ozone reanalysis from 1970–2012
103 produced by van der A et al. (2015) and the Japanese Reanalysis for Aerosols (JRAero) from 2011–2015 produced by
104 Yumimoto et al. (2017). These reanalysis datasets promote our understanding of the atmospheric composition and also
105 facilitate the air quality research. However, these datasets are all global datasets with coarse horizontal resolutions (> 50 km),
106 which may be insufficient to capture the high spatial variability of air pollutants at the regional scale. In addition, some of
107 these reanalysis datasets only provide air quality data prior to year 2012 and only focus on specific species. There is still no
108 high-resolution air quality reanalysis dataset in China capturing its dramatic air quality change during recent years.

109 In view of these discrepancies, in this study, we develop a high-resolution regional air quality reanalysis dataset in China
110 from 2013 to 2018 (which will be extended in the future on a yearly basis) by assimilating surface observations from China
111 National Environmental Monitoring Centre (CNEMC). The developed reanalysis dataset may help mitigate the lack of high-
112 resolution air quality datasets in China by providing surface concentration fields of all six conventional air pollutants in China
113 at high spatial ($15\text{ km}\times 15\text{ km}$) and temporal (hourly) resolutions, which is of great value to (1) retrospective air quality analysis
114 in China, (2) health and environmental impact assessment of air pollution at fine scales, (3) model evaluation and satellite
115 calibration, (4) optimization of monitoring sites and (5) provision of basic training datasets for statistical or artificial
116 intelligence (AI)-based forecasting.

117 **2 Description of the chemical data assimilation system**

118 The Chinese air quality reanalysis (CAQRA) dataset was produced with the chemical data assimilation system
119 (ChemDAS) developed by the Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP, CAS) (Tang et al., 2011).
120 This system consists of (i) a three-dimensional CTM called the Nested Air Quality Prediction Modeling System (NAQPMS)
121 developed by Wang et al. (2000), (ii) an ensemble Kalman filter (EnKF) assimilation algorithm, and (iii) surface observations
122 from CNEMC with the automatic outlier detection method developed by Wu et al. (2018). We adopted an offline analysis
123 scheme in this study since there are no previous experiences with online chemical data assimilation at such a high horizontal
124 resolution. The lessons learned from this offline analysis application could also facilitate future implementation of online
125 analysis. In the offline analysis scheme, a free ensemble simulation is first conducted, and the observations are then assimilated
126 using the EnKF. A similar offline analysis scheme has also been applied in previous reanalysis studies, such as Candiani et al.
127 (2013) and Kumar et al. (2012). Detailed descriptions of the ensemble simulation, observations and data assimilation algorithm
128 used in this study are presented below.

129 **2.1 Air pollution prediction model**

130 The NAQPMS model was used as the forecast model to represent the atmospheric chemistry, which has been applied in
131 previous assimilation studies (Tang et al., 2011; Tang et al., 2013). The model is driven by the hourly meteorological fields

132 produced by the Weather Research and Forecasting (WRF) model (Skamarock, 2008). Gas phase chemistry is simulated with
133 the carbon bond mechanism Z (CBM-Z) developed by Zaveri and Peters (1999). Aqueous-phase chemistry and wet deposition
134 are simulated based on the Regional Acid Deposition Model (RADM) mechanism in the Community Multi-scale Air Quality
135 (CMAQ) model version 4.6. In regard to aerosol processes, the thermodynamic model ISORROPIA 1.7 (Nenes et al., 1998)
136 is applied for the simulations of inorganic atmospheric aerosols. Six secondary organic aerosols (SOAs) are explicitly treated
137 in the NAQPMS model based on Li et al. (2011). To simulate the interactions between particles and gases, 28 heterogeneous
138 reactions involving sulfate, soot, dust and sea salt particles are included based on previous studies (Li et al., 2015; Li et al.,
139 2012). Size-resolved mineral dust emissions are calculated online as a function of the relative humidity, frictional velocity,
140 mineral particle size distribution and surface roughness (Li et al., 2012). Sea salt emissions are calculated with the scheme of
141 Athanasopoulou et al. (2008). The dry deposition of gases and aerosols is modelled based on the scheme of Wesely (1989),
142 and advection is simulated with the accurate mass conservation algorithm of Walcek and Aleksic (1998).

143 Figure 1 shows the modelling domain of this study, which covers most parts of East Asia with a fine horizontal resolution
144 of 15 km. The vertical coordinate system consists of 20 terrain-following levels with the model top reaching up to 20000 m
145 and the first layer at approximately 50 m. Nine vertical layers are set within 2 km of the surface to better characterize the
146 vertical mixing process within the boundary layers. The emissions of air pollutants considered in this study include the monthly
147 anthropogenic emissions retrieved from the Hemispheric Transport of Air Pollution (HTAP) v2.2 emission inventory with a
148 base year of 2010 (Janssens-Maenhout et al., 2015), biomass burning emissions retrieved from the Global Fire Emissions
149 Database (GFED) version 4 (Randerson et al., 2017; van der Werf et al., 2010), biogenic volatile organic compound (BVOC)
150 emissions retrieved from the Model of Emissions of Gases and Aerosols from Nature (MEGAN)-MACC (Sindelarova et al.,
151 2014), marine VOC emissions retrieved from the POET database (Granier et al., 2005), soil NO_x emissions retrieved from the
152 Regional Emission Inventory in Asia (Yan et al., 2003) and lightning NO_x emissions retrieved from Price et al., 1997. Clean
153 initial conditions are used in the air quality simulations with a two-week free run of the NAQPMS model as the spin-up time.
154 The top and boundary conditions are provided by the Model for Ozone and Related Chemical Tracers (MOZART; Brasseur et
155 al., 1998; Hauglustaine et al., 1998) model, and the meteorological fields are provided by the WRF model. In each daily
156 meteorology simulation, a 36-h free run of the WRF model is conducted with the first 12-h simulation period as the spin-up
157 run and the remaining 24-h period providing the meteorologic inputs for the NAQPMS model. The initial and boundary
158 conditions for the meteorology simulations are provided by the NCAR/NCEP 1° × 1° reanalysis data.

159 **2.2 Generation of ensemble simulation**

160 The EnKF uses an ensemble of model simulation to represent the forecast uncertainty which should include the most
161 model uncertain aspects. Considering that the emissions are a major source of uncertainty in air quality prediction (Carmichael
162 et al., 2008; Hanna et al., 1998; Li et al., 2017), in this study the ensemble were generated by perturbing the emissions based
163 on their error probability distribution functions (PDFs) which were assumed to be Gaussian distributions. Table 1 lists the
164 perturbed species considered in this study as well as their corresponding emission uncertainties obtained from previous studies.

165 The perturbed emissions were parameterized by multiplying the base emissions with a perturbation factor β , as expressed in
166 Eq. (1):

$$167 \mathbf{E}_i = \mathbf{E} \circ \beta_i, i = 1, 2, \dots, N \quad (1)$$

168 where \mathbf{E} denotes the vector of base emissions, \circ denotes the Schur product and N denotes the ensemble size. The performance
169 of the EnKF is strongly related to the ensemble size which determines the accuracy to which the background error covariance
170 is approximated (Constantinescu et al., 2007; Miyazaki et al., 2012). A large ensemble size is important in capturing the proper
171 background error covariance structure, especially in high-resolution data assimilation application due to the fine-scale
172 variability and large degree of freedoms. However, a large ensemble is computationally expensive as the cost of EnKF linearly
173 increases with ensemble size while the accuracy of covariance estimate improves by its square root (Constantinescu et al.,
174 2007). Thus, an appropriate ensemble should keep a good balance between accuracy and computational cost. Constantinescu
175 et al. (2007) in their ideal experiments showed that a 50-member ensemble has significant improvement against smaller
176 ensembles, and Miyazaki et al. (2012) in their real chemical assimilation experiments showed that the improvement was much
177 less significantly by further increasing the ensemble size from 48 to 64. Thus, the ensemble size was chosen as 50 in this study
178 by referencing pervious publications and also our previous high-resolution regional assimilation work (Tang et al., 2011; Tang
179 et al., 2013; Tang et al., 2016) which showed that a 50-member ensemble keeps good balance between assimilation
180 performance and computational efficiency. However, it should be noted that our application has higher horizontal resolution
181 than that of Constantinescu et al. (2007) and Miyazaki et al. (2012), which may require larger ensemble size due to the larger
182 degree of freedoms in our application. Thus, to reduce the degree of freedoms in our high-resolution data assimilation work,
183 we assumed that the emission errors were spatially correlated, and an isotropic correlation model was assumed in the
184 covariance of the emission errors, which is written as:

$$185 \rho(i, j) = \exp \left\{ -\frac{1}{2} \left[\frac{h(i, j)}{l} \right]^2 \right\} \quad (2)$$

186 where $\rho(i, j)$ represents the correlation between grids i and j , $h(i, j)$ is the distance between these two points and l is the
187 decorrelation length, which was specified as 150 km in this study. According to the PDF of the emission errors, β follows the
188 same Gaussian distribution as that of the emission errors except that its mean equals 1. Using the method of Evensen (1994),
189 fifty smooth pseudorandom perturbation fields of β were generated for each perturbed species. In addition, the emission
190 perturbations were kept independent from each other to prevent pseudo-correlation among the different species.

191 2.3 Observations

192 Surface observations of the hourly ambient PM_{2.5}, PM₁₀, SO₂, NO₂, CO and O₃ concentrations retrieved from the CNEMC
193 were used in this study. The number of observation sites was approximately 510 in 2013 and increased to 1436 in 2015. Real-
194 time observations of these six air pollutants at each monitoring site are routinely gathered by the CNEMC and released to the
195 public (available at <http://www.cnemc.cn/>; last accessed: 17 April 2020) at hourly intervals. A challenge that should be

196 overcome in the assimilations of surface observation is that there are occasional outliers occurring in these observations due
 197 to the instrument malfunctions, influences of harsh environments and limitation of measurement method. Filtering out these
 198 outliers is necessary before the assimilation, otherwise these outliers may cause unrealistic spatial and temporal variations in
 199 the reanalysis. To address this issue, a fully automatic outlier detection method was developed by Wu et al. (2018) to filter out
 200 the observation outliers. An automatic outlier detection method is very important in chemical data assimilation since there is
 201 a large amount of observation data on multiple species. Four types of outliers characterized by temporal and spatial
 202 inconsistencies, instrument-induced low variances, periodic calibration exceptions and lower PM₁₀ concentrations than those
 203 of PM_{2.5} were detected and removed before the assimilation. Figure A1 in Appendix A shows the removal ratios of the six air
 204 pollutants from 2013 to 2018, which are generally around 1.5% for most air pollutants throughout the assimilation period. The
 205 PM₁₀ observations have a high removal ratio (9–13%) during 2013–2015 with most of these outliers marked by lower PM₁₀
 206 concentrations than those of PM_{2.5}. However, there was a sharp decrease in removal ratios of PM₁₀ in 2016 (~1.5%) because
 207 of the implementation of a compensation algorithm for the loss of semi-volatile materials in the PM₁₀ measurements (Wu et
 208 al., 2018). To assess the potential impacts of outlier detection on the assimilations, the differences in annual concentrations
 209 caused by quality control are shown in Fig. A2. The differences were generally positive for PM_{2.5}, SO₂, NO₂ and CO
 210 concentrations, indicating a lower tendency of these species' concentrations due to the use of outlier detection. Negative
 211 differences were mainly found in the PM₁₀ concentrations in south China and the O₃ concentrations throughout China.
 212 According to estimation, the impacts of outlier detection were generally small in most stations. The differences were less than
 213 5 µg/m³ (1 µg/m³) for PM_{2.5} concentrations over most stations in north (south) China and less than 1 µg/m³ for the gaseous
 214 air pollutants for most stations throughout China. The differences were shown to be relative larger for PM₁₀ concentrations
 215 over northwest China which can be over 20 µg/m³ in stations around Taklimakan Desert. This would be due to the higher
 216 outlier ratios in the observations over the remote areas. More details on the outlier detection method were available in Wu et
 217 al. (2018).

218 A proper estimate of the observation error is important in regard to the filter performance since the observation and
 219 background errors determine the relative weights of the observation and background values in the analysis. The observation
 220 error includes measurement and representativeness errors. For each species, the measurement error was given by their
 221 respective instruments, namely, 5% for PM_{2.5} and PM₁₀, 2% for SO₂, NO₂ and CO, and 4% for O₃ according to officially
 222 released documents of the Chinese Ministry of Ecology and Environmental Protection (HJ 193–2013 and HJ 654–2013,
 223 available at <http://www.cnemc.cn/jcgf/dqhj/>; last accessed: 17 April 2020). The representativeness error arises from the
 224 different spatial scales that the gridded model results and discrete observations represent, which is parameterized by the
 225 formula proposed by Elbern et al., (2007) in this study:

$$226 \quad r_{repr} = \sqrt{\frac{\Delta x}{L_{repr}}} \times \epsilon^{abs} \quad (3)$$

227 where r_{repr} represents the representativeness error, Δx represents the model resolution, L_{repr} represents the characteristic
 228 representativeness length of the observation site and ϵ^{abs} represents the error characteristic parameters for different species.

229 The estimation of L_{repr} is dependent on the types of observation sites with urban sites usually having smaller representative
 230 length than the rural sites have due to the larger representativeness errors. Considering that the observation sites from CNEMC
 231 were almost city (urban) sites (>90%), the L_{repr} was assigned to be 2km in this study according to Elbern et al., 2007.

232 For the estimations of ε^{abs} , previous studies (Chen et al., 2019; Feng et al., 2018; Jiang et al., 2013; Ma et al., 2019;
 233 Pagowski and Grell, 2012; Peng et al., 2017; Werner et al., 2019) usually assigned the ε^{abs} empirically to be half of the
 234 measurement error following the study by Pagowski et al. (2010). In this study, the ε^{abs} was obtained from Li et al. (2019)
 235 who estimated the ε^{abs} based on a dense observation network in Beijing-Tianjin-Hebei region. In their study, the
 236 representativeness error of each species' observation was first estimated by the spatiotemporal averaged standard deviation of
 237 the observed values within a 30km×30km grid:

$$238 \quad r_{repr,i} = \frac{1}{MT} \sum_{m=1}^M \sum_{t=1}^T S_{m,t,i} \quad (4)$$

239 where $r_{repr,i}$ represents the representativeness errors of the observations for species i , $S_{m,t,i}$ represents the standard deviation
 240 of the observed values of species i at different sites that are located in a same grid m at time t , M and T represents the total
 241 number of grid and observation time. After the estimations of $r_{repr,i}$, the ε_i^{abs} for species i were estimated by a transformation
 242 of Eq. (3):

$$243 \quad \varepsilon_i^{abs} = r_{repr,i} / \sqrt{\frac{\Delta x}{L_{repr}}} \quad (5)$$

244 where Δx is equal to 30km. Based on the estimated L_{repr} and the ε_i^{abs} for different species, the representativeness errors are
 245 estimated using Eq. (3) by specifying the Δx to be 15km.

246 2.4 Data assimilation algorithm

247 We used a variant of the EnKF approach, i.e., the local ensemble transform Kalman filter (LETKF; Hunt et al., 2007), to
 248 assimilate the observations into the model state. The LETKF has several advantages over the original EnKF (e.g., Miyazaki
 249 et al., 2012). As a kind of deterministic filter, it does not need to perturb the observations, which avoids introducing additional
 250 sampling errors. In addition, the LETKF performs the analysis locally in space and time, which not only alleviates the rank
 251 problem of the EnKF method but also suppresses the long-distance spurious correlation caused by the limited ensemble size.
 252 The formulation of the LETKF can be written as:

$$253 \quad \bar{\mathbf{x}}^a = \bar{\mathbf{x}}^b + \mathbf{X}^b \bar{\mathbf{w}}^a \quad (6)$$

$$254 \quad \bar{\mathbf{w}}^a = \tilde{\mathbf{P}}^a (\mathbf{H}\mathbf{X}^b)^T \mathbf{R}^{-1} (\mathbf{y}^o - \mathbf{H}\bar{\mathbf{x}}^b) \quad (7)$$

$$255 \quad \tilde{\mathbf{P}}^a = \left[\frac{(N_{ens}-1)I}{1+\lambda} + (\mathbf{H}\mathbf{X}^b)^T \mathbf{R}^{-1} (\mathbf{H}\mathbf{X}^b) \right]^{-1} \quad (8)$$

$$256 \quad \bar{\mathbf{x}}^b = \frac{1}{N_{ens}} \sum_{i=1}^{N_{ens}} \mathbf{x}_i^b ; \mathbf{X}_i^b = \frac{1}{\sqrt{N-1}} (\mathbf{x}_i^b - \bar{\mathbf{x}}^b) \quad (9)$$

257 where $\bar{\mathbf{x}}^a$ is the analysis state, $\bar{\mathbf{x}}^b$ is the background state, \mathbf{X}^b represents the background perturbations, $\bar{\mathbf{w}}^a$ is the analysis in
 258 the ensemble space spanned by \mathbf{X}^b , $\bar{\mathbf{P}}^a$ is the analysis error covariance in the ensemble space with dimensions of $N_{ens} \times N_{ens}$,
 259 \mathbf{y}^o is the vector of observations used in the analysis of this grid, \mathbf{R} is the observation error covariance matrix, and \mathbf{H} is the
 260 linear observational operator that maps the model space to the observation space. The scalar λ in Eq. (8) denotes the inflation
 261 factor for the background covariance matrix, which was estimated with the algorithm proposed by Wang and Bishop (2003):

$$262 \quad \lambda = \frac{(\mathbf{R}^{-1/2}\mathbf{d})^T \mathbf{R}^{-1/2}\mathbf{d} - p}{\text{trace}\{\mathbf{R}^{-1/2}\mathbf{H}\mathbf{P}^b(\mathbf{R}^{-1/2}\mathbf{H})^T\}} \quad (10)$$

$$263 \quad \mathbf{d} = \mathbf{y}^o - \mathbf{H}\bar{\mathbf{x}}^b \quad (11)$$

$$264 \quad \mathbf{P}^b = \mathbf{X}^b(\mathbf{X}^b)^T \quad (12)$$

265 where \mathbf{d} represents the residuals, p is the number of observations, \mathbf{P}^b is the ensemble-estimated background error covariance
 266 matrix, and the trace of the covariance matrix is used to approximate covariance on a globally averaged basis. The inflation is
 267 necessary for the ensemble-based assimilation algorithm since the ensemble-estimated background error covariance is very
 268 likely to underestimate the true background error covariance due to the limited ensemble size and occurrence of the model
 269 error (Liang et al., 2012). Without any treatment to prevent background error covariance underestimation, the model forecast
 270 would be overconfident and eventually result in filter divergence. Using Eq. (10), the hourly inflation factor was calculated
 271 for each species. In addition, the inflation factor was calculated locally in this study. Thus, the inflation factor used in this
 272 assimilation is not only species specific, but also varies with time and space, which reflects different error characteristics of
 273 the different species in different time and places.

274 Besides, the inter-species correlation was neglected in the background error covariance similar to previous chemical data
 275 assimilation studies (e.g., Inness et al., 2015; Inness et al., 2019; Ma et al., 2019) although Miyazaki et al. (2012) has shown
 276 the benefits of including correlations between the background errors of different chemical species. This is, on the one hand, to
 277 avoid the effects of the spurious correlation between non or weakly related variables. On the other hand, different from
 278 Miyazaki et al., 2012, this study concentrated on the assimilations of primary air pollutants (except of O_3) whose errors are
 279 more related to the errors in their emissions. Since the emission errors of these species were considered to be independent in
 280 this study (Sect. 2.2), thus the correlation between background errors of different species were generally near zero for most
 281 cases as shown in Figs. B1-2 in Appendix B. The high correlations only occur in background errors of $\text{PM}_{2.5}$ and PM_{10} as well
 282 as those of NO_2 and O_3 . The high positive correlation between $\text{PM}_{2.5}$ and PM_{10} is just because $\text{PM}_{2.5}$ is a part of PM_{10} , and
 283 there would be redundant information in the observations of $\text{PM}_{2.5}$ and PM_{10} concentrations, thus we did not include the
 284 correlation between the $\text{PM}_{2.5}$ and PM_{10} concentrations in the assimilation. The negative correlation between the O_3 and NO_2
 285 is due to the NO_x -OH- O_3 chemical reactions in the NO_x saturated conditions that the increases of NO_2 concentrations would
 286 reduce the O_3 concentrations due to the enhanced NO titration effect. However, the relationship between O_3 and NO_2
 287 concentrations is actually nonlinear depending on the NO_x limited or saturated conditions (Sillman, 1999), and previous study
 288 by Tang et al. 2016 has shown the limitations of the EnKF under strong nonlinear relationships. The cross-variable data

289 assimilations of O₃ and NO₂ may come up with inefficient or even wrong adjustments. Considering the nonlinear relationship
 290 between the O₃ and NO₂ concentrations and their unexpected effects on EnKF, we took a conservative way in the assimilations
 291 of NO₂ and O₃ by neglecting their error correlations. This would also make different species be assimilated in a consistent way.
 292 Therefore, in this study each air pollutant is assimilated independently by only using the observations of this pollutant.

293 Figure 2 shows the local scheme we used in the assimilation, where the plus and dot symbols indicate the centres of the
 294 model grids and locations of the observation sites, respectively. In each model grid, only the observation sites located within
 295 a $(2l + 1)$ by $(2l + 1)$ rectangular area centred at this model grid were considered in the calculations of its analysis. The cut-
 296 off radius l was chosen as 12 model grids, approximately 180 km at the 15-km horizontal resolution. The use of a cut-off
 297 radius, however, could cause analysis discontinuities when an observation enters or leaves the local domain when moving
 298 from one model grid to another (Sakov and Bertino, 2011). To increase the smoothness of the analysis state, following Hunt
 299 et al. (2007), we artificially reduced the impact of the observations close to the boundary of the local domain by multiplying
 300 the entries in \mathbf{R}^{-1} by a factor decaying from one to zero with increasing distance of the observation from the central model
 301 grid. The decay factors used in this study are calculated by:

$$302 \quad \rho(i) = \exp\left\{-\frac{h(i)^2}{2L^2}\right\} \quad (13)$$

303 where $\rho(i)$ is the decay factor for observation i , $h(i)$ is the distance between observation i and the central model grid point,
 304 and L is the decorrelation length chosen as 80 km, smaller than the cut-off radius, to increase the smoothness of the analysis
 305 state. Typically, only the state of the central model grid is updated and used to construct the global analysis field. However,
 306 experience has shown that an observable discontinuity remains in the analysis over certain regions. To address this issue,
 307 following the method of Ott et al. (2004), we simultaneously updated the state of a small patch ($l=1$) around the central model
 308 grid (the updated region in Fig. 2) at each local analysis step. The final analysis of a given model grid was then obtained as the
 309 weighted mean of all the analysis values of this model grid. A weighted mean was necessary since the analysis of the different
 310 patches adopted different decay factors for the observation error. The weight of each analysis value in model grid i is calculated
 311 by Eq. (14):

$$312 \quad W_{i,j} = \frac{\exp\left(-\frac{h(i,j)^2}{L^2}\right)}{\sum_{j=1}^m \exp\left(-\frac{h(i,j)^2}{L^2}\right)} \quad (14)$$

313 where $h(i,j)$ is the distance of model grid i to the central model grid of the patch generating the j th analysis value of this grid,
 314 m is the number of patches containing this model grid and L is the decorrelation length, which was chosen as 80 km in this
 315 study.

316

317 3 Data assimilation statistics

318 3.1 χ^2 diagnosis

319 We first applied the χ^2 test to demonstrate the performance of our data assimilation system, which is important in
320 evaluating the reanalysis (Miyazaki et al., 2015). The χ^2 diagnosis is a robust criterion for validating the estimated background
321 and observation error covariance in the data assimilation (e.g. Menard et al., 2000; Miyazaki et al., 2015; Miyazaki et al.,
322 2012), which is estimated by comparing the sample covariance of observation minus forecast (OmF) with the sum of estimated
323 background and observation error covariance in the observational space ($\mathbf{HBH}^T + \mathbf{R}$):

$$324 \mathbf{Y} = \frac{1}{\sqrt{m}} (\mathbf{HBH}^T + \mathbf{R})^{-\frac{1}{2}} (\mathbf{y}^o - \mathbf{HX}^b) \quad (15)$$

$$325 \chi^2 = \mathbf{Y}^T \mathbf{Y} \quad (16)$$

326 where m is the number of observations. According to the Kalman filtering theory, the mean of χ^2 should approach 1 if the
327 background and observation error covariances are properly specified, while values greater (lower) than 1 indicates the
328 underestimation (overestimation) of the observation and/or background error covariance.

329 Figure 3 shows the time series of the monthly χ^2 values (black lines) for different species as well as the number of
330 assimilated observations per month (blue bars). The mean values of χ^2 are generally within 50% difference from the ideal
331 value of 1 for PM_{2.5}, PM₁₀, NO₂ and O₃, which suggests that the observation and background error covariance are generally
332 well specified in the analysis of these species. Although the χ^2 values for these species showed pronounced seasonal variations
333 that reflects the different error characteristics in different seasons, the χ^2 values were roughly stable for PM_{2.5} and O₃
334 throughout the assimilation periods, and for NO₂ and PM₁₀ after 2015 when the number of assimilated observations become
335 stable, which generally shows the long-term stability of the performance of data assimilation. The χ^2 values for SO₂ were
336 nevertheless greater than 1 in most cases, especially before 2017. This would be more relevant to the underestimations of
337 background error covariance of SO₂ as we only specified 12% uncertainty in the SO₂ emissions, suggesting that the emission
338 uncertainty of SO₂ may be underestimated by Zhang et al. (2009). There were also pronounced annual trends in the χ^2 values
339 of SO₂, which may be attributed to the increases of observation number from 2013 to 2014 and the substantial decreases of
340 SO₂ observations from 2013 to 2018. Although smaller than the χ^2 values of SO₂, the values for CO were greater than 1 in
341 most cases, suggesting the underestimations of the error covariances. Similar to the χ^2 values of SO₂, obvious decreasing trend
342 can also be found in the χ^2 values of CO. These results suggest that our data assimilation system has relatively poor
343 performance in the analysis of CO and SO₂ concentrations than the other four species, which is consistent with the cross-
344 validation results (Sect. 4.2.2) that showed smaller R^2 values for the reanalysis data of CO and SO₂ concentrations. The annual
345 trend of χ^2 values in CO and SO₂ also indicates relatively weak stability in the performance of data assimilation system on
346 assimilating CO and SO₂ observations, which may influence the analysis of the annual trends in these two species.

347 **3.2 OmF & OmA analysis**

348 Spatial distributions of six-year averaged OmF and observation minus analysis (OmA) for each species in the observation
349 space were then analysed to investigate the structure of forecast bias and to measure the improvement in the reanalysis (Fig.
350 4). The analysis increment, which is estimated from the differences between the analysis and forecast, is also plotted to measure
351 the adjustments made in the model space. The OmF values have showed persistent positive model biases (i.e., negative OmF)
352 in the PM_{2.5} and SO₂ concentrations in east China, as well as PM₁₀ and O₃ concentrations in south China. The negative model
353 biases (i.e., positive OmF) were mainly found in the PM_{2.5} concentrations in west China, the PM₁₀ concentrations in north
354 China, the O₃ concentrations in central-east China, as well as the concentrations of CO and NO₂ throughout the whole China.

355 The OmA values suggest that the data assimilation removes most of the model biases for each species, which confirms
356 the good performance of our data assimilation system. According to Fig. C1 in Appendix C, the monthly mean OmF biases
357 were almost completely removed in each regions of China because of the assimilation, with mean OmF biases reducing by
358 32–94% for PM_{2.5}, 33–83% for PM₁₀, 25–96% for SO₂, 53–88% for NO₂, 88–97% for CO and 54–90% for O₃ concentrations
359 in different regions of China. The mean OmF root mean square error (RMSE) were also reduced substantially by 80–93% for
360 PM_{2.5}, 80–86% for PM₁₀, 73–96% for SO₂, 76–91% for NO₂, 88–96% for CO and 76–87% for O₃ concentrations in different
361 regions of China (Fig. C2). In addition, despite the mean OmF bias and OmF RMSE exhibit significant annual trend, the OmA
362 bias and OmA RMSE are relatively stable during the assimilation period, which generally confirms the long-term stability of
363 our data assimilation system.

364 The spatial patterns of analysis increment were in good agreement with those of the OmF values for each species, which
365 generally shows negative (positive) increments for PM_{2.5} concentrations in east (west) China, negative (positive) increments
366 for PM₁₀ concentrations in south (north) China, negative increments for SO₂ throughout the China, positive increments for CO
367 and NO₂ concentrations throughout the China, and the positive (negative) increments for O₃ concentrations in central-east
368 (south) China. These results confirm that the data assimilation can effectively propagate the observation information into the
369 model state and reduce the model errors.

370 **4 Evaluation Results**

371 In this section, we present the fields of the CAQRA dataset and compare them to the observations. It aims to provide a
372 brief introduction to the CAQRA dataset and gives a first assessment of the quality of this dataset. The cross-validation (CV)
373 method was applied in the assessment of the CAQRA dataset, in which a proportion of the observation data was withheld from
374 the data assimilation process and adopted as a validation dataset. We conducted five CV experiments by randomly dividing
375 the observation sites of the CNEMC into five groups (with 20% of the observation sites in each group). In each experiment,
376 the analysis was performed with one group of the observation data omitted in the assimilation process. Analysis results at the
377 validation sites, i.e., the observation sites not used in the assimilation process, were then collected and used to validate the
378 assimilation results. For convenience, the analysis results at the validation sites of the five CV experiments were combined

379 and comprised a validation dataset containing all observation sites (the CV run). This dataset was then evaluated against the
380 observations to assess the quality of the CAQRA dataset. In addition, independent PM_{2.5} observations retrieved from the U.S.
381 Department State Air Quality Monitoring Program over China were also employed in the assessment of the PM_{2.5} reanalysis
382 field. The quality of the CAQRA dataset was assessed at different spatial and temporal scales to better understand the CAQRA
383 dataset. Additionally, the validation results of the ensemble mean of the simulations without assimilation (the base simulation)
384 are provided to highlight the impacts of assimilation.

385 **4.1 Particulate matter (PM)**

386 **4.1.1 Spatial distribution of the PM reanalysis data over China**

387 We first present the reanalysis fields of the PM concentrations (PM_{2.5} and PM₁₀) in China. Figure 5 shows the six-year
388 mean (2013–2018) spatial distribution of the PM_{2.5} concentration in China obtained from the CAQRA dataset, base simulation
389 and observations. The CAQRA dataset provides a continuous map of the PM_{2.5} concentration in China and suitably reproduces
390 the observed magnitude of the PM_{2.5} concentration in China. The highest PM_{2.5} concentrations were observed in the NCP
391 region due to its intensive industrial activities and the associated high emissions of PM_{2.5} and its precursors (Qi et al., 2017).
392 High PM_{2.5} concentrations were also found in the SE region, where the PM_{2.5} concentration is influenced by both local
393 emissions and the long-range transport of air pollutants from northern China (Lu et al., 2017). In the NW region, in addition
394 to hotspots exhibiting high PM_{2.5} concentrations in large cities, high PM_{2.5} concentrations were observed in the Taklimakan
395 Desert due to the influences of dust emissions. The observed magnitude and spatial variability of the PM₁₀ concentration were
396 also represented well by the PM₁₀ reanalysis field. In general, the spatial distributions of the PM₁₀ reanalysis were similar to
397 those of the PM_{2.5} reanalysis except in Gansu and Ningxia provinces, where high PM₁₀ concentrations and relatively low PM_{2.5}
398 concentrations occurred. This may be related to the large contributions of dust emissions in these areas. The base simulation
399 notably overestimated the PM_{2.5} and PM₁₀ concentrations in China. This may occur due to the systematic biases in the emission
400 inventory (Kong et al., 2019) and because negative trends of PM and its precursor emissions were not considered in our
401 simulations. In addition, the PM_{2.5} concentration hotspots in the NW region and Tibetan Plateau were not captured in the base
402 simulation, possibly due to the absence of emissions in these remote regions.

403 Seasonal maps of the PM_{2.5} and PM₁₀ concentrations are shown in Figs. D1–2 in the Appendix D, which reveal profound
404 seasonal variations. Both the PM_{2.5} and PM₁₀ concentrations exhibit maximum values in winter in most regions of China due
405 to the increased anthropogenic emissions related to enhanced power generation, industrial activities and fossil fuel burning for
406 heating purposes (Li et al., 2017). Unfavourable meteorological conditions with stable boundary conditions also contribute to
407 the high PM concentrations in winter. In contrast, due to the low emission rate and intense mixing processes, the PM
408 concentrations are the lowest in summer. The PM concentrations in the Taklimakan Desert exhibit a different seasonality, with
409 the highest PM concentrations occurring in spring and the lowest levels occurring in winter. This occurs because the major
410 PM sources in the Taklimakan Desert are not anthropogenic emissions but dust emissions, which are usually the highest in

411 spring due to the frequent strong dust storms. Figure 6 further shows an example of the hourly PM reanalysis results, including
412 a year-round time series of the site mean hourly PM concentrations in Beijing. This figure shows that PM reanalysis suitably
413 captures the hourly evolution of the PM concentrations. Both the heavy haze episodes during the wintertime and the strong
414 dust storms during the springtime are represented well in PM reanalysis.

415 4.1.2 Assessment of the PM reanalysis data over China

416 The CV method was used to assess the quality of the PM reanalysis data over China. Table 2 summarizes the site-based
417 CV results for the reanalysis data from 2013 to 2018 at the different temporal scales. It should be mentioned that these sites
418 are all validation sites not used in the data assimilation process. The validation results indicated that by assimilating the surface
419 PM concentrations, the reanalysis data exhibit a relatively high performance in reproducing the magnitude and variability of
420 the surface PM concentrations in China. The CV R^2 values were up to 0.81 and 0.72 in regard to the hourly $PM_{2.5}$ and PM_{10}
421 concentrations, respectively, which were much higher than the values of 0.26 and 0.17, respectively, in the base simulation.
422 The bias was substantially reduced in the $PM_{2.5}$ and PM_{10} reanalysis data with CV mean bias (MBE) values of approximately
423 $-2.6 \mu\text{g}/\text{m}^3$ (-4.9%) and $-6.8 \mu\text{g}/\text{m}^3$ (-8.7%), respectively, at the hourly scale, much smaller than the large bias in the base
424 simulation. The CV RMSE values were only approximately half of the base simulation RMSE values, which were
425 approximately 17.6 and $39.3 \mu\text{g}/\text{m}^3$ for the hourly $PM_{2.5}$ and PM_{10} concentrations, respectively. The reanalysis data showed
426 a good performance at the daily, monthly and yearly scales, with CV RMSE values ranging from 9.0 to $15.1 \mu\text{g}/\text{m}^3$ for the
427 $PM_{2.5}$ concentration and from 19.1 to $28.8 \mu\text{g}/\text{m}^3$ for the PM_{10} concentration.

428 The quality of the $PM_{2.5}$ and PM_{10} reanalysis data in the different regions of China is further summarized in Appendix E
429 Tables E1-2. At the hourly scale, small negative biases of the $PM_{2.5}$ reanalysis data were found in the NCP (-4.8%), NE (-
430 5.8%), SE (-3.8%) and SW (-3.4%) regions. The biases in the NW and central regions were relatively large, with CV
431 normalized mean bias (CV NMB) values of approximately -7.3% and -8.2%, respectively. Two reasons might explain the
432 large biases in these two regions. First, the observation sites are sparse in the NW and central regions. As a result, the $PM_{2.5}$
433 concentration is not suitably constrained at certain sites in the CV method. Second, the emissions of $PM_{2.5}$ and its precursors
434 might be very low in these two regions, leading to underestimation of the background errors since we only considered the
435 emission uncertainty in the ensemble simulations. Although this problem was alleviated by using the inflation technique to
436 compensate for the missing errors, the overconfident model results still degraded the assimilation performance to a certain
437 extent, making the analysis less influenced by the observations. The errors of the $PM_{2.5}$ reanalysis data exhibited apparent
438 spatial differences (Table E1). The CV RMSE values were the smallest in the SE ($14.9 \mu\text{g}/\text{m}^3$) and SW ($16.5 \mu\text{g}/\text{m}^3$) regions
439 and increased to $\sim 25 \mu\text{g}/\text{m}^3$ in the NCP, NE and central regions. Consistent with the bias distributions, the largest CV RMSE
440 value was found in the NW region, which reached $52.1 \mu\text{g}/\text{m}^3$ but was still much smaller than the RMSE value of the base
441 simulation ($73.0 \mu\text{g}/\text{m}^3$). The errors of the $PM_{2.5}$ reanalysis data were small at the daily, monthly and yearly scales, with CV
442 RMSE values of approximately $10.6\text{--}39.4 \mu\text{g}/\text{m}^3$ at the daily scale, $7.4\text{--}26.9 \mu\text{g}/\text{m}^3$ at the monthly scale and $6.1\text{--}23.5 \mu\text{g}/\text{m}^3$

443 at the yearly scale. In terms of the hourly PM₁₀ reanalysis data, the CV results (Table E2) indicated that small negative biases
444 occurred in the NCP, NE, SE and SW regions, ranging from -9.6% (NE region) to -5.9% (SE region). The biases were larger
445 in the NW and central regions, with the CV NBM values increasing to approximately 18.0% and 14.1%, respectively. The
446 errors of the PM₁₀ reanalysis data also exhibited a spatial heterogeneity. The CV RMSE value was the smallest in the SE (26.0
447 $\mu\text{g}/\text{m}^3$) and SW (30.2 $\mu\text{g}/\text{m}^3$) regions and increased to approximately 39.8 and 43.7 $\mu\text{g}/\text{m}^3$ in the NE and NCP regions,
448 respectively. The largest errors were found in the central and NW regions, with CV RMSE values of approximately 105.5 and
449 57.3 $\mu\text{g}/\text{m}^3$, respectively. The PM₁₀ reanalysis data revealed small errors at the daily, monthly and yearly scales, with CV
450 RMSE values of approximately 18.6–85.5 $\mu\text{g}/\text{m}^3$ at the daily scale, 13.7–64.0 $\mu\text{g}/\text{m}^3$ at the monthly scale and 12.3–55.8
451 $\mu\text{g}/\text{m}^3$ at the yearly scale.

452 **4.1.3 Trend study of the PM reanalysis data over China**

453 A realistic representation of the observed interannual change is another important aspect of the reanalysis dataset. The
454 performance of the reanalysis data in representing the observed interannual changes in the PM_{2.5} and PM₁₀ concentrations was
455 thus evaluated nationwide and in the different regions of China. Figures 7–8 show time series of the monthly mean PM_{2.5} and
456 PM₁₀ concentrations nationwide and in the different regions. The observed national PM_{2.5} concentration revealed a profound
457 seasonal cycle with the highest concentration in winter and the lowest level in summer. The annual trends of the PM_{2.5} and
458 PM₁₀ concentrations were also calculated using the Mann-Kendall (M-K) trend test and the Theil-Sen trend estimation method,
459 which are summarized in Table 3. A significant negative trend was observed in the PM_{2.5} concentration nationwide, with a
460 calculated annual trend of approximately -5.8 ($p < 0.05$) $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{yr}^{-1}$. The NE and NCP regions exhibited the highest
461 negative trends among the six regions, with calculated trends of approximately -7.5 ($p < 0.05$) and -7.0 ($p < 0.05$) $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{yr}^{-1}$,
462 respectively. In the other regions, the negative trends ranged from -6.3 to -5.2 $\mu\text{g} \cdot \text{m}^{-3} \cdot \text{yr}^{-1}$. The base simulation suitably
463 reproduced the observed seasonal cycle of the PM_{2.5} concentration in all regions. The magnitude of the PM_{2.5} concentration in
464 2013 was also captured well in the different regions, suggesting that the emission inventories of 2010 were generally reasonable
465 for the simulation of the PM_{2.5} concentration in 2013. However, starting from 2014, the base simulation tended to overestimate
466 the observations in the NCP, SE and SW regions, indicating that the emission inventory of 2010 may be too high for the
467 simulation of the PM_{2.5} concentration in these regions after 2014. In contrast, the base simulation significantly underestimated
468 the PM_{2.5} concentration in the NW region. The model performance of the base simulation was relatively good in the NE and
469 central regions throughout the six years. Although the base simulation captured the negative trends of the observed PM_{2.5}
470 concentration in China and the different regions, the simulated trends were much lower than those indicated by the observations.
471 Since we adopted the same emission inventory in the simulations of the air pollutants in the different years, the simulated
472 trends in the base simulation were only driven by the variations in meteorological conditions. This suggests that the change in
473 meteorological conditions only explained a small proportion of the negative trends in the PM_{2.5} concentration in China and
474 that emission reductions contributed more to the decline in the PM_{2.5} concentration. The CV run agreed better with the
475 observations. The observed trends of the PM_{2.5} concentration in China and each subregion were all suitably captured by the

476 reanalysis in the CV run. Similar results were obtained for the analysis of the trend of the PM₁₀ concentration, as shown in Fig.
477 8. The observed PM₁₀ concentration also exhibited significant negative trends, which were captured well by the PM₁₀ reanalysis
478 in the CV run. The base simulation attained a better performance in reproducing the PM₁₀ concentration in China than in
479 reproducing the PM_{2.5} concentration, while significant underestimations of the PM₁₀ concentration occurred in the NW and
480 central regions. The calculated negative trends of the base simulation were still lower than those indicated by the observations.
481 This again highlights the large contributions of emission reduction to the improvement of the air quality in China in these years.

482 **4.1.4 Independent validation of the PM_{2.5} reanalysis data**

483 In addition to the CV method, the PM_{2.5} reanalysis data were further validated against an independent dataset acquired
484 from the U.S. Department State Air Quality Monitoring Program over China (<http://www.stateair.net/>; last accessed: 17 April
485 2020), which contains the hourly PM_{2.5} concentration in Beijing, Chengdu, Guangzhou, Shanghai and Shenyang cities. Table
486 4 presents a comparison of the observed PM_{2.5} concentrations to those obtained from the CAQRA dataset and base simulation.
487 The results indicated that the magnitude and variability of the PM_{2.5} reanalysis data agreed better with those of the observed
488 PM_{2.5} concentrations in all cities. Both the MBE and RMSE values were greatly reduced in the CAQRA dataset, which only
489 ranged from -7.1 to -0.3 $\mu\text{g} \cdot \text{m}^{-3}$ and from 16.8 to 33.6 $\mu\text{g} \cdot \text{m}^{-3}$, respectively, in these cities. The correlation coefficient was
490 also greatly improved in CAQRA ($R^2 = 0.74\text{--}0.86$) over the base simulation ($R^2 = 0.09\text{--}0.38$). These results confirm that the
491 CAQRA dataset attains a high quality performance in representing the PM_{2.5} pollution in China in these years.

492 **4.1.5 Comparison to the satellite-estimated PM_{2.5} concentration**

493 Previous studies have shown that estimating the ground-based PM_{2.5} concentration from the satellite-derived AOD is an
494 effective way to map the PM_{2.5} concentration with a good accuracy. To further demonstrate the accuracy of our PM_{2.5} reanalysis
495 data, we also compared the accuracy to that of satellite-estimated PM_{2.5} concentrations. Table 5 summarizes several
496 representative studies focusing on the estimation of the ground-based PM_{2.5} concentration in China at the national level using
497 different kinds of methods. Most of these studies estimated the ground-based PM_{2.5} concentration at the daily scale since they
498 employed polar-orbiting satellite data (e.g., MODIS) that only provide daily AOD observations. The estimation conducted by
499 Liu et al. (2019) was an exception which exhibited an hourly resolution due to the use of AOD measurements from a
500 geostationary satellite (Himawari-8). The horizontal resolution in these studies was mainly approximately 10 km except that
501 of Lin et al. (2018), which revealed the finest horizontal resolution (1 km), and that of Zhan et al., 2017, which revealed the
502 coarsest horizontal resolution (0.5°). Few studies have provided long-term PM_{2.5} data covering recent years. In comparison,
503 our PM_{2.5} reanalysis data provide long-term data in China at a fine temporal resolution (1 hour) and a high accuracy. A fine
504 temporal resolution is important for epidemiological studies, especially for the assessment of the acute health effects of air
505 pollution. Furthermore, the accuracy of our reanalysis data (CV $R^2 = 0.86$ and CV RMSE = 15.1 $\mu\text{g} \cdot \text{m}^{-3}$) was also higher
506 than that of most of these satellite estimates (CV $R^2 = 0.56\text{--}0.86$ and CV RMSE = 15.0–20.2 $\mu\text{g} \cdot \text{m}^{-3}$).

507 4.2 Gases

508 4.2.1 Spatial distribution of the reanalysis data of gaseous air pollutants over China

509 Next, we present the reanalysis fields for gaseous air pollutants in China, namely, SO₂, CO, NO₂ and O₃. Figure 9 shows
510 the spatial distribution of the six-year average SO₂ and CO concentrations in China obtained from the CAQRA dataset, base
511 simulation and observations. The SO₂ reanalysis data captured the magnitude and spatial distribution of the SO₂ concentration
512 in China well, while the base simulation greatly overestimated the SO₂ concentration due to the positive biases of the SO₂
513 emissions in the simulations. Consistent with the observations, the SO₂ reanalysis data exhibited high spatial heterogeneity,
514 with the highest values located in the NCP region, especially in Shandong, Shanxi and Hebei provinces. Several SO₂
515 concentration hotspots were also found in the NE region. SO₂ is mainly emitted from fossil fuel consumption, especially coal
516 burning (Lu et al., 2010). Shandong, Shanxi, Inner Mongolia and Hebei provinces are the four largest consumers of coal in
517 China according to the China Energy Statistical Yearbook (NBSC 2017a, b), which explains the high SO₂ concentrations in
518 these provinces. The spatial distribution of the CO reanalysis data was similar to that of the SO₂ reanalysis data and agreed
519 well with the observed spatial distribution. In contrast, the base simulation highly underestimated the CO concentration,
520 especially in the NCP region. In addition, both the observations and reanalysis data showed CO concentration hotspots in the
521 NW region and Xizang Province, while these hotspots were largely underestimated or even missing in the base simulation.
522 According to previous studies, such underestimation might be related to underestimated CO emissions in China (Kong et al.,
523 2020; Tang et al., 2013). In regard to NO₂ (Fig. 10), both the reanalysis data and base simulation captured the observed
524 magnitude and spatial distribution of the NO₂ concentration in China. High NO₂ concentrations generally occurred in the NCP
525 region and the major city clusters in China. However, the base simulation generally revealed an underestimated NO₂
526 concentration in China. The spatial distribution of the O₃ concentration (Fig. 10) demonstrated a lower spatial heterogeneity
527 than that of the other gases. The O₃ reanalysis data suitably captured the observed magnitude and spatial distribution of the O₃
528 concentration in China, while the base simulation generally underestimated the O₃ concentration in China. Figures D3–6 in
529 Appendix D further show seasonal maps of the reanalysis fields of these gases. All gases exhibited a profound seasonal cycle,
530 with maximum values observed in winter and the lowest values in summer except O₃, which demonstrated the opposite
531 seasonal cycle. The highest SO₂, CO and NO₂ concentrations in winter could occur due to the increased anthropogenic
532 emissions and the more stable atmospheric conditions during this season. Regarding O₃, the highest value in summer was
533 closely related to the enhanced photochemical reactions in summer associated with the high temperature and solar radiance.

534 4.2.2 Assessment of the gas reanalysis data over China

535 Evaluation results of the above gas reanalysis data are provided in Table 2. The table indicates that the reanalysis data
536 attain an excellent performance in representing the magnitude and variability of these gaseous air pollutants in China, with CV
537 R² values ranging from 0.51 for SO₂ to 0.76 for O₃ and CV MBE (CV NMB) values of approximately -2.0 μg · m⁻³ (-8.5%),
538 -2.3 μg · m⁻³ (-6.9%), -0.06 mg · m⁻³ (-6.1%) and -2.3 μg · m⁻³ (-4.0%) for the hourly SO₂, NO₂, CO and O₃ reanalysis data,

539 respectively. Compared to the base simulation, the errors were reduced by approximately half in the reanalysis data with CV
540 RMSE values of approximately $24.9 \mu\text{g} \cdot \text{m}^{-3}$, $16.4 \mu\text{g} \cdot \text{m}^{-3}$, $0.54 \text{mg} \cdot \text{m}^{-3}$ and $21.9 \mu\text{g} \cdot \text{m}^{-3}$ for the hourly SO_2 , NO_2 , CO
541 and O_3 reanalysis data, respectively. The reanalysis data achieved a good performance at the daily, monthly and yearly scales.
542 The CV RMSE values of the daily SO_2 and NO_2 reanalysis data were also smaller than those of the SO_2 and NO_2 concentration
543 datasets in China previously developed by Zhan et al. (2018) and Zhang et al. (2019), respectively, based on the random-forest-
544 spatiotemporal-kriging model wherein the RMSE values of the daily SO_2 and NO_2 concentrations were estimated to be 19.5
545 and $13.3 \mu\text{g} \cdot \text{m}^{-3}$, respectively.

546 In terms of the different regions (Tables E3–6, Appendix E), the hourly SO_2 reanalysis data indicated small negative
547 biases (approximately 2–10%) in all regions except in the central region, where the negative bias was relatively large (17.0%).
548 The smallest CV RMSE values of the SO_2 reanalysis data were observed in the SE, SW and NW regions (smaller than $25 \mu\text{g} \cdot \text{m}^{-3}$)
549 m^{-3}), while in the other regions, the CV RMSE values exceeded $30 \mu\text{g} \cdot \text{m}^{-3}$. The hourly NO_2 reanalysis data showed small
550 negative biases in all regions, which were relatively small in the NE, NCP and SE regions (ranging from -5.9 to -3.5%) and
551 were relatively large in the SW, NW and central regions (ranging from -15.1 to -12.9%). The CV RMSE for the hourly NO_2
552 reanalysis data was approximately $15 \mu\text{g} \cdot \text{m}^{-3}$ in all regions except in the NW ($24.3 \mu\text{g} \cdot \text{m}^{-3}$) and central ($20.5 \mu\text{g} \cdot \text{m}^{-3}$)
553 regions. The hourly CO reanalysis data exhibited small negative biases in all regions. The largest biases were still found in the
554 NW region, which reached approximately 15.0%, while in the other regions, the biases ranged from -11.2% to -2.5%. The CV
555 RMSE values for the hourly CO reanalysis data were the smallest in South China (approximately 0.39 and $0.46 \text{mg} \cdot \text{m}^{-3}$ in
556 the SE and SW regions, respectively) and increased to 0.64 and $0.59 \text{mg} \cdot \text{m}^{-3}$ in the NCP and NE regions, respectively. The
557 largest CV RMSE was observed in the NW region, which amounted to approximately $1.13 \text{mg} \cdot \text{m}^{-3}$. The biases of the hourly
558 O_3 reanalysis data were uniformly distributed in the different regions, with the CV NMB value ranging from -6.1% to 1.4%.
559 Similarly, the CV RMSE value of the O_3 reanalysis data was approximately $20 \mu\text{g} \cdot \text{m}^{-3}$ in all regions except in the NW region
560 ($28.3 \mu\text{g} \cdot \text{m}^{-3}$).

561 4.2.3 Trend study of the gas reanalysis data over China

562 Figure 11 shows time series of the monthly mean SO_2 concentration in China obtained from the CV run, base simulation
563 and observations. Additionally, time series of the monthly mean SO_2 concentration in the different regions are shown. The
564 observed SO_2 concentrations showed significant negative trends ($P < 0.05$) in China ($-6.2 \mu\text{g} \cdot \text{m}^{-3} \cdot \text{yr}^{-1}$, Table 6) and in all
565 regions (ranging from -2.3 to $-9.5 \mu\text{g} \cdot \text{m}^{-3} \cdot \text{yr}^{-1}$, Table 6) due to the large reductions in SO_2 emissions across China. During
566 the 11th-13rd Five-Year Plans (FYPs) and the Air Pollution Prevention and Control Plan, the Chinese government invested
567 great efforts to reduce SO_2 emissions, such as the installation of flue-gas desulfurization (FGD) and selective catalytic
568 reduction systems, construction of large units, decommissioning of small units and replacement of coal with cleaner energies
569 (Li et al., 2017; Zheng et al., 2018b). As a result, the SO_2 emissions substantially decreased in China, especially in the industrial
570 and power sectors. The base simulation significantly overestimated the SO_2 concentration in all regions, especially after 2013.

571 The negative trends of the SO₂ concentration were also largely underestimated in the base simulation. In contrast, the SO₂
572 reanalysis data captured the magnitude and negative trends of the observed SO₂ concentrations in China and in all regions well.
573 The NO₂ observations showed negative trends in China as well (Fig. 12). However, the negative trend was not significant
574 except in the NE region (Table 6). This is consistent with the small reductions in NO_x emissions (21%) in China due to the
575 small changes in the emissions originating from the transportation sector, accounting for almost one-third of the NO_x emissions
576 in China. The pollution controls applied in the transportation section were exactly offset by the growing emissions related to
577 vehicle growth (Zheng et al., 2018b). The base simulation generally underestimated the NO₂ concentration during the
578 wintertime, and the observed negative trends of the NO₂ concentration were also underestimated in all regions. By assimilating
579 the observed NO₂ concentrations, the reanalysis data agreed better with the observations both in regard to the magnitude and
580 negative trends. The CO observations exhibited significant negative trends in all regions except in the NW region (Fig. 13),
581 with calculated negative trends ranging from -0.18 to -0.06 μg · m⁻³ · yr⁻¹. Such negative trends have also been observed in
582 satellite measurements, such as MOPITT observations (Zheng et al., 2018a), which are mainly attributed to the reduced
583 anthropogenic emissions in China, as suggested by both bottom-up and top-down methods (Zheng et al., 2019). The base
584 simulation largely underestimated the CO concentration in all regions. In addition, the negative trends of the CO concentration
585 were also notably underestimated in the base simulation, which highlights the major contribution of emission reduction to the
586 decreased CO concentration in these regions. The CO reanalysis data agreed well with the observations and captured the
587 negative trends of the CO concentration in all regions. The O₃ concentration exhibited the opposite trend to that exhibited by
588 the other air pollutants (Fig. 14), which revealed significant positive trends in all regions, ranging from 2.3 to 5.4 μg · m⁻³ ·
589 yr⁻¹ and indicating enhanced photochemical pollution in China. This phenomenon has been observed and investigated by Li
590 et al. (2019), who suggested that the rapid decrease in the PM_{2.5} concentration and the resultant reduction in the aerosol sink
591 of hydroperoxyl (HO₂) radicals were important factors contributing to the enhanced O₃ concentration in China. The base
592 simulation generally captured the magnitude of the O₃ concentration in the SE, SW, NW and central regions but underestimated
593 the O₃ concentration in the NCP and NE regions, especially in spring and summer. In addition, the base simulation
594 underestimated the observed positive trends of the O₃ concentration in all regions, which suggests that meteorological
595 variability only contributed a small proportion of the observed O₃ trend in China. Again, the O₃ reanalysis data are substantially
596 better than the base simulation and suitably reproduce the observed trends of the O₃ concentration in all regions.

597 **4.2.4 Comparison to the CAMS reanalysis data**

598 To further evaluate the accuracy of our reanalysis dataset for gaseous air pollutants, the CAMSRA dataset produced by
599 the ECMWF (Inness et al., 2019) was employed as a reference in a comparison to our reanalysis dataset. The CAMSRA dataset
600 is the latest global reanalysis dataset of the atmospheric composition, which assimilates satellite retrievals of O₃, CO, NO₂ and
601 AOD. Three-hour reanalysis data of the SO₂, NO₂, CO and O₃ concentrations at the surface model level from 2013 to 2018
602 were adopted in this study, which were downloaded from [https://atmosphere.copernicus.eu/copernicus-releases-new-global-](https://atmosphere.copernicus.eu/copernicus-releases-new-global-reanalysis-data-set-atmospheric-composition)
603 [reanalysis-data-set-atmospheric-composition](https://atmosphere.copernicus.eu/copernicus-releases-new-global-reanalysis-data-set-atmospheric-composition) (last accessed: 17 April 2020) at a resolution of 1 degree by 1 degree. Here, we

604 only focus on a comparison of the gaseous pollutants since the CAMSRA dataset does not provide PM_{2.5} and PM₁₀
605 concentrations.

606 Figure 15 shows the spatial distribution of the six-year average concentration of these gaseous air pollutants in China
607 obtained from the CAMSRA dataset. Compared to the spatial distributions determined with the CAQRA dataset and
608 observations (Figs. 9–10), the CAMSRA dataset greatly overestimates the surface SO₂ and O₃ concentrations in China. In
609 addition, due to the higher spatial resolution (15 km) of the CAQRA dataset than that of the CAMSRA dataset (approximately
610 50 km), our products provide more detailed spatial patterns of the surface air pollutants in China, which are better suited for
611 air quality studies at the regional scale. Table 7 quantitatively compares the accuracy of the CAQRA dataset to that of the
612 CAMSRA dataset in the estimation of the surface concentrations of gaseous air pollutants in China. Compared to CAMSRA
613 ($R^2 = 0.00\text{--}0.23$), CAQRA attains a better performance in capturing the spatiotemporal variability in the surface concentrations
614 of gaseous air pollutants in China, with R^2 values ranging from 0.53 to 0.77. The MBE and RMSE values are also smaller in
615 the CAQRA dataset than those in the CAMSRA dataset, especially for the SO₂ and O₃ concentrations. This is attributed to the
616 assimilation of surface observations in CAQRA, while CAMSRA only assimilates satellite retrievals. These results suggest
617 that the CAQRA dataset provides surface air quality datasets in China of a higher quality than the air quality datasets provided
618 by the CAMSRA dataset, which is especially valuable for future relevant studies with high demands in spatiotemporal
619 resolution and accuracy.

620 **5 Data availability**

621 The CAQRA dataset can be freely downloaded at <https://doi.org/10.11922/sciencedb.00053> (Tang et al., 2020a), and the
622 prototype product, which contains the monthly and annual means of the CAQRA dataset, is available at
623 <https://doi.org/10.11922/sciencedb.00092> (Tang et al., 2020b). When you click the first Science DB link, you will see basic
624 descriptions of CAQRA dataset, and 2192 zip files listed in the DATA FILES column at the website. The total file sizes are
625 approximately 318.81 GB as of the time of this writing. Each zip file is named by the date and contains one day's reanalysis
626 data, which is composed of 24 Network Common Data Form (NetCDF) files. Each NetCDF file contains one hour's reanalysis
627 data and is named by the date. The time zone of the reanalysis data is Beijing Time and the description on the content of each
628 NetCDF file is available in README.txt at the website. The monthly and annual version of the CAQRA dataset contains two
629 zip files which are the monthly and annual mean of the reanalysis data, respectively. The total file sizes of this product are
630 approximately 480.67MB which is more easily to be downloaded and suitable for users who only need air quality data at
631 monthly or yearly scales.

632

633 6 Conclusions

634 A high-resolution CAQRA dataset was produced in this study by assimilating surface observations of the PM_{2.5}, PM₁₀,
635 SO₂, NO₂, CO and O₃ concentrations retrieved from the CNEMC. This dataset provides time-consistent concentration fields
636 of PM_{2.5}, PM₁₀, SO₂, NO₂, CO and O₃ in China from 2013 to 2018 (will be extended in the future on a yearly basis) at high
637 spatial (15 km) and temporal (1 hour) resolutions. The CAQRA dataset was produced with the ChemDAS, which applied the
638 NAQPMS model as the forecast model, and the LETKF to assimilate the observations in the postprocessing mode. The
639 background error covariance was calculated from ensemble simulations, which considered the emission uncertainties of the
640 major air pollutants. An inflation technique was also applied to dynamically inflate the background error to prevent
641 underestimation of the true background error covariance.

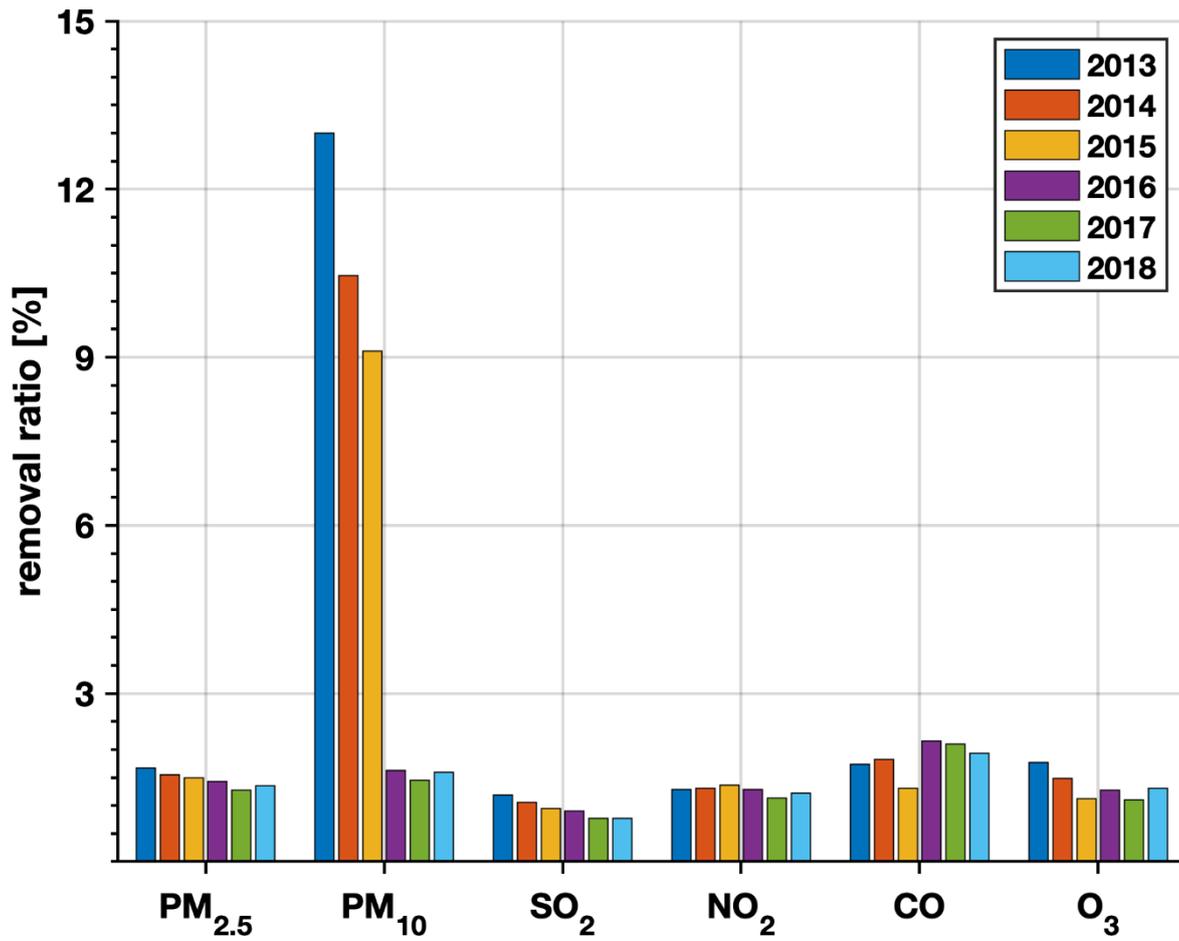
642 The fivefold CV method was employed to validate the reanalysis dataset, which provided us with the first indication of
643 the quality of the CAQRA dataset. The validation results suggested that the CAQRA dataset attains an excellent performance
644 in representing the spatiotemporal variability of surface air pollutants in China, with CV R² values ranging from 0.52 for the
645 hourly SO₂ concentration to 0.81 for the hourly PM_{2.5} concentration. The CV MBE values of the reanalysis data were -2.6 μg ·
646 m⁻³, -6.8 μg · m⁻³, -2.0 μg · m⁻³, -2.3 μg · m⁻³, -0.06 mg · m⁻³ and -2.3 μg · m⁻³ for the hourly concentrations of PM_{2.5},
647 PM₁₀, SO₂, NO₂, CO and O₃, respectively. The CV RMSE values of the reanalysis data for these air pollutants were estimated
648 to be approximately 21.3 μg · m⁻³, 39.3 μg · m⁻³, 24.9 μg · m⁻³, 16.4 μg · m⁻³, 0.54 mg · m⁻³ and 21.9 μg · m⁻³,
649 respectively. In the different regions of China, the NW and central regions exhibited relatively large biases and errors, which
650 mainly occurred due to the relatively sparse observations and underestimated background errors. The Chinese air quality has
651 substantially changed over the last six years. The observations indicate significant decreasing trends for all air pollutants except
652 O₃, which shows an increasing trend over the last six years. The reanalysis data reveal an excellent performance in representing
653 the trends of all air pollutants in China, suggesting the suitability of the reanalysis data for air pollutant trend analysis in China.

654 In addition to the CV method, the PM_{2.5} reanalysis data were also evaluated against independent observations retrieved
655 from the U.S. Department State Air Quality Monitoring Program over China. The results suggested that the reanalysis data
656 suitably reproduce the magnitude and variability of the observed PM_{2.5} concentration in all cities, with the MBE and RMSE
657 values only ranging from -7.1 to -0.3 μg · m⁻³ and from 16.8 to 33.6 μg · m⁻³, respectively. The reanalysis data of the gaseous
658 air pollutants were also compared to the latest global reanalysis data contained in the CAMSRA dataset produced by the
659 ECMWF. The CAMSRA dataset is of great value in providing three-dimensional distributions of multiple chemical species
660 globally. As a regional dataset, our products attain a higher spatial resolution than does the CAMSRA dataset, which could
661 better suit air quality studies at the regional scale. Although our products only provide the surface concentrations of six
662 conventional air pollutants in China, the accuracy of the CAQRA dataset was estimated to be higher than that of the CAMSRA
663 dataset due to the assimilation of surface observations. Hence, our products exhibit their own value in regional air quality
664 studies with high demands in spatiotemporal resolution and accuracy. We also compared our PM_{2.5} reanalysis data to previous

665 satellite estimates of the surface PM_{2.5} concentration, which revealed that the PM_{2.5} reanalysis data are more accurate than
666 most satellite estimates and exhibit a relatively fine temporal resolution.

667 As the first version of the CAQRA dataset, certain limitations remain that potential users should be aware of. First, the
668 discontinuities in the availability and coverage of assimilated observations will affect the reanalysis quality and the estimated
669 interannual trends. As shown in Sect.3.1, there has been a consistent increase in the number of assimilated observations from
670 2013 to 2015 due to the increases of observation sites. The smaller number of assimilated observations in 2013 and 2014 would
671 provide less constrains on the background state and thus degrade the reanalysis in these two years. This may cause spurious
672 interannual changes and trends from 2013 to 2018. Thus, cautions are needed when using the reanalysis for long-term air
673 quality change from 2013 to 2018. However, this problem would be not serious after 2015 when the number of assimilated
674 observations become stable. In addition, the observation sites used in the assimilation are mainly urban or suburban sites that
675 do not provide enough information on the air pollution in rural areas, which may influence the quality of CAQRA in rural
676 areas. Secondly, we only perturbed the emissions to represent the forecast uncertainty in this study, which may underestimate
677 the forecast uncertainty due to the omitting of other error sources, such as the uncertainty in poorly parameterized physical or
678 chemical processes, and the uncertainty in meteorological simulation. The limited ensemble size would also lead to
679 underestimation of the forecast error especially in the high-resolution assimilation applications. Although the inflation method
680 is used to compensate for the missing errors, the underestimated forecast uncertainty would still degrade the assimilation
681 performance to a certain extent as exemplified by the larger biases in the reanalysis over NW and Central regions. Thirdly, we
682 did not consider the annual trend of emissions in the ensemble simulation. This would lead to temporal changes in the statistics
683 of innovation due to the substantial changes of observations, which would influence the long stability of the data assimilation
684 as suggested by the χ^2 test although the OmA statistics generally confirms a passable stability in our assimilation system. Last
685 but not least, the current CAQRA only contains the surface concentrations of the air pollutants in China which cannot provide
686 the information on the vertical structure of the air pollutants. to further improve the accuracy of our air quality reanalysis
687 dataset, in the future, an online EnKF run could be conducted to simultaneously correct the emissions and concentrations.
688 More observation types, such as observation data of the PM_{2.5} composition, could also be assimilated to provide PM_{2.5}
689 composition fields in China, which could support both epidemiological studies and climate research.

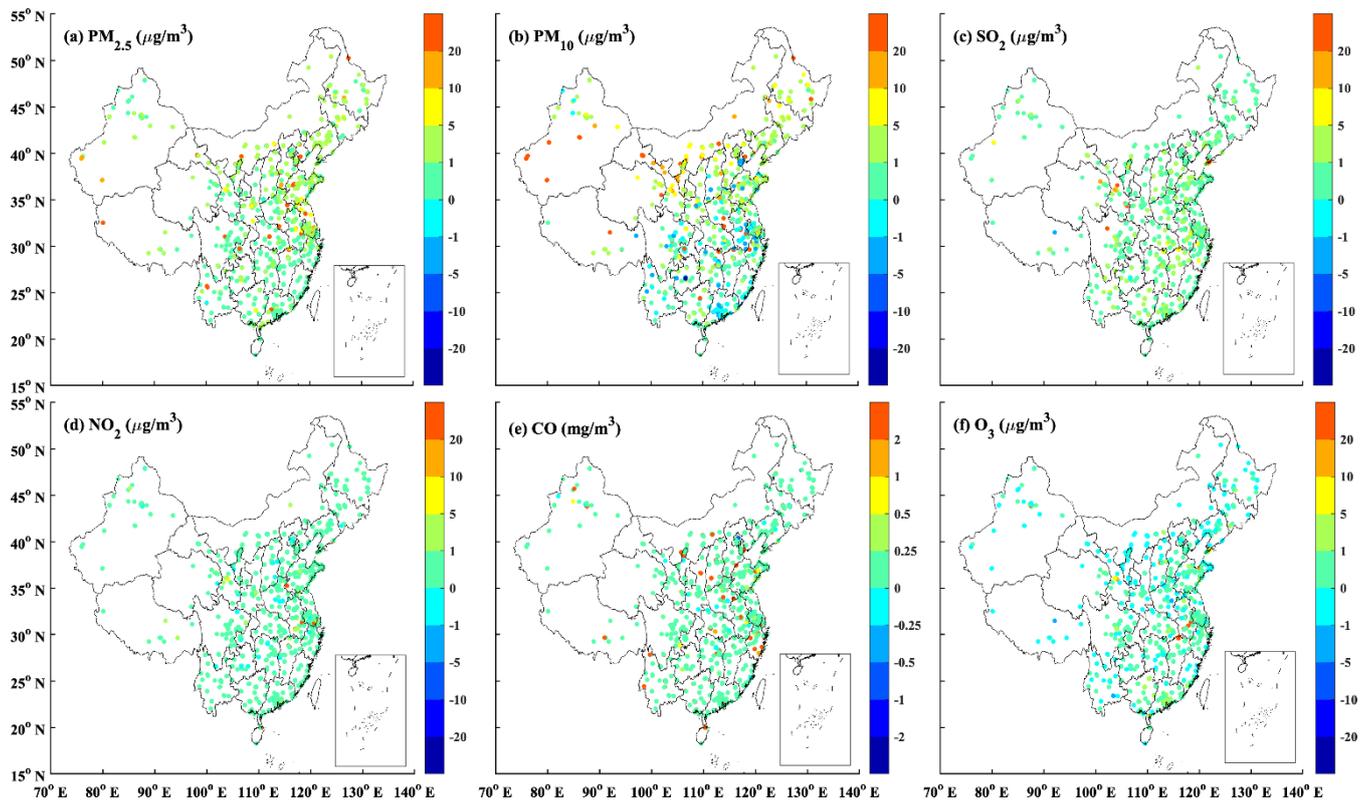
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701 Figure A1: Removal ratio of the observations in China from 2013 to 2018 for different species detected by the automatic
 702 outlier detection method.

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705 **Figure A2: Spatial distributions of differences in annual concentrations of six air pollutants in China before and after**
 706 **quality control averaged from 2013 to 2018.**

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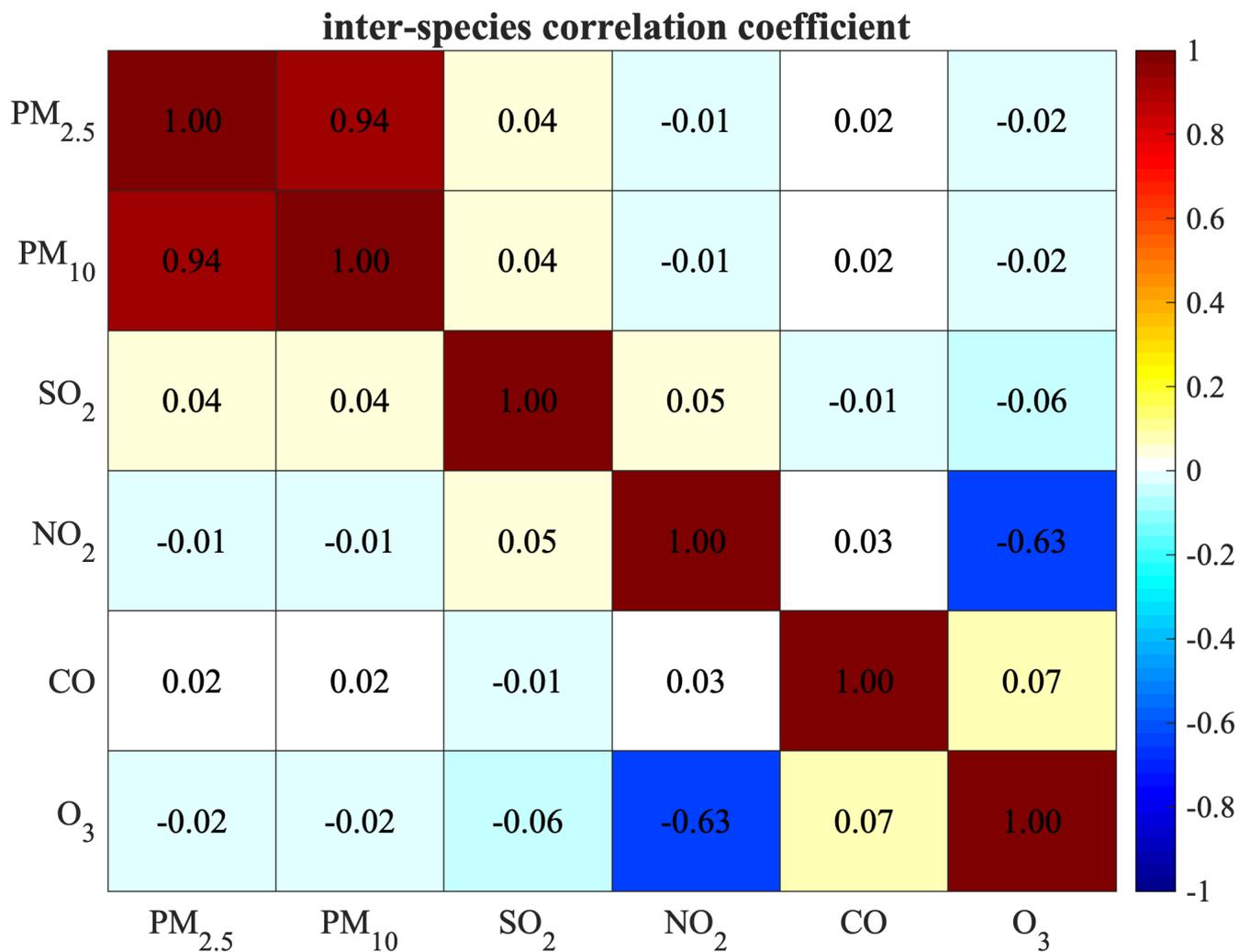
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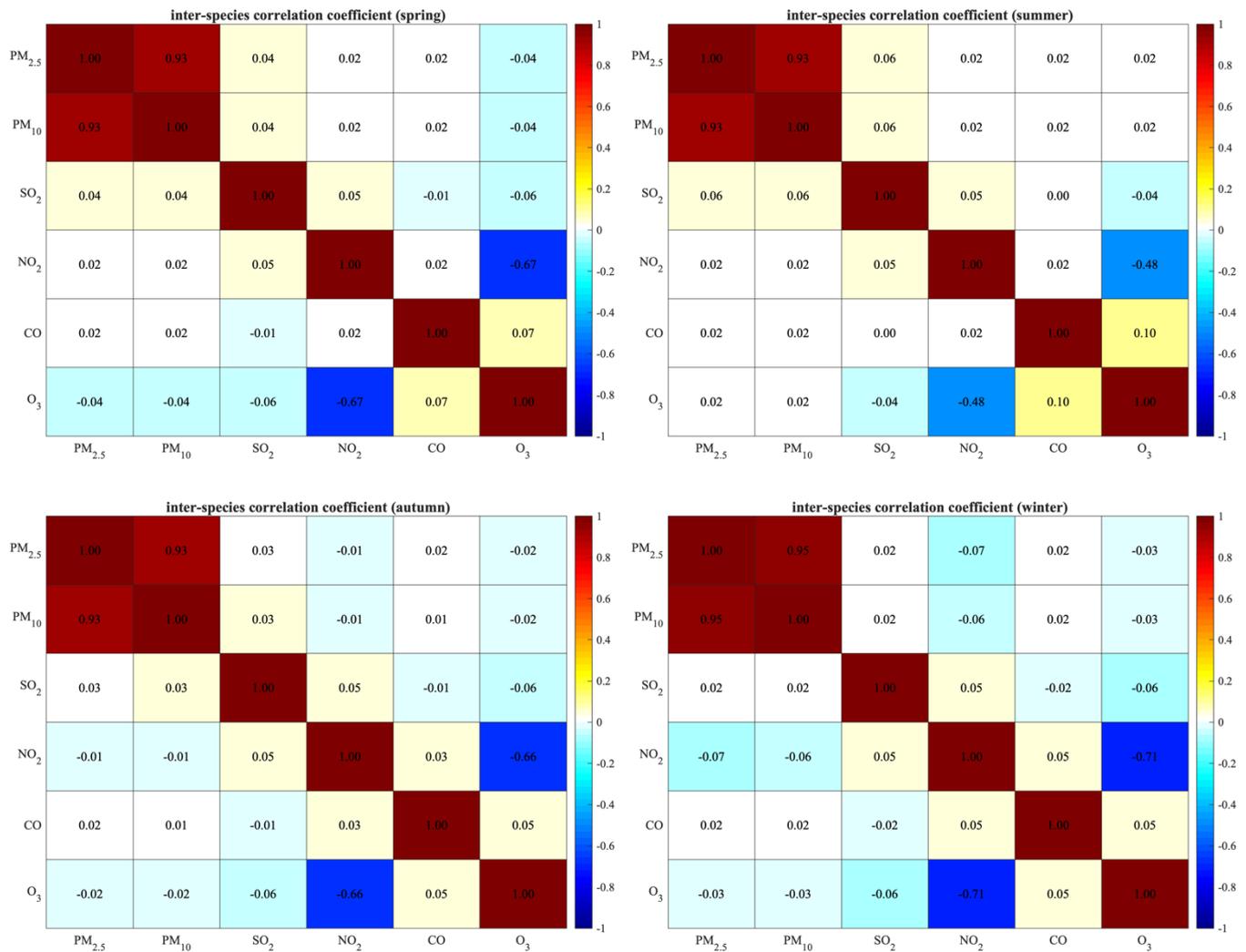
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723 **Figure B1: Correlations between species in the background error covariance matrix, estimated from the LETKF**
 724 **ensemble averaged from 2013 to 2018. The global mean of the covariance estimated for each station is plotted.**

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727 **Figure B2: Correlations between species in the background error covariance matrix, estimated from the LETKF**
 728 **ensemble averaged in different seasons from 2013 to 2018. The global mean of the covariance estimated for each station**
 729 **is plotted.**

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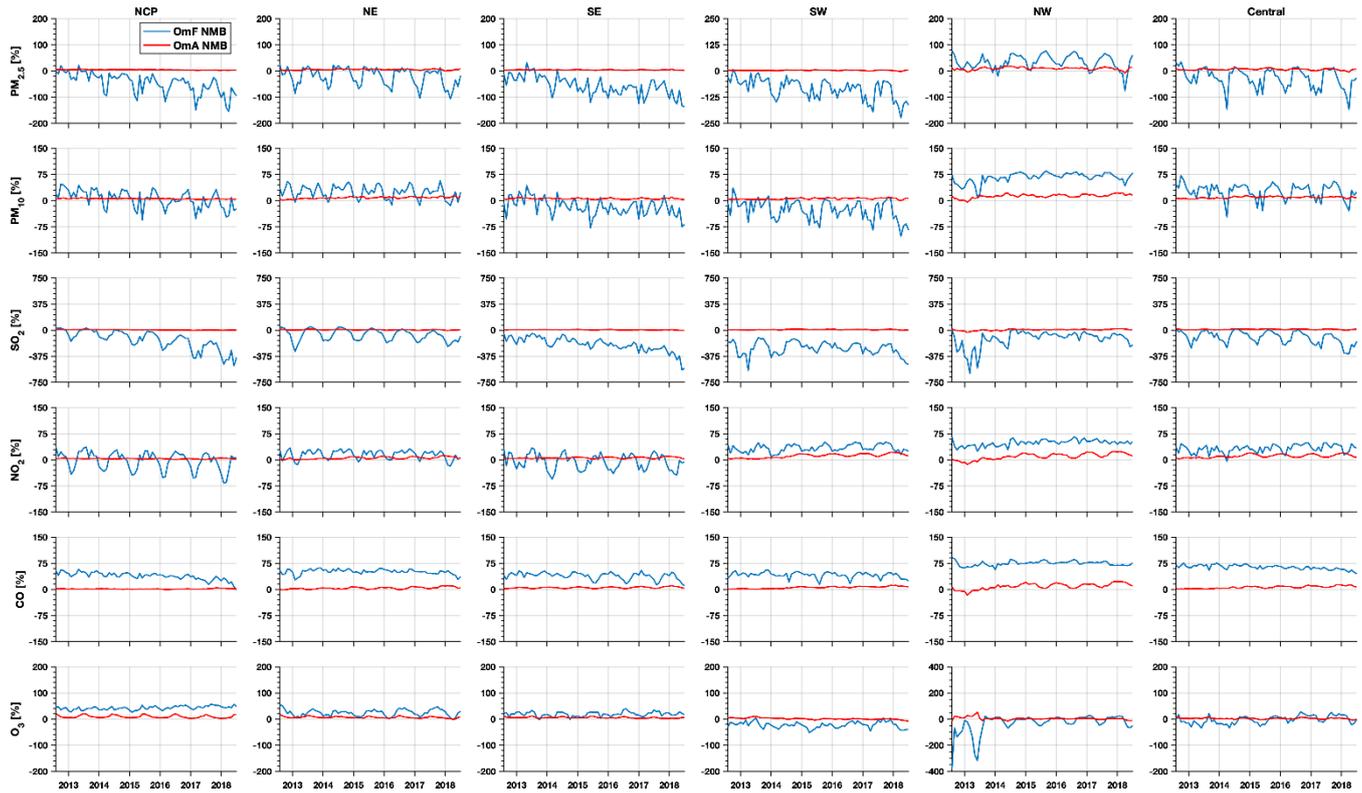
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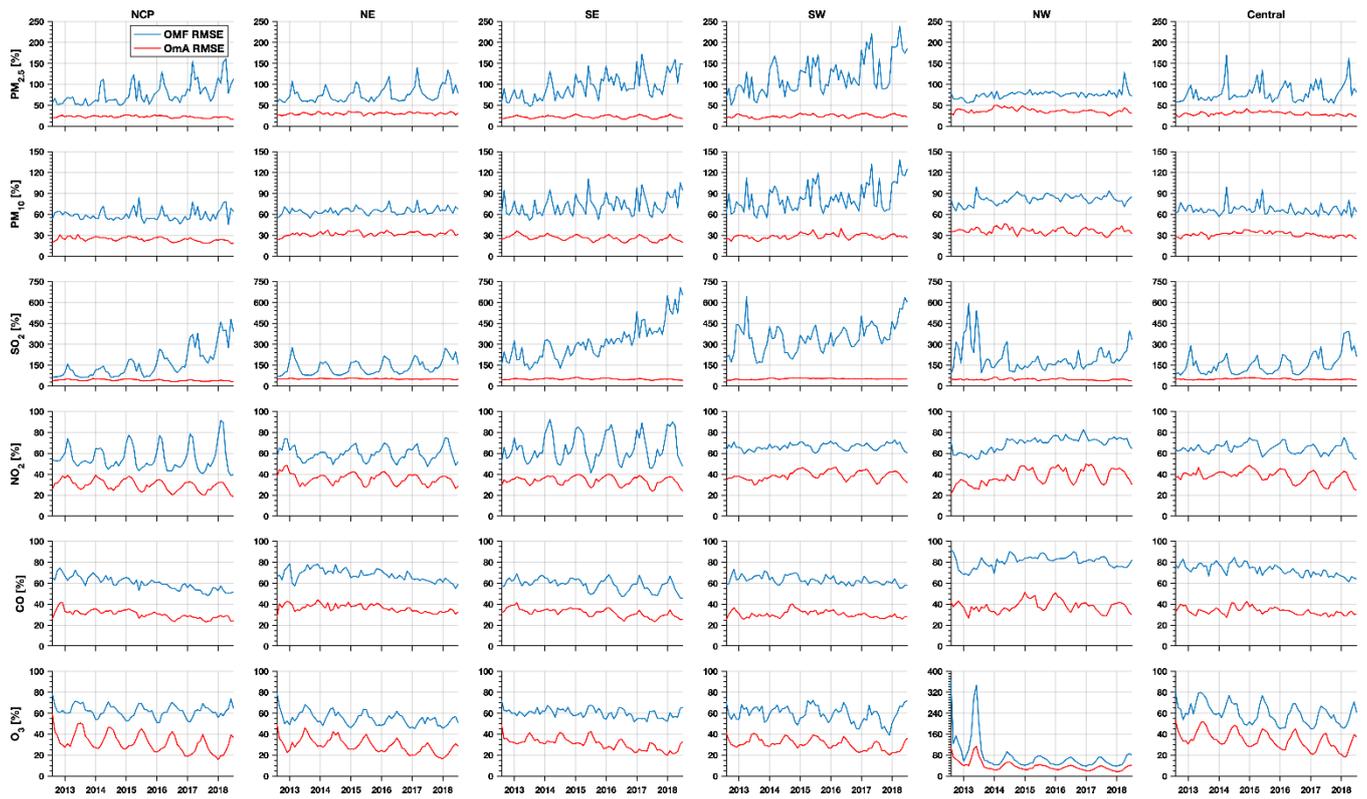
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740 Figure C1: Time series of monthly mean OmF and OmA normalized mean bias in different regions of China for
 741 different species.

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744 **Figure C2: Time series of monthly mean OmF and OmA normalized root mean square error in different regions of**
 745 **China for different species.**

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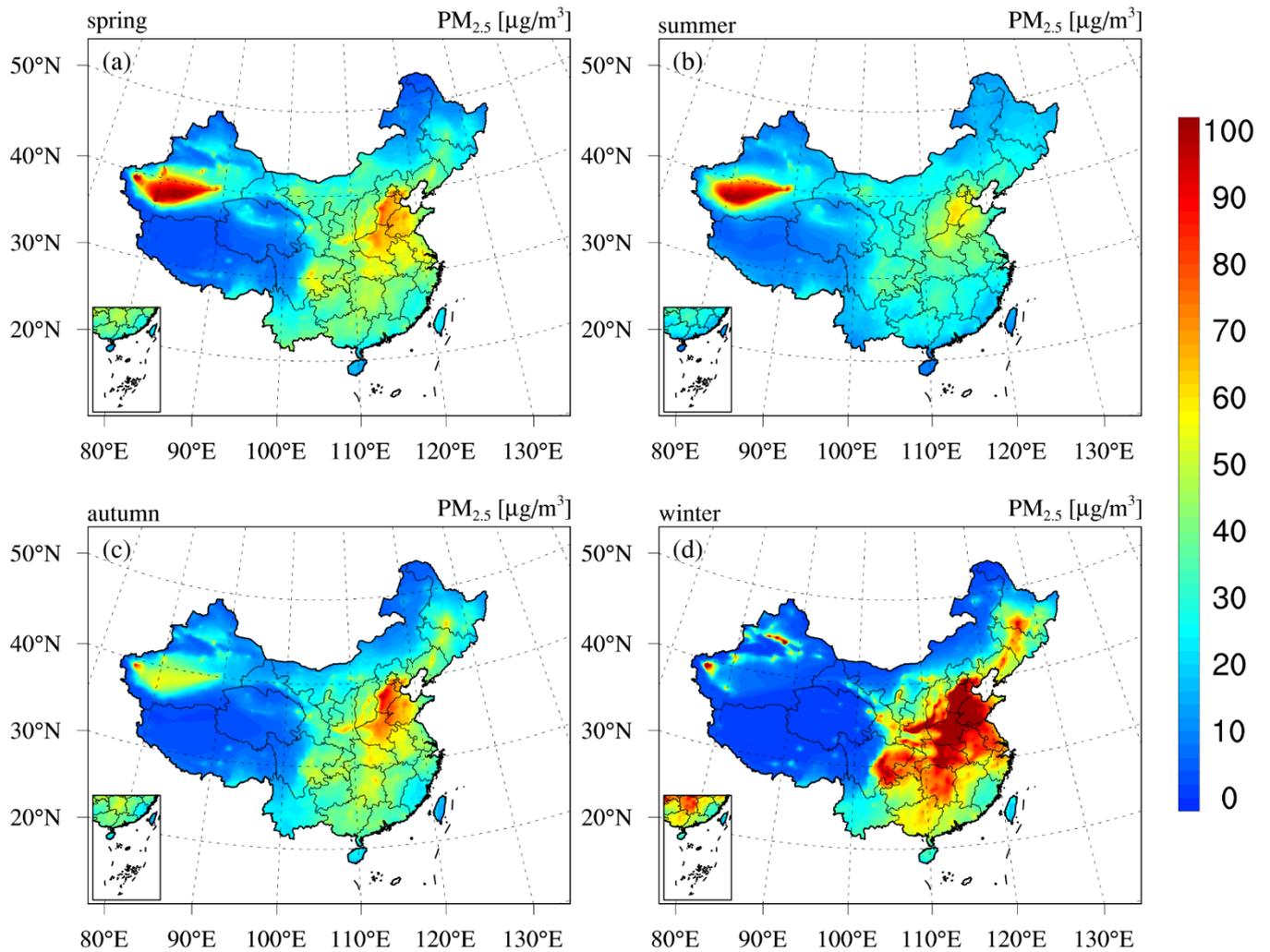
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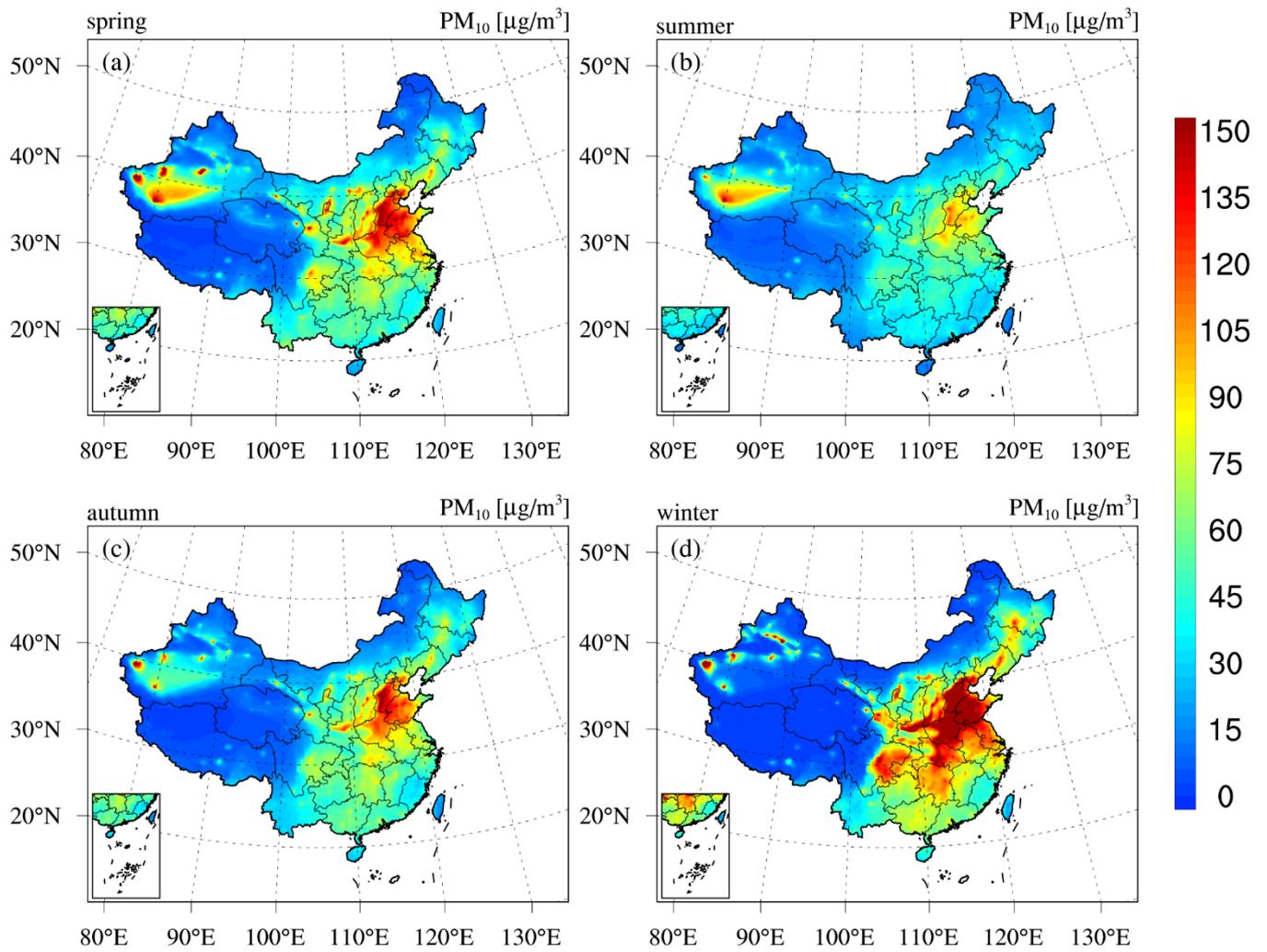
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762 Figure D1: Spatial distributions of the PM_{2.5} reanalysis in China during (a) spring, (b) summer, (c) autumn and (d)

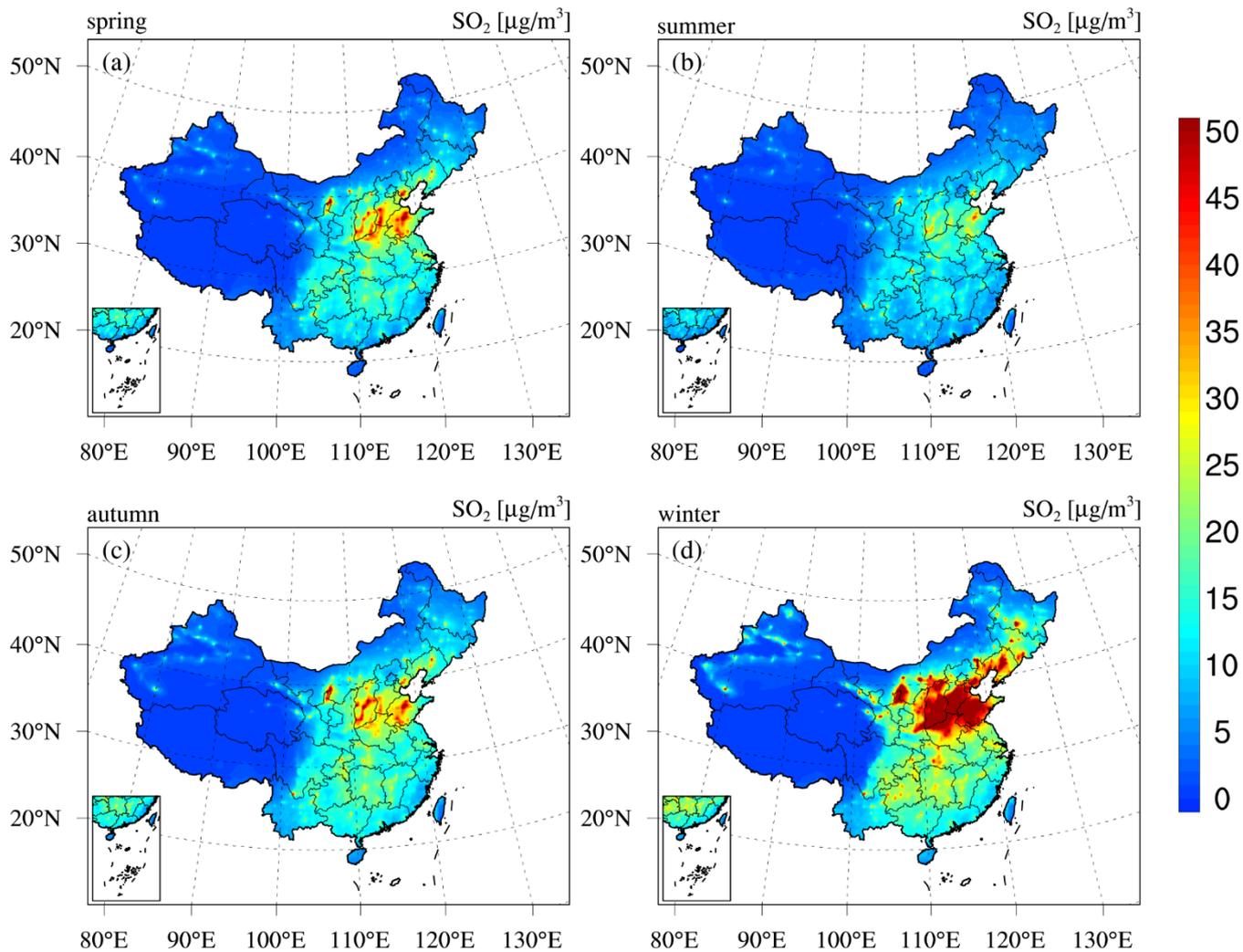
763 winter averaged from 2013 to 2018.



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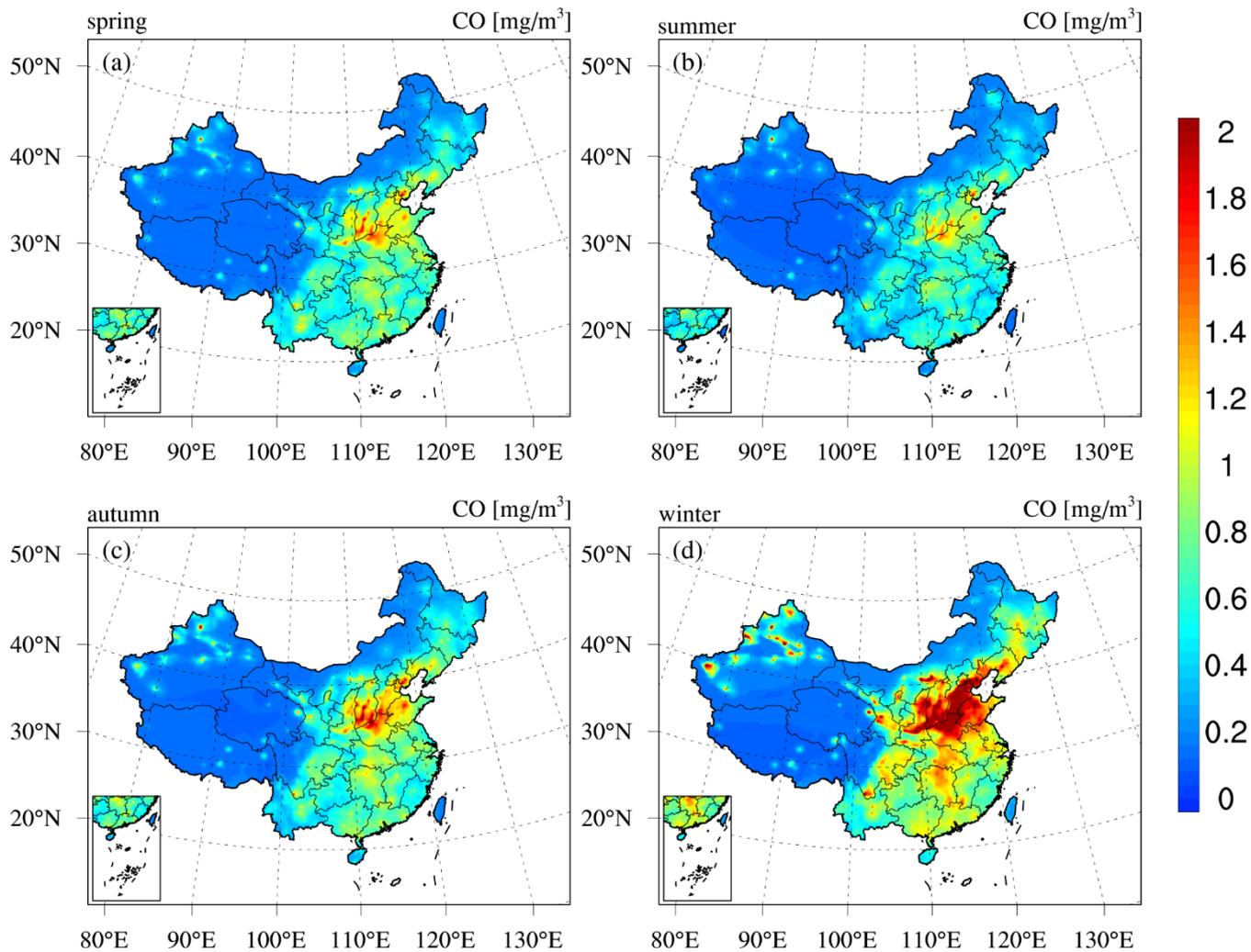
765 **Figure D2: Same as Fig. D1 but for PM₁₀.**

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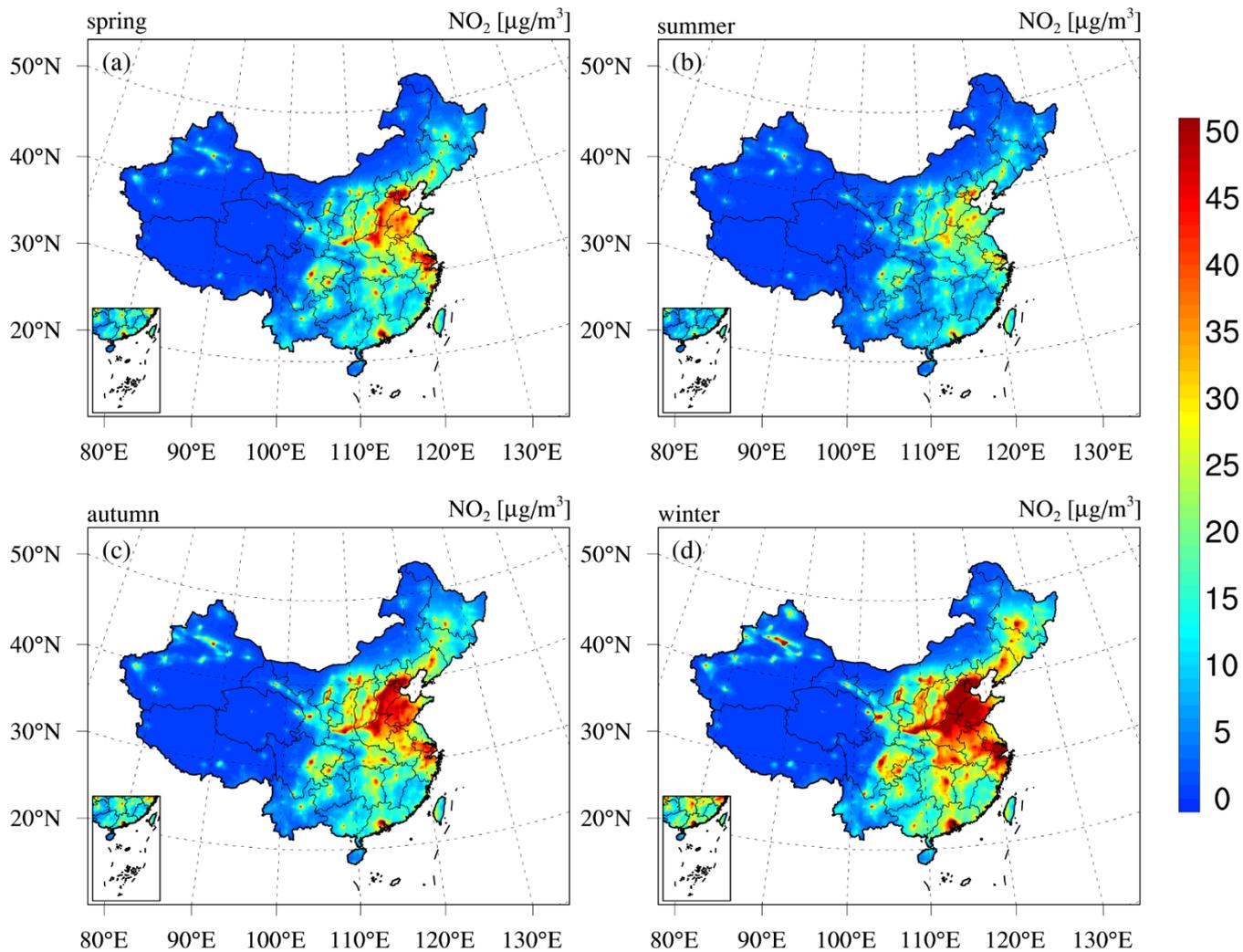
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768 **Figure D3: Same as Fig. D1 but for SO₂.**



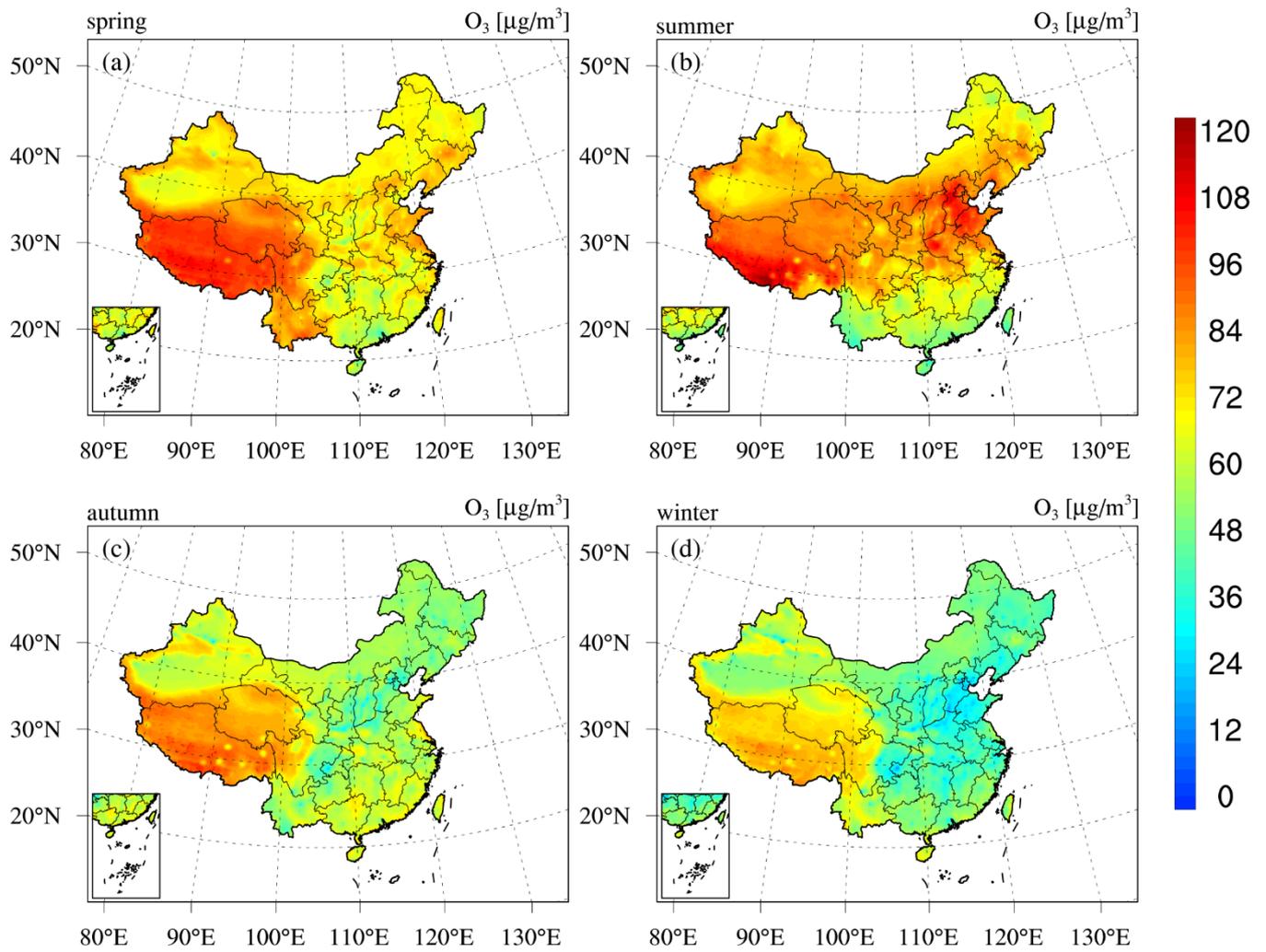
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770 **Figure D4: Same as Fig. D1 but for CO.**



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772 **Figure D5: Same as Fig. D1 but for NO₂.**



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774 **Figure D6: Same as Fig. D1 but for O₃.**

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785 **Appendix E: CV results of the reanalysis data in different regions of China**786 **Table E1: CV results of the reanalysis (outside bracket) and base simulation (in bracket) for PM_{2.5} concentrations in**
787 **different regions of China at different temporal scales**

PM _{2.5} (µg/m ³)	NCP				NE			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.85 (0.33)	-3.3 (22.4)	-4.8 (32.8)	25.1 (62.6)	0.77 (0.25)	-2.6 (2.8)	-5.8 (6.5)	22.6 (44.5)
Daily	0.90 (0.44)	-3.4 (22.3)	-4.9 (32.4)	17.5 (51.2)	0.86 (0.32)	-2.6 (2.6)	-5.9 (6.0)	14.7 (35.1)
Monthly	0.92 (0.56)	-3.4 (22.2)	-4.9 (32.4)	11.4 (34.1)	0.86 (0.38)	-2.6 (2.7)	-5.9 (6.0)	9.7 (21.4)
Yearly	0.92 (0.56)	-3.6 (20.8)	-5.0 (29.2)	8.7 (27.3)	0.79 (0.35)	-3.1 (0.4)	-6.6 (0.8)	8.8 (16.7)
	SE				SW			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.85 (0.25)	-1.8 (22.2)	-3.8 (47.6)	14.9 (51.5)	0.79 (0.22)	-1.4 (30.3)	-3.4 (74.7)	16.5 (57.4)
Daily	0.90 (0.31)	-1.8 (22.2)	-3.8 (47.4)	10.6 (45.4)	0.86 (0.29)	-1.4 (30.0)	-3.4 (74.2)	12.1 (51.6)
Monthly	0.92 (0.45)	-1.8 (22.1)	-3.8 (47.2)	7.4 (33.7)	0.86 (0.49)	-1.5 (29.8)	-3.7 (73.3)	9.7 (42.8)
Yearly	0.90 (0.37)	-2.0 (20.5)	-4.0 (42.0)	6.1 (29.3)	0.79 (0.47)	-2.2 (27.2)	-5.0 (63.2)	9.5 (38.8)
	NW				Central			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.52 (0.11)	-7.3 (-28.7)	-13.1 (-51.1)	52.1 (73.0)	0.72 (0.23)	-4.1 (0.8)	-8.2 (1.6)	26.6 (47.5)
Daily	0.66 (0.15)	-7.5 (-29.0)	-13.2 (-51.3)	39.4 (66.0)	0.83 (0.30)	-4.2 (0.7)	-8.3 (1.4)	19.1 (39.9)
Monthly	0.72 (0.28)	-7.4 (-28.9)	-13.1 (-51.3)	26.9 (50.3)	0.85 (0.42)	-4.2 (0.7)	-8.2 (1.4)	13.1 (26.1)
Yearly	0.64 (0.40)	-9.8 (-33.5)	-16.1 (-54.9)	23.5 (43.1)	0.77 (0.31)	-5.4 (-3.6)	-10.1 (-6.7)	12.5 (24.3)

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794 **Table E2: CV results of the reanalysis (outside bracket) and base simulation (in bracket) for PM₁₀ concentrations in**
 795 **different regions of China at different temporal scales**

PM ₁₀ (µg/m ³)	NCP				NE			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.79 (0.23)	-7.7 (-14.6)	-6.4 (-12.1)	43.7 (88.3)	0.71 (0.18)	-7.6 (-23.6)	-9.6 (-29.8)	39.8 (70.8)
Daily	0.86 (0.31)	-7.6 (-14.2)	-6.3 (-11.7)	30.9 (71.8)	0.79 (0.25)	-7.6 (-23.6)	-9.7 (-30.0)	27.1 (56.8)
Monthly	0.86 (0.38)	-7.6 (-14.2)	-6.3 (-11.8)	21.4 (44.9)	0.76 (0.29)	-7.7 (-23.6)	-9.8 (-30.0)	19.4 (39.6)
Yearly	0.85 (0.46)	-7.6 (-15.8)	-6.2 (-12.8)	17.6 (33.0)	0.67 (0.31)	-8.3 (-26.5)	-10.3 (-32.6)	18.4 (36.2)
	SE				SW			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.77 (0.18)	-4.4 (6.9)	-5.9 (9.4)	26.0 (61.2)	0.69 (0.15)	-5.1 (13.0)	-7.5 (19.1)	30.2 (66.2)
Daily	0.85 (0.23)	-4.1 (8.1)	-5.6 (11.1)	18.6 (52.0)	0.77 (0.21)	-5.0 (13.1)	-7.4 (19.6)	22.4 (56.5)
Monthly	0.85 (0.38)	-4.2 (7.5)	-5.7 (10.2)	13.7 (33.3)	0.76 (0.38)	-5.2 (12.5)	-7.8 (18.5)	18.7 (41.4)
Yearly	0.81 (0.36)	-4.7 (4.9)	-6.1 (6.5)	12.3 (26.3)	0.62 (0.38)	-6.8 (8.7)	-9.6 (12.2)	19.3 (35.7)
	NW				Central			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.46 (0.08)	-21.5 (-88.5)	-18.0 (-74.1)	105.5 (150.2)	0.61 (0.11)	-14.6 (-45.6)	-14.1 (-43.9)	57.3 (96.4)
Daily	0.56 (0.11)	-21.5 (-89.3)	-17.9 (-74.1)	85.5 (141.6)	0.72 (0.14)	-14.6 (-45.5)	-14.1 (-43.8)	42.1 (84.6)
Monthly	0.59 (0.17)	-20.8 (-89.5)	-17.2 (-74.0)	64.0 (118.9)	0.74 (0.28)	-14.6 (-45.3)	-14.1 (-43.8)	30.2 (62.5)
Yearly	0.58 (0.23)	-23.8 (-92.3)	-19.3 (-74.7)	55.8 (110.2)	0.67 (0.25)	-16.4 (-50.1)	-15.4 (-46.8)	28.0 (60.4)

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800 **Table E3: CV results of the reanalysis (outside bracket) and base simulation (in bracket) for SO₂ concentrations in**
 801 **different regions of China at different temporal scales**

SO ₂ (µg/m ³)	NCP				NE			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.62 (0.10)	-3.6 (26.4)	-9.4 (69.4)	31.5 (63.1)	0.46 (0.08)	-2.0 (5.1)	-6.9 (17.5)	34.8 (53.2)
Daily	0.74 (0.16)	-3.6 (26.4)	-9.4 (69.6)	22.8 (52.7)	0.62 (0.13)	-2.0 (5.1)	-7.0 (17.6)	23.8 (42.2)
Monthly	0.79 (0.19)	-3.7 (26.2)	-9.6 (68.4)	17.1 (43.6)	0.71 (0.14)	-2.0 (5.0)	-6.9 (17.3)	17.9 (34.9)
Yearly	0.81 (0.18)	-4.2 (23.7)	-10.2 (56.9)	13.3 (36.1)	0.56 (0.14)	-2.4 (2.7)	-7.6 (8.7)	15.9 (27.7)
	SE				SW			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.42 (0.01)	-1.0 (29.8)	-5.7 (169.6)	14.6 (69.3)	0.27 (0.01)	-1.9 (44.2)	-12.1 (277.2)	16.7 (88.1)
Daily	0.55 (0.01)	-1.0 (29.9)	-5.7 (170.2)	10.5 (63.3)	0.38 (0.01)	-1.9 (44.1)	-12.2 (276.5)	11.8 (80.3)
Monthly	0.61 (0.01)	-1.0 (29.7)	-5.7 (168.6)	7.8 (55.8)	0.46 (0.02)	-2.0 (43.9)	-12.4 (273.7)	9.1 (73.7)
Yearly	0.66 (0.01)	-1.4 (28.0)	-7.1 (144.5)	7.9 (52.7)	0.53 (0.01)	-2.7 (41.2)	-15.2 (231.3)	9.5 (68.3)
	NW				Central			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.31 (0.01)	-0.3 (9.4)	-2.3 (61.6)	22.7 (40.4)	0.30 (0.02)	-4.4 (13.2)	-17.0 (51.3)	36.0 (58.9)
Daily	0.42 (0.01)	-0.3 (9.4)	-1.8 (62.2)	17.8 (36.2)	0.49 (0.03)	-4.4 (13.2)	-17.0 (51.5)	23.6 (49.1)
Monthly	0.48 (0.03)	-0.3 (9.3)	-2.2 (61.1)	13.4 (30.3)	0.59 (0.03)	-4.4 (13.1)	-17.0 (51.0)	18.2 (43.2)
Yearly	0.29 (0.00)	-1.9 (6.6)	-10.5 (35.9)	15.8 (28.0)	0.50 (0.00)	-5.6 (8.6)	-19.0 (29.3)	18.6 (40.2)

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807 **Table E4: CV results of the reanalysis (outside bracket) and base simulation (in bracket) for NO₂ concentrations in**
 808 **different regions of China at different temporal scales**

NO ₂ (µg/m ³)	NCP				NE			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.67 (0.20)	-1.4 (-3.0)	-3.5 (-7.1)	16.8 (26.5)	0.61 (0.27)	-1.6 (-5.9)	-5.0 (-19.1)	15.8 (22.4)
Daily	0.72 (0.22)	-1.4 (-2.9)	-3.3 (-7.1)	12.4 (20.8)	0.66 (0.34)	-1.5 (-5.9)	-4.9 (-19.0)	11.7 (17.2)
Monthly	0.72 (0.24)	-1.4 (-2.9)	-3.3 (-7.1)	9.3 (15.5)	0.64 (0.37)	-1.5 (-5.9)	-5.0 (-19.1)	9.3 (13.7)
Yearly	0.67 (0.36)	-1.4 (-3.8)	-3.3 (-9.0)	7.5 (11.0)	0.64 (0.45)	-1.5 (-6.4)	-4.8 (-20.3)	7.8 (11.7)
	SE				SW			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.64 (0.23)	-1.9 (-1.3)	-5.9 (-4.0)	14.9 (24.1)	0.49 (0.19)	-3.9 (-9.9)	-14.0 (-35.7)	16.4 (23.2)
Daily	0.71 (0.28)	-1.8 (-1.3)	-5.8 (-4.0)	11.2 (19.2)	0.55 (0.28)	-3.9 (-9.9)	-14.0 (-35.7)	12.8 (18.7)
Monthly	0.72 (0.36)	-1.8 (-1.2)	-5.8 (-3.9)	8.8 (14.7)	0.48 (0.32)	-4.0 (-10.0)	-14.4 (-36.0)	12.6 (17.2)
Yearly	0.66 (0.49)	-1.9 (-2.2)	-6.0 (-6.6)	7.8 (11.7)	0.46 (0.37)	-4.6 (-11.0)	-16.1 (-38.7)	11.8 (16.4)
	NW				Central			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.46 (0.20)	-4.3 (-18.0)	-12.9 (-54.4)	24.3 (31.5)	0.50 (0.20)	-5.2 (-16.7)	-15.1 (-48.3)	20.5 (29.2)
Daily	0.55 (0.27)	-4.1 (-18.0)	-12.5 (-54.4)	18.3 (27.0)	0.58 (0.28)	-5.2 (-16.7)	-15.0 (-48.3)	15.4 (24.7)
Monthly	0.59 (0.40)	-4.2 (-18.0)	-12.7 (-54.3)	15.3 (23.8)	0.61 (0.40)	-5.2 (-16.6)	-15.1 (-48.3)	12.8 (21.6)
Yearly	0.40 (0.36)	-6.0 (-19.7)	-17.3 (-56.3)	16.5 (23.8)	0.55 (0.36)	-5.6 (-17.6)	-16.0 (-50.1)	12.2 (21.4)

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816 **Table E5: CV results of the reanalysis (outside bracket) and base simulation (in bracket) for CO concentrations in**
 817 **different regions of China at different temporal scales**

CO (mg/m ³)	NCP				NE			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.67 (0.25)	-0.03 (-0.59)	-2.49 (-43.4)	0.64 (1.13)	0.50 (0.20)	-0.05 (-0.51)	-5.3 (-51.9)	0.59 (0.88)
Daily	0.72 (0.31)	-0.03 (-0.59)	-2.15 (-43.3)	0.50 (0.99)	0.56 (0.25)	-0.05 (-0.51)	-4.9 (-51.7)	0.46 (0.78)
Monthly	0.74 (0.34)	-0.03 (-0.59)	-2.24 (-43.5)	0.38 (0.85)	0.59 (0.25)	-0.05 (-0.51)	-5.2 (-52.0)	0.37 (0.70)
Yearly	0.71 (0.14)	-0.04 (-0.64)	-2.75 (-45.1)	0.32 (0.85)	0.55 (0.14)	-0.06 (-0.56)	-5.9 (-54.0)	0.35 (0.74)
	SE				SW			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.42 (0.13)	-0.06 (-0.36)	-6.4 (-38.2)	0.39 (0.62)	0.36 (0.07)	-0.08 (-0.32)	-9.4 (-36.4)	0.46 (0.65)
Daily	0.45 (0.15)	-0.06 (-0.36)	-6.1 (-38.0)	0.34 (0.57)	0.40 (0.08)	-0.08 (-0.31)	-9.1 (-36.3)	0.39 (0.59)
Monthly	0.44 (0.14)	-0.06 (-0.36)	-6.2 (-38.1)	0.28 (0.51)	0.40 (0.08)	-0.08 (-0.32)	-9.4 (-36.7)	0.34 (0.54)
Yearly	0.38 (0.05)	-0.06 (-0.38)	-6.5 (-39.3)	0.25 (0.50)	0.36 (0.01)	-0.09 (-0.36)	-10.1 (-39.1)	0.36 (0.57)
	NW				Central			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.38 (0.12)	-0.19 (-1.02)	-15.0 (-79.3)	1.13 (1.55)	0.44 (0.22)	-0.13 (-0.76)	-11.2 (-65.2)	0.73 (1.11)
Daily	0.45 (0.18)	-0.19 (-1.01)	-14.6 (-79.2)	0.92 (1.43)	0.49 (0.27)	-0.13 (-0.76)	-10.8 (-65.1)	0.62 (1.04)
Monthly	0.50 (0.29)	-0.19 (-1.02)	-15.1 (-79.3)	0.75 (1.32)	0.53 (0.32)	-0.13 (-0.76)	-11.1 (-65.2)	0.52 (0.97)
Yearly	0.13 (0.12)	-0.31 (-1.18)	-21.1 (-80.8)	0.85 (1.35)	0.19 (0.08)	-0.17 (-0.84)	-13.3 (-67.3)	0.69 (1.08)

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825 **Table E6: CV results of the reanalysis (outside bracket) and base simulation (in bracket) for O₃ concentrations in**
 826 **different regions of China at different temporal scales**

O ₃ (µg/m ³)	NCP				NE			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.83 (0.50)	-3.9 (-24.5)	-6.1 (-39.1)	22.1 (44.3)	0.76 (0.38)	-3.6 (-15.4)	-6.0 (-25.6)	21.0 (36.7)
Daily	0.83 (0.48)	-3.8 (-24.5)	-6.0 (-39.1)	16.3 (37.0)	0.76 (0.34)	-3.6 (-15.3)	-5.9 (-25.5)	16.4 (30.8)
Monthly	0.85 (0.62)	-3.8 (-24.4)	-6.1 (-39.1)	12.6 (31.6)	0.76 (0.42)	-3.6 (-15.2)	-5.9 (-25.4)	13.1 (25.2)
Yearly	0.72 (0.29)	-3.7 (-23.1)	-6.2 (-38.6)	9.2 (26.8)	0.62 (0.18)	-3.5 (-14.4)	-6.1 (-25.0)	10.0 (20.4)
	SE				SW			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.77 (0.57)	-2.3 (-10.0)	-3.9 (-17.4)	21.1 (38.2)	0.69 (0.40)	0.8 (9.6)	1.4 (18.0)	22.2 (33.6)
Daily	0.69 (0.44)	-2.2 (-10.0)	-3.8 (-17.3)	15.8 (31.0)	0.64 (0.34)	0.8 (9.7)	1.5 (18.1)	17.4 (26.7)
Monthly	0.69 (0.43)	-2.2 (-10.0)	-3.9 (-17.3)	12.4 (24.2)	0.64 (0.44)	0.8 (9.7)	1.6 (18.1)	14.4 (21.1)
Yearly	0.45 (0.07)	-2.4 (-10.1)	-4.2 (-17.7)	10.0 (20.7)	0.42 (0.28)	1.3 (10.0)	2.6 (19.4)	12.0 (17.7)
	NW				Central			
	R ²	MBE	NMB(%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.52 (0.31)	-2.7 (-2.2)	-4.6 (-3.8)	28.3 (33.2)	0.71 (0.45)	-1.5 (-0.8)	-2.5 (-1.3)	23.9 (32.5)
Daily	0.50 (0.31)	-2.6 (-2.1)	-4.5 (-3.6)	22.9 (26.6)	0.67 (0.42)	-1.4 (-0.7)	-2.4 (-1.1)	17.8 (23.9)
Monthly	0.58 (0.42)	-2.6 (-2.1)	-4.5 (-3.6)	19.1 (22.1)	0.72 (0.56)	-1.4 (-0.7)	-2.4 (-1.2)	13.9 (17.6)
Yearly	0.37 (0.24)	-1.6 (-0.6)	-2.9 (-1.1)	15.7 (17.1)	0.53 (0.30)	-0.8 (0.2)	-1.4 (0.4)	11.6 (14.1)

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834 **Author contributions**

835 X.T., J.Z., and Z.W. conceived and designed the project; H.W., L.K., X.T., and L.W. established the data assimilation system;
836 Q.W. and L.K. performed the meteorology simulations; X.T., L.K., H.C., H.W., H.Z., G.J. and M.L. conducted the ensemble
837 simulations with the NAQPMS model; J.L., L.Z., W.W., B.L., Q.W., D.C. and T.S. provided the air quality monitoring data;
838 W.H. executed the quality control of the observation data; F.L. estimated the representativeness error of the observations; and
839 L.K. carried out the CAQRA calculations, generated the figures and wrote the paper with comments provided by G.C.

840 **Competing interests**

841 The authors declare that they have no conflicts of interest.

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847 valuable comments.

848 **Tables**

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850 **Table 1: Uncertainties in the emissions of the different species**

Species	SO ₂ ^a	NO _x ^a	CO ^a	Non- methane volatile organic compounds (NMVOCs) ^a	NH ₃ ^b	PM ₁₀ ^a	PM _{2.5} ^a	Black carbon (BC) ^a	Organic carbon (OC) ^a
Emission Uncertainty	12%	31%	70%	68%	53%	132%	130%	208%	258%

851 ^a Emission uncertainty obtained from Zhang et al. (2009)

852 ^b Emission uncertainty obtained from Streets et al. (2003)

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854 **Table 2: Site-based cross-validation results for the reanalysis data (outside brackets) and base simulation (inside**
 855 **brackets) from 2013 to 2018 at the different temporal scales**

	PM _{2.5} (µg/m ³)				PM ₁₀ (µg/m ³)			
	R ²	MBE	NMB (%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.81 (0.26)	-2.6 (17.6)	-4.9 (34.7)	21.3 (54.1)	0.72 (0.17)	-6.8 (-7.6)	-7.8 (-8.7)	39.3 (75.7)
Daily	0.86 (0.32)	-2.5 (17.4)	-4.9 (34.3)	15.1 (46.4)	0.81 (0.22)	-6.7 (-7.0)	-7.7 (-8.1)	28.8 (64.1)
Monthly	0.88 (0.40)	-2.5 (17.4)	-5.0 (34.1)	10.3 (33.6)	0.83 (0.28)	-6.7 (-7.3)	-7.7 (-8.4)	21.1 (44.4)
Yearly	0.86 (0.37)	-3.0 (15.2)	-5.6 (28.7)	9.0 (28.9)	0.79 (0.27)	-7.5 (-10.2)	-8.3 (-11.3)	19.1 (38.2)
	SO ₂ (µg/m ³)				NO ₂ (µg/m ³)			
	R ²	MBE	NMB (%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.52 (0.03)	-2.0 (25.5)	-8.5 (106.6)	24.9 (67.2)	0.61 (0.22)	-2.3 (-5.0)	-6.9 (-14.8)	16.4 (24.9)
Daily	0.67 (0.04)	-2.0 (25.6)	-8.5 (106.9)	17.5 (59.3)	0.67 (0.27)	-2.3 (-5.0)	-6.8 (-14.8)	12.3 (19.9)
Monthly	0.74 (0.04)	-2.1 (25.4)	-8.6 (105.7)	13.2 (52.0)	0.67 (0.34)	-2.3 (-5.0)	-6.8 (-14.8)	10.0 (15.9)
Yearly	0.71 (0.04)	-2.6 (23.1)	-9.9 (87.2)	12.0 (47.5)	0.62 (0.42)	-2.5 (-5.9)	-7.3 (-17.3)	9.1 (13.6)
	CO (mg/m ³)				O ₃ (µg/m ³)			
	R ²	MBE	NMB (%)	RMSE	R ²	MBE	NMB (%)	RMSE
Hourly	0.55 (0.17)	-0.06 (-0.47)	-6.1 (-44.7)	0.54 (0.87)	0.76 (0.35)	-2.3 (-10.5)	-4.0 (-17.8)	21.9 (38.3)
Daily	0.61 (0.20)	-0.06 (-0.47)	-5.8 (-44.6)	0.44 (0.77)	0.74 (0.25)	-2.3 (-10.4)	-3.9 (-17.8)	16.6 (31.3)
Monthly	0.62 (0.21)	-0.06 (-0.47)	-6.0 (-44.7)	0.36 (0.69)	0.74 (0.28)	-2.3 (-10.4)	-3.9 (-17.8)	13.1 (25.3)
Yearly	0.52 (0.09)	-0.08 (-0.51)	-6.9 (-46.7)	0.37 (0.72)	0.53 (0.03)	-2.2 (-9.8)	-3.8 (-17.2)	10.4 (21.2)

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863 **Table 3: Calculated annual trends of the PM_{2.5} and PM₁₀ concentrations in China**

	PM _{2.5} (µg/m ³)			PM ₁₀ (µg/m ³)		
	Observation	Cross-validation	Base simulation	Observation	Cross-validation	Base simulation
China	-5.8 (-13.4, -3.5)^a	-5.0 (-12.6, -3.1)	-2.0 (-3.6, -0.7)	-7.2 (-18.4, -3.2)	-6.0 (-17.0, -2.9)	-2.5 (-3.6, -0.7)
NCP	-7.0 (-15.7, -5.5)	-6.6 (-14.5, -4.8)	-3.5 (-4.7, -1.9)	-8.3 (-20.4, -5.1)	-7.6 (-19.2, -4.4)	-4.2 (-4.7, -1.9)
NE	-7.5 (-11.0, -3.9)	-6.7 (-10.0, -3.5)	-3.2 (-5.8, -1.2)	-11.2 (-17.4, -4.7)	-10.4 (-16.4, -4.7)	-3.7 (-5.8, -1.2)
SE	-5.2 (-11.3, -2.8)	-4.9 (-10.6, -2.7)	-0.9 (-3.1, 1.3)	-6.0 (-14.9, -2.4)	-5.8 (-13.2, -1.9)	-1.6 (-3.1, 1.3)
SW	-6.3 (-12.8, -2.6)	-4.9 (-12.2, -2.4)	-1.4 (-7.5, 0.4)	-7.9 (-19.9, -2.2)	-5.5 (-17.5, -2.1)	-1.3 (-7.5, 0.4)
NW	-5.7 (-11.6, 2.1) ^b	-3.3 (-10.7, 1.8)	-1.3 (-4.9, 2.9)	-0.5 (-14.4, 1.6)	-2.2 (-8.5, 3.4)	-2.3 (-4.9, 2.9)
Central	-5.8 (-19.8, -0.8)	-3.6 (-17.7, 0.2)	-0.6 (-5.9, 0.9)	-8.9 (-28.5, 0.2)	-6.8 (-26.9, 0.5)	-2.0 (-5.9, 0.9)

864 ^a The bold font denotes that the calculated trend is significant at the 0.05 significance level, and the values in brackets denote
865 the 95% confidence interval.

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871 **Table 4: Independent validation results of the CAQRA dataset (outside brackets) and base simulation (inside brackets)**
872 **against the observation data retrieved from the U.S. Department State Air Quality Monitoring Program over China**

	R ²	MBE (µg/m ³)	NMB (%)	RMSE (µg/m ³)
Beijing	0.86 (0.37)	-0.3 (11.4)	-0.3 (13.2)	33.6 (75.6)
Shanghai	0.86 (0.34)	5.5 (39.6)	10.9 (78.3)	17.1 (64.8)
Chengdu	0.85 (0.19)	-7.1 (59.3)	-8.9 (74.7)	23.1 (91.5)
Guangzhou	0.74 (0.09)	-3.3 (11.1)	-7.5 (25.1)	16.8 (38.8)
Shenyang	0.85 (0.29)	-2.2 (16.8)	-3.2 (24.3)	24.8 (59.1)

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876 **Table 5 Comparison of the accuracy of our PM_{2.5} reanalysis data to that of satellite estimates**

Reference	Spatial resolution	Temporal resolution	Temporal coverage	CV R ²	CV RMSE	Method
Ma et al. (2016)	0.1° × 0.1°	daily	2004–2013	0.79	27.4	LME + GAM
Xue et al. (2019)	0.1° × 0.1°	daily	2000–2016	0.56	30.2	CTM + HD-expansion + GAM
Xue et al. (2017)	0.1° × 0.1°	daily	2014	0.72	23.0	CTM + LME + spatiotemporal kriging
Chen et al. (2018)	0.1° × 0.1°	daily	2005–2016	0.83	18.1	RF
Lin et al. (2018)	1 km × 1km	daily	2001 – 2015	0.78 ^a	19.3 ^a	Semi-empirical
Chen et al. (2019)	3 km × 3 km	daily	2014 – 2015	0.86	15.0	XGBoost + NELRM
Yao et al. (2019)	6 km × 6 km	daily	2014	0.60	21.8	TEFR + GWR
You et al. (2016)	0.1° × 0.1°	daily	2014	0.79	18.6	GWR
Zhan et al. (2017)	0.5° × 0.5°	daily	2014	0.76	23.0	GW-GBM
Li et al. (2017b)	0.1° × 0.1°	daily	2015	0.82	16.4	Geoi-DBN
Liu et al. (2019)	0.125° × 0.125°	hourly	2016	0.86	17.3	RF
This study	15 km × 15km	hourly	2013–2018	0.81	21.3	EnKF
		daily	2013–2018	0.86	15.1	EnKF

877 ^a The accuracy of the PM_{2.5} estimates of Lin et al. (2018) was assessed at the monthly scale.

878 LME: Linear mixed-effect model

879 GWR: Geographically weighted regression model

880 GAM: Generalized additive model

881 HD-expansion: High-dimensional expansion

882 RF: Random forest

883 XGBoost: Extreme gradient boosting

884 NELRM: Non-linear exposure-lag-response model

885 TEFR: Time fixed-effects regression model

886 GW-GBM: Geographically weighted gradient boosting machine

887 Geoi-DBN: Geographical deep belief network

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889 **Table 6: Calculated annual trends of the SO₂, NO₂, CO and O₃ concentrations in China**

	SO ₂ (µg/m ³)			NO ₂ (µg/m ³)		
	Observation	Cross-validation	Base simulation	Observation	Cross-validation	Base simulation
China	-6.2 (-12.0, -3.9)^a	-4.9 (-10.3, -3.0)	-1.7 (-6.2, -0.8)	-2.6 (-5.9, 0.1)	-2.1 (-5.9, 0.1)	-0.9 (-3.0, -0.3)
NCP	-9.5 (-16.5, -7.2)	-8.1 (-14.5, -5.9)	-1.7 (-4.1, -1.4)	-2.0 (-5.9, 0.0)	-2.1 (-5.6, 0.1)	-0.6 (-1.6, -0.3)
NE	-6.8 (-14.6, -4.9)	-5.9 (-12.1, -4.1)	-1.8 (-7.6, -0.6)	-3.0 (-4.9, -1.1)	-3.3 (-5.4, -1.2)	-1.3 (-3.8, -0.3)
SE	-4.4 (-6.7, -2.5)	-3.7 (-5.6, -2.0)	-1.0 (-2.9, -0.1)	-2.4 (-5.3, 0.1)	-2.5 (-5.1, 0.1)	-1.0 (-1.8, -0.3)
SW	-4.2 (-8.8, -1.9)	-2.8 (-7.6, -1.3)	-3.4 (-15.6, -1.9)	-1.8 (-6.2, 0.3)	-1.6 (-6.5, 0.2)	-0.7 (-3.9, -0.2)
NW	-2.3 (-11.1, 0.6)	-4.2 (-7.7, -1.1)	-1.9 (-13.7, 1.0)	-3.4 (-8.4, 2.3)	-1.7 (-9.5, 1.3)	-1.0 (-6.5, 0.3)
Central	-7.9 (-17.5, -3.3)	-5.5 (-15.7, -2.3)	-0.6 (-10.2, 0.0)	-2.0 (-6.6, 1.9)	-1.0 (-8.0, 2.2)	-0.5 (-3.8, 0.1)
	CO (mg/m ³)			O ₃ (µg/m ³)		
	Observation	Cross-validation	Base simulation	Observation	Cross-validation	Base simulation
China	-0.12 (-0.17, -0.06)	-0.12 (-0.18, -0.07)	-0.02 (-0.05, -0.01)	3.5 (2.1, 5.0)	3.8 (2.1, 5.0)	2.0 (0.1, 5.9)
NCP	-0.18 (-0.25, -0.11)	-0.17 (-0.24, -0.11)	-0.03 (-0.05, -0.02)	5.3 (2.5, 8.7)	5.5 (2.4, 8.8)	1.4 (-0.5, 5.0)
NE	-0.13 (-0.21, -0.05)	-0.13 (-0.20, -0.06)	-0.03 (-0.07, -0.01)	4.8 (1.5, 10.0)	4.6 (1.4, 9.5)	2.8 (-0.4, 8.0)
SE	-0.06 (-0.09, -0.04)	-0.06 (-0.08, -0.04)	-0.01 (-0.02, -0.01)	2.3 (0.3, 3.4)	2.6 (0.8, 3.5)	1.7 (0.3, 3.0)
SW	-0.11 (-0.19, -0.04)	-0.09 (-0.21, -0.04)	-0.02 (-0.06, -0.01)	3.2 (1.2, 5.0)	3.5 (1.8, 5.4)	2.7 (-0.9, 7.1)
NW	-0.14 (-0.46, 0.04)	-0.14 (-0.30, 0.04)	-0.03 (-0.06, 0.00)	5.4 (1.6, 9.8)	4.0 (1.4, 10.1)	2.6 (-0.2, 8.8)
Central	-0.16 (-0.27, -0.09)	-0.17 (-0.25, -0.10)	-0.01 (-0.06, 0.00)	5.3 (2.3, 9.2)	4.5 (1.4, 7.8)	2.2 (-0.3, 7.7)

890 ^a The bold font denotes that the calculated trend is significant at the 0.05 significance level, and the values in brackets denote
891 the 95% confidence interval.

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895 **Table 7: Comparison of the data accuracy of CAQRA and CAMSRA in China**

	CAQRA				CAMSRA			
	SO ₂ (µg/m ³)	NO ₂ (µg/m ³)	CO (mg/m ³)	O ₃ (µg/m ³)	SO ₂ (µg/m ³)	NO ₂ (µg/m ³)	CO (mg/m ³)	O ₃ (µg/m ³)
R ²	0.53	0.61	0.55	0.77	0.04	0.23	0.13	0.00
MBE	-2.0	-2.3	-0.1	-2.3	19.4	1.7	-0.2	30.6
NMB (%)	-8.5	-6.9	-6.1	-4.0	81.2	5.2	-17.5	52.1
RMSE	24.8	16.4	0.5	21.9	54.5	27.3	0.9	55.2

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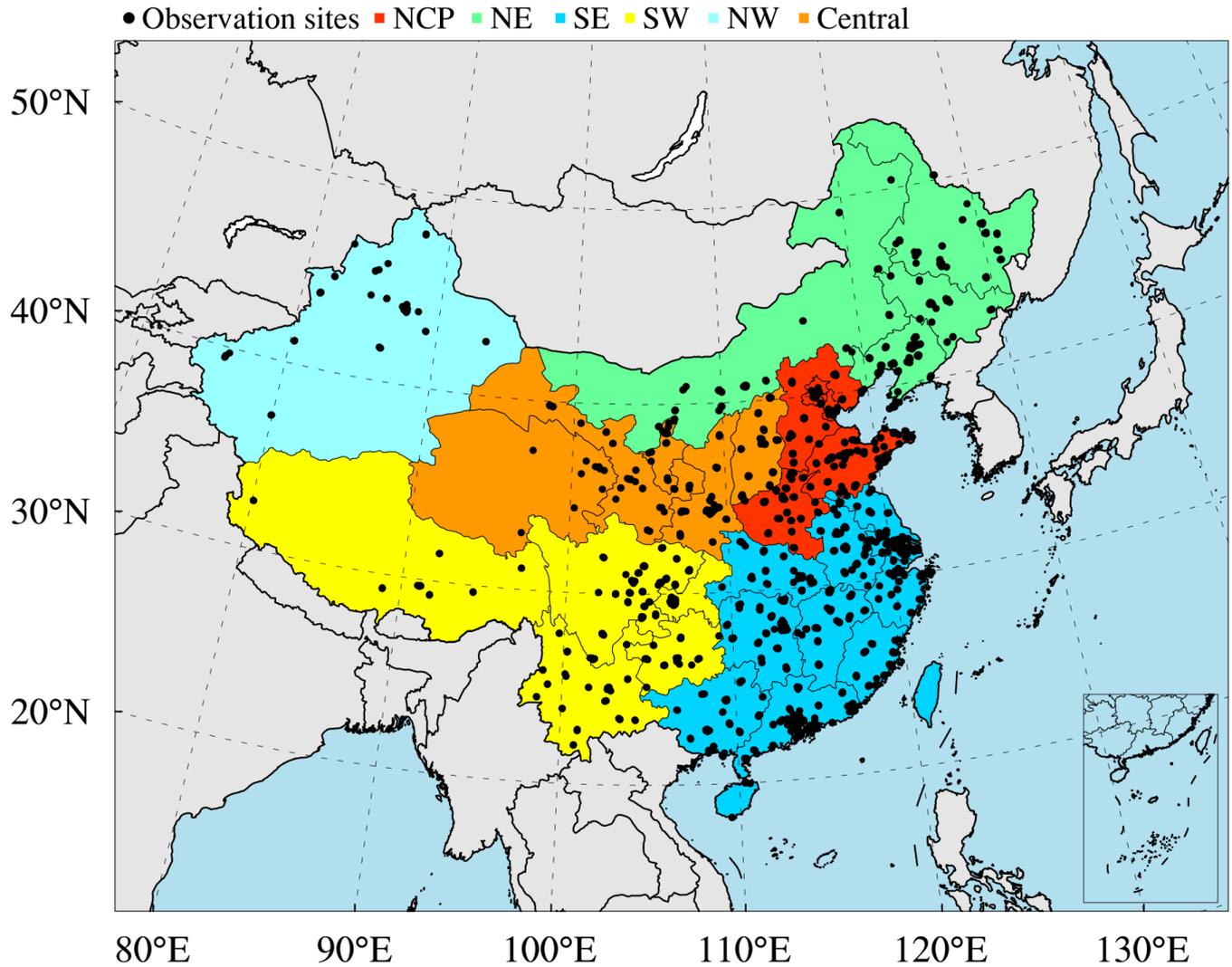
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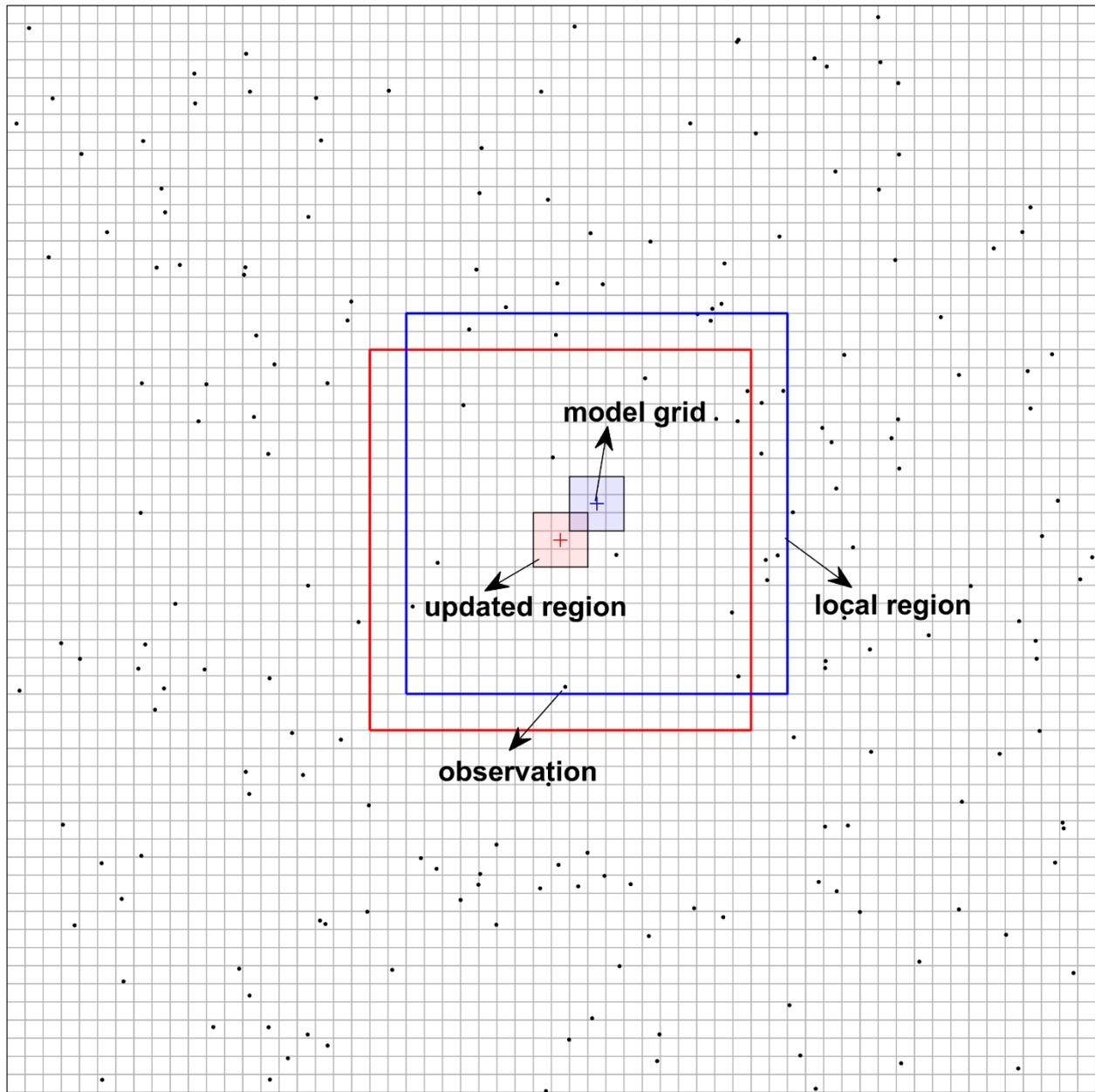
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922 **Figure 1: Modelling domain of the ensemble simulation overlain on the distribution of the observation sites of the CNEMC. The**
 923 **different colours denote the different regions in China, namely, the North China Plain (NCP), Northeast China (NE), Southwest**
 924 **China (SW), Southeast China (SE), Northwest China (NW) and Central China.**



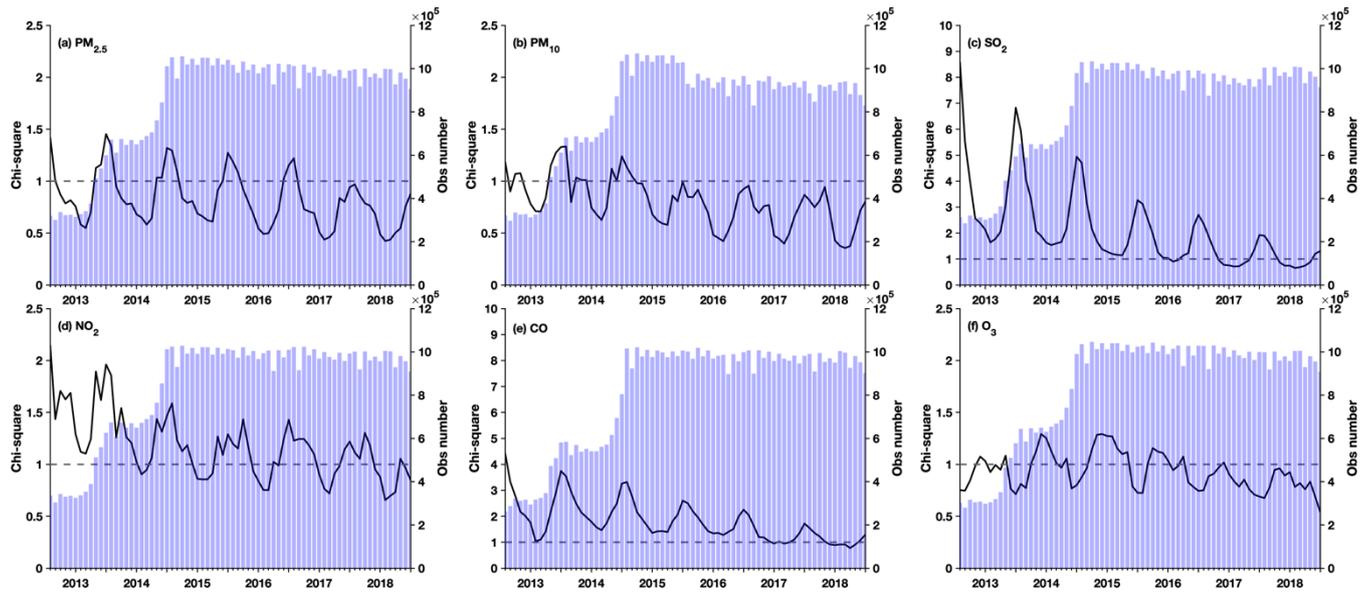
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926 **Figure 2: Illustration of the local analysis scheme used in the assimilation. The plus and dot symbols denote the centres**
 927 **of the model grids and the location of the observation sites, respectively. The large rectangular region denotes the local**
 928 **region, and the shaded region denotes the updated region.**

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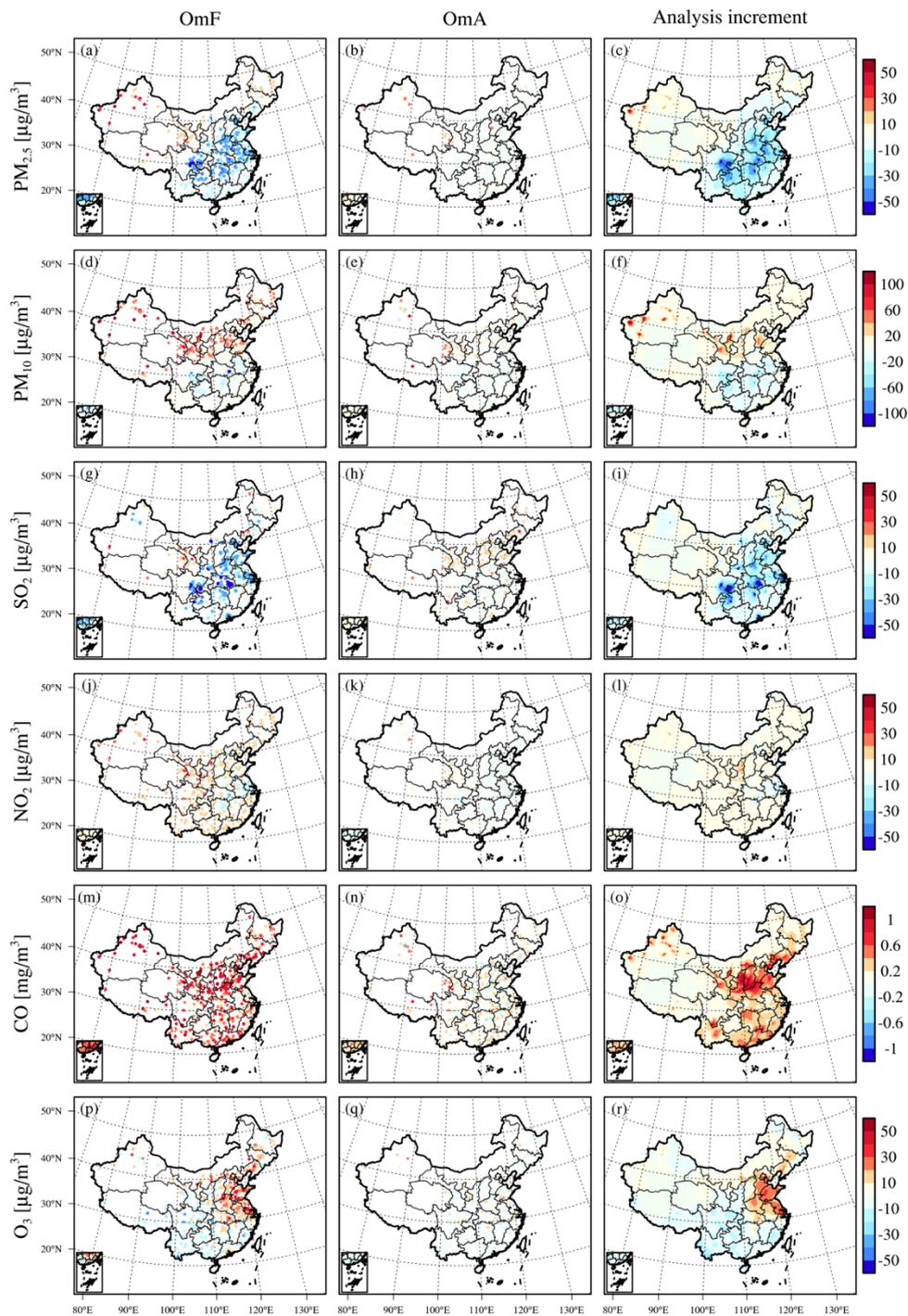
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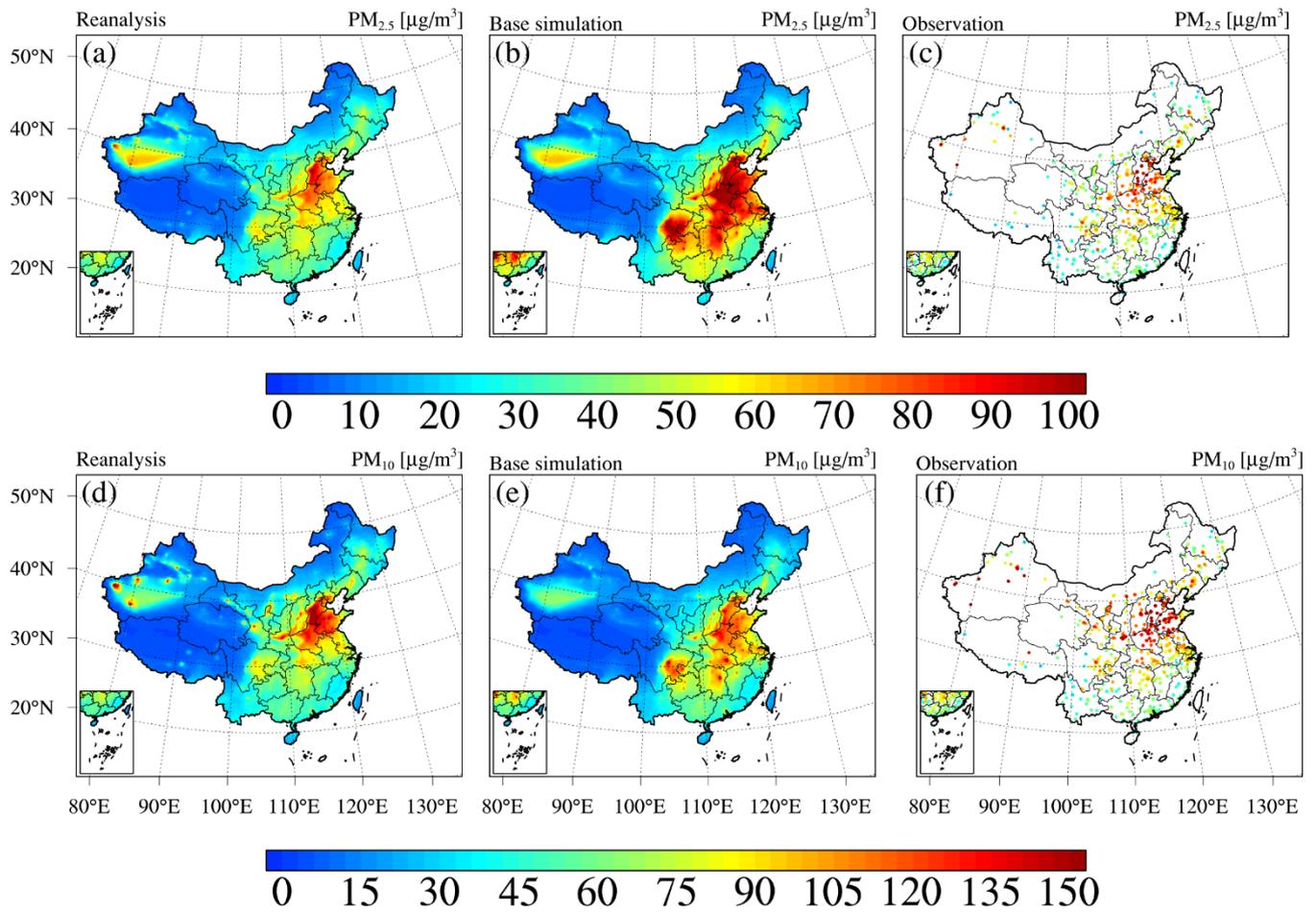
934 **Figure 3: Time series of the monthly mean χ^2 values (black line) and the number of assimilated observations per month**935 **(blue bars) for (a) $PM_{2.5}$, (b) PM_{10} , (c) SO_2 , (d) NO_2 , (e) CO and (f) O_3 .**

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938 **Figure 4: Spatial distributions of the six-year mean OmF (left panel), OmA (middle panel) and analysis increment**
 939 **(right panel) for different species in China.**

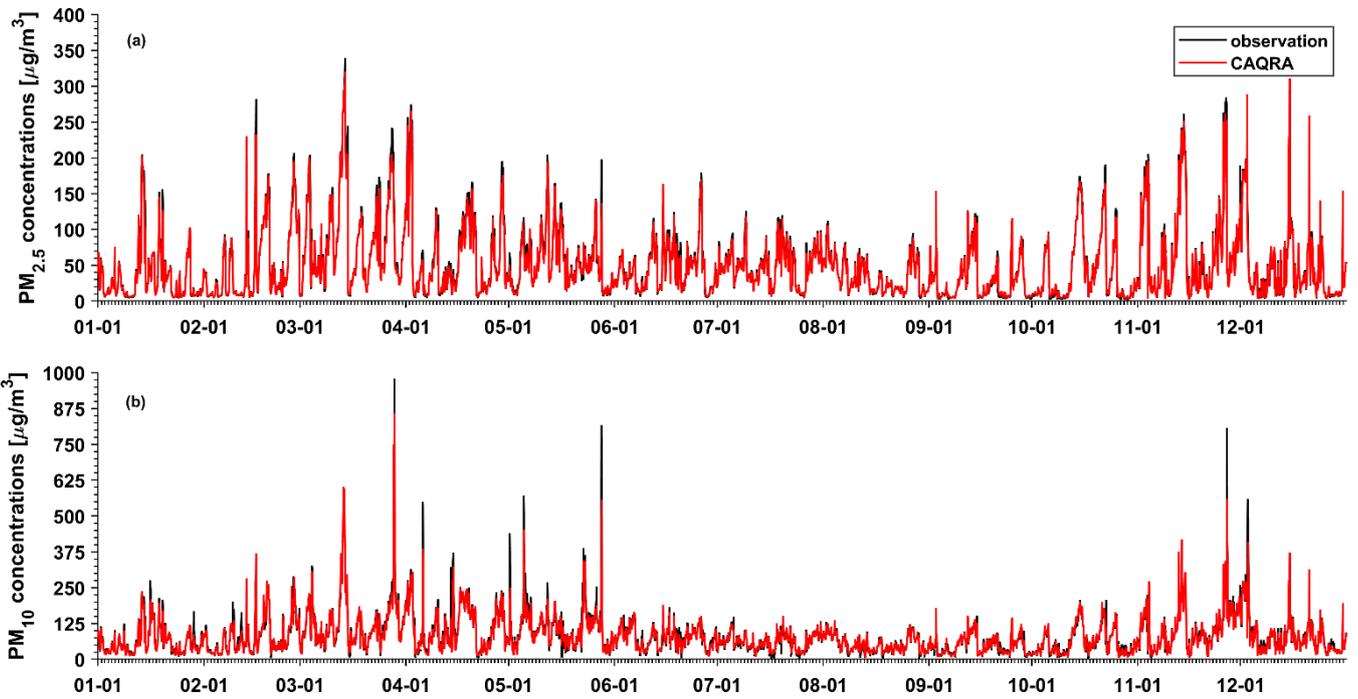


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941 **Figure 5: Spatial distributions of the (a–c) $PM_{2.5}$ and (d–f) PM_{10} concentrations in China from (a, d) CAQRA, (b, e)**

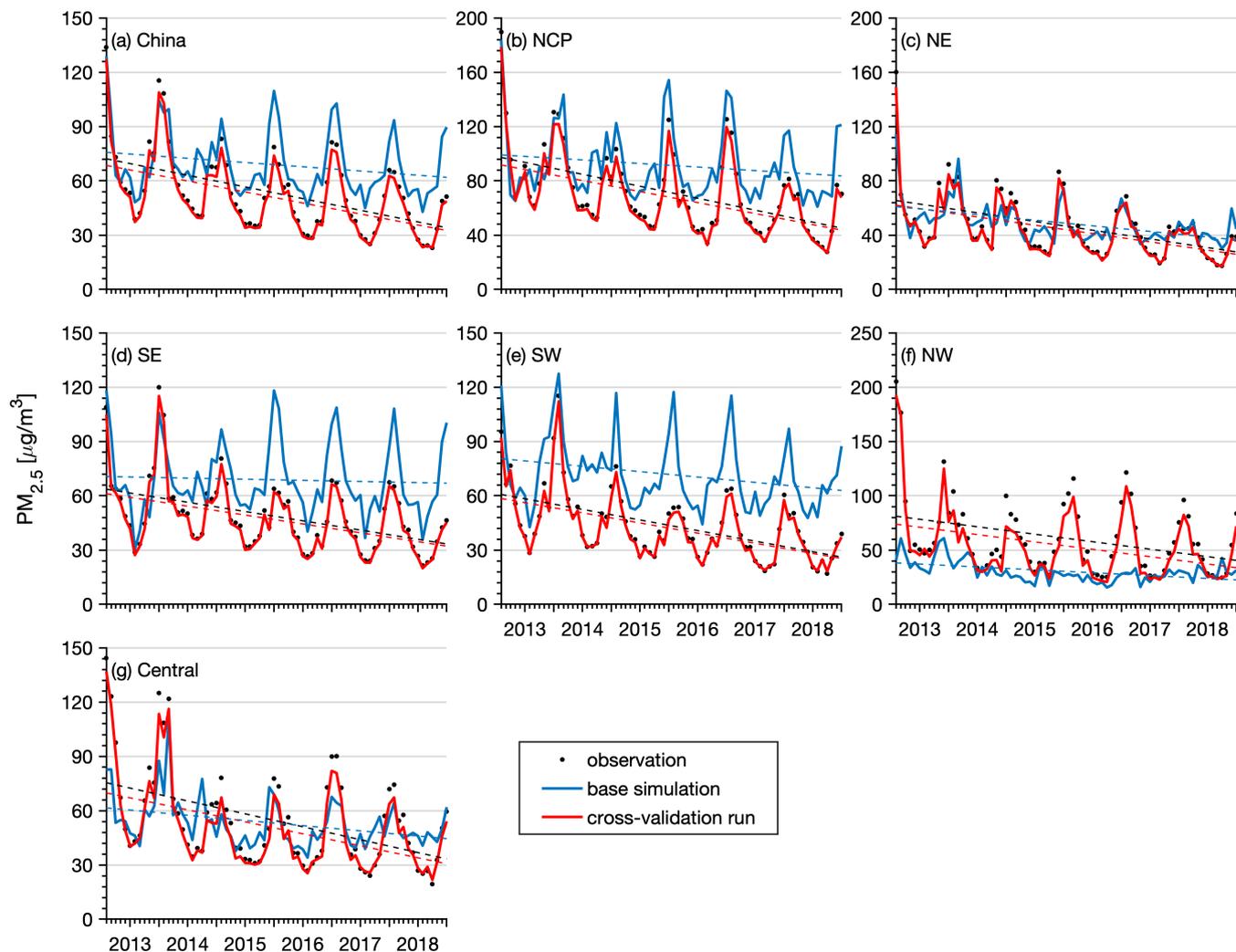
942 **base simulation and (c, f) observations averaged from 2013 to 2018.**

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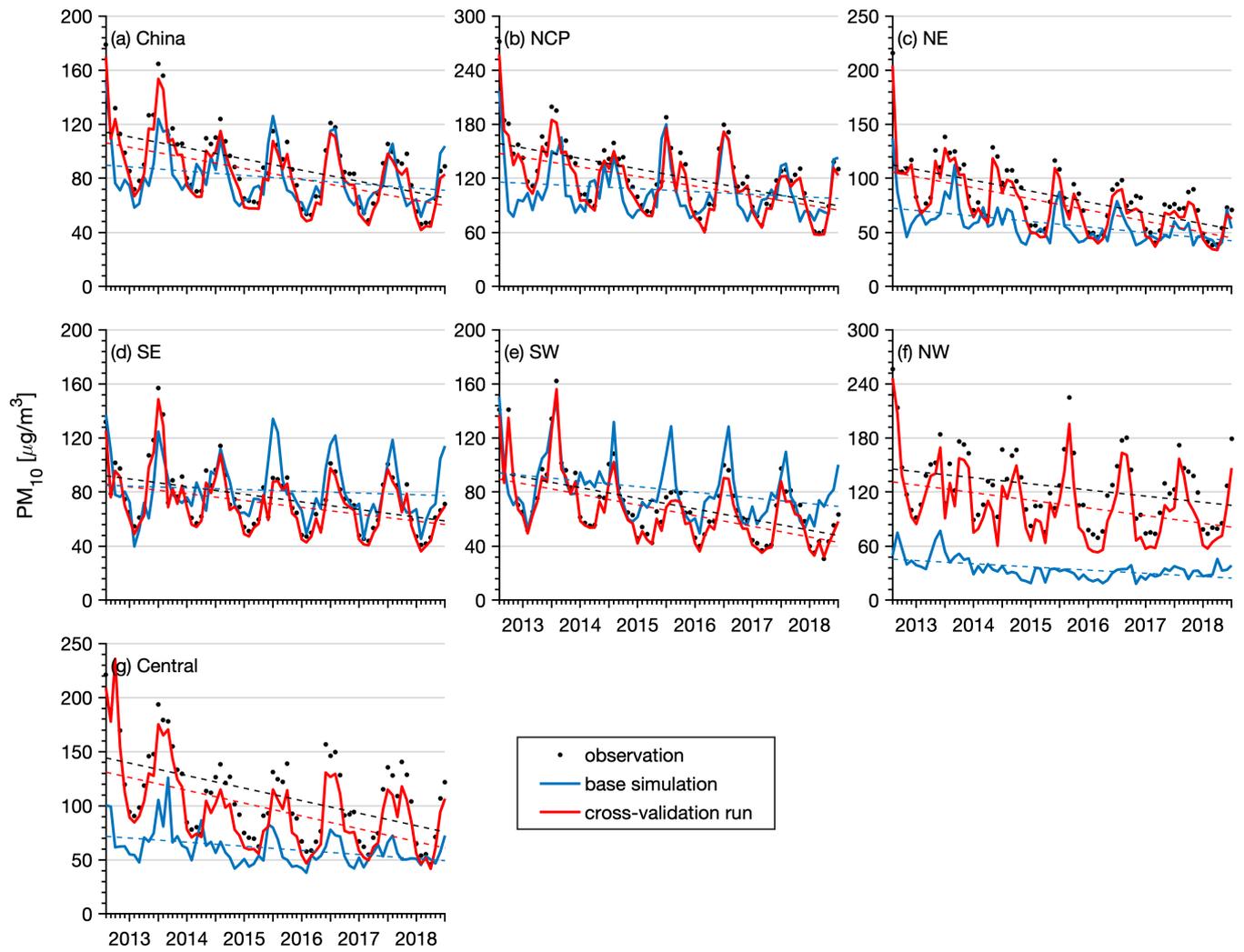
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945 **Figure 6: Time series of the site mean hourly (a) PM_{2.5} and (b) PM₁₀ concentrations in Beijing obtained from the**
946 **observations and CAQRA.**



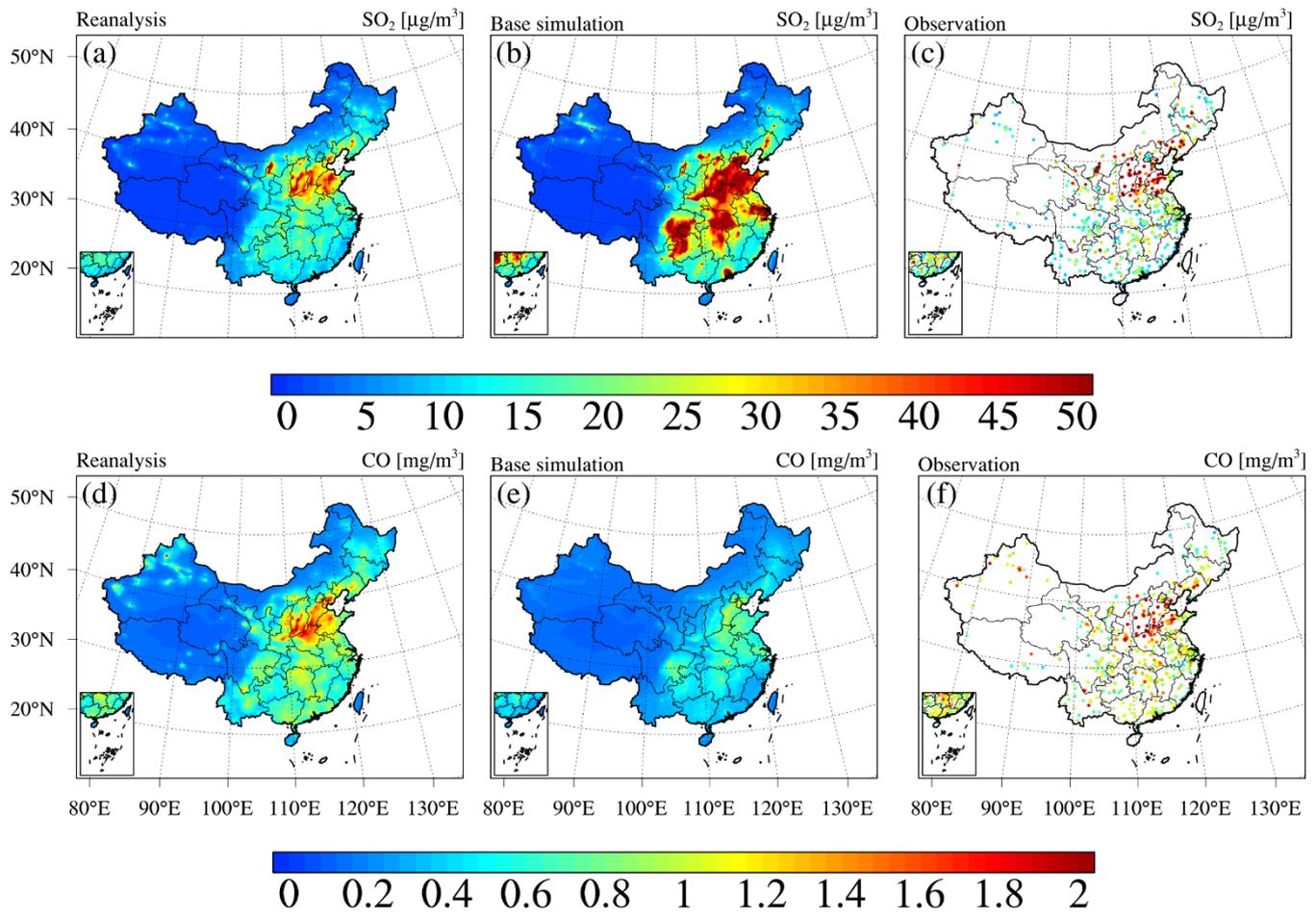
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948 **Figure 7: Time series of the monthly mean PM_{2.5} concentrations in (a) China, (b) NCP, (c) NE, (d) SE, (e) SW, (f) NW**
 949 **and (g) central regions obtained from the cross-validation run (red line), base simulation (blue line) and observations**
 950 **(black dots).**



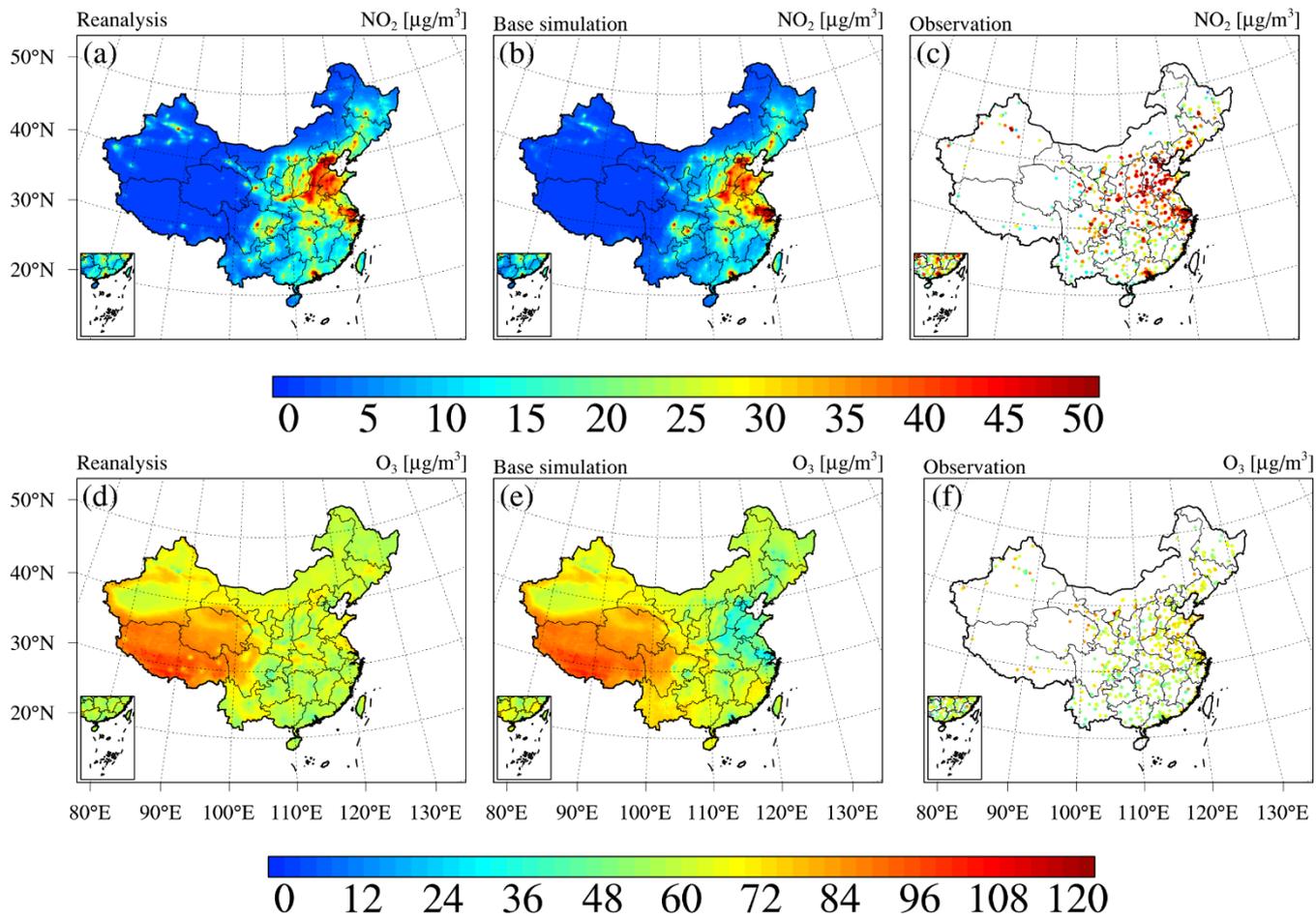
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952 **Figure 8: Same as Fig. 7 but for the PM₁₀ concentration.**



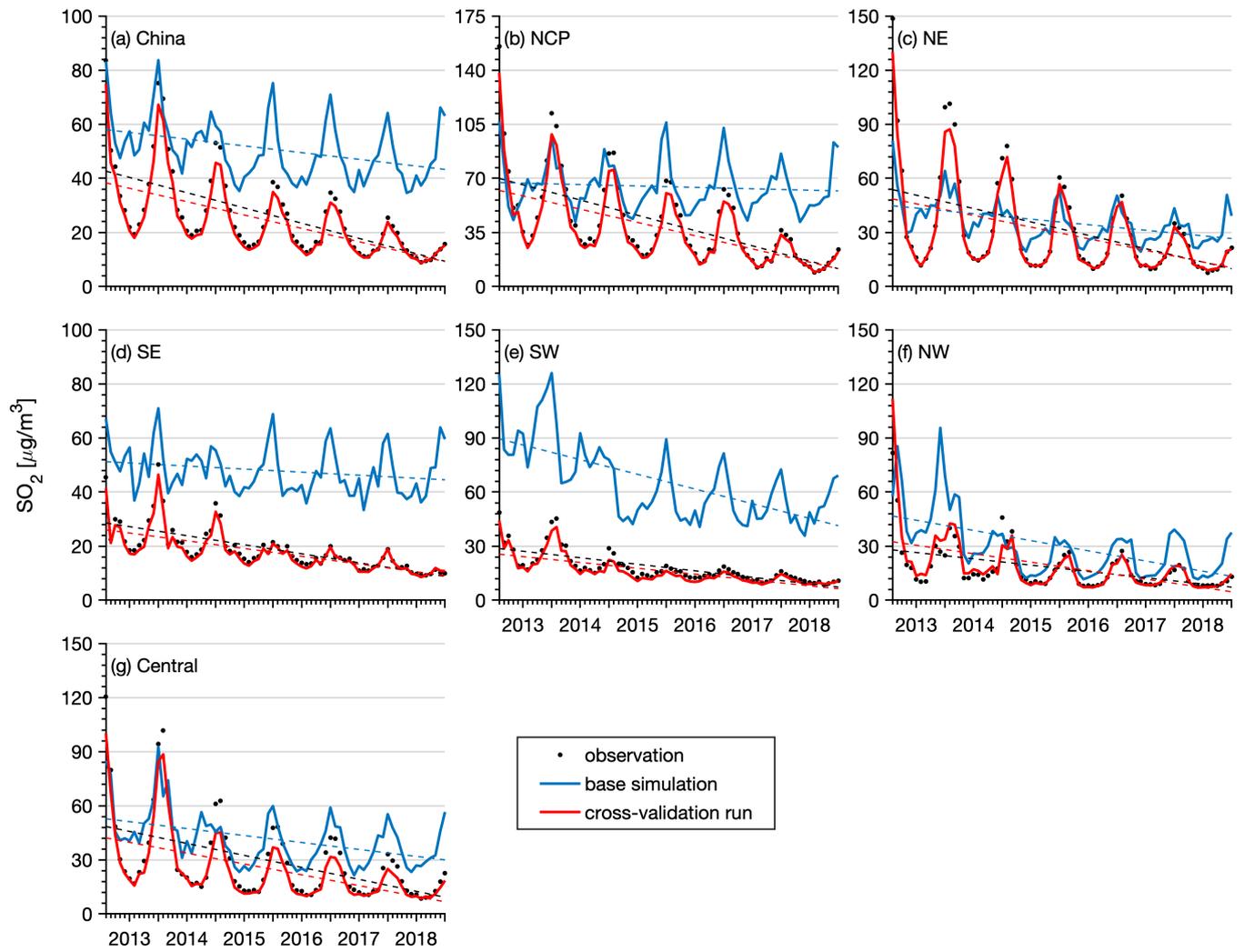
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954 **Figure 9: Same as Fig. 5 but for the SO_2 and CO concentrations.**



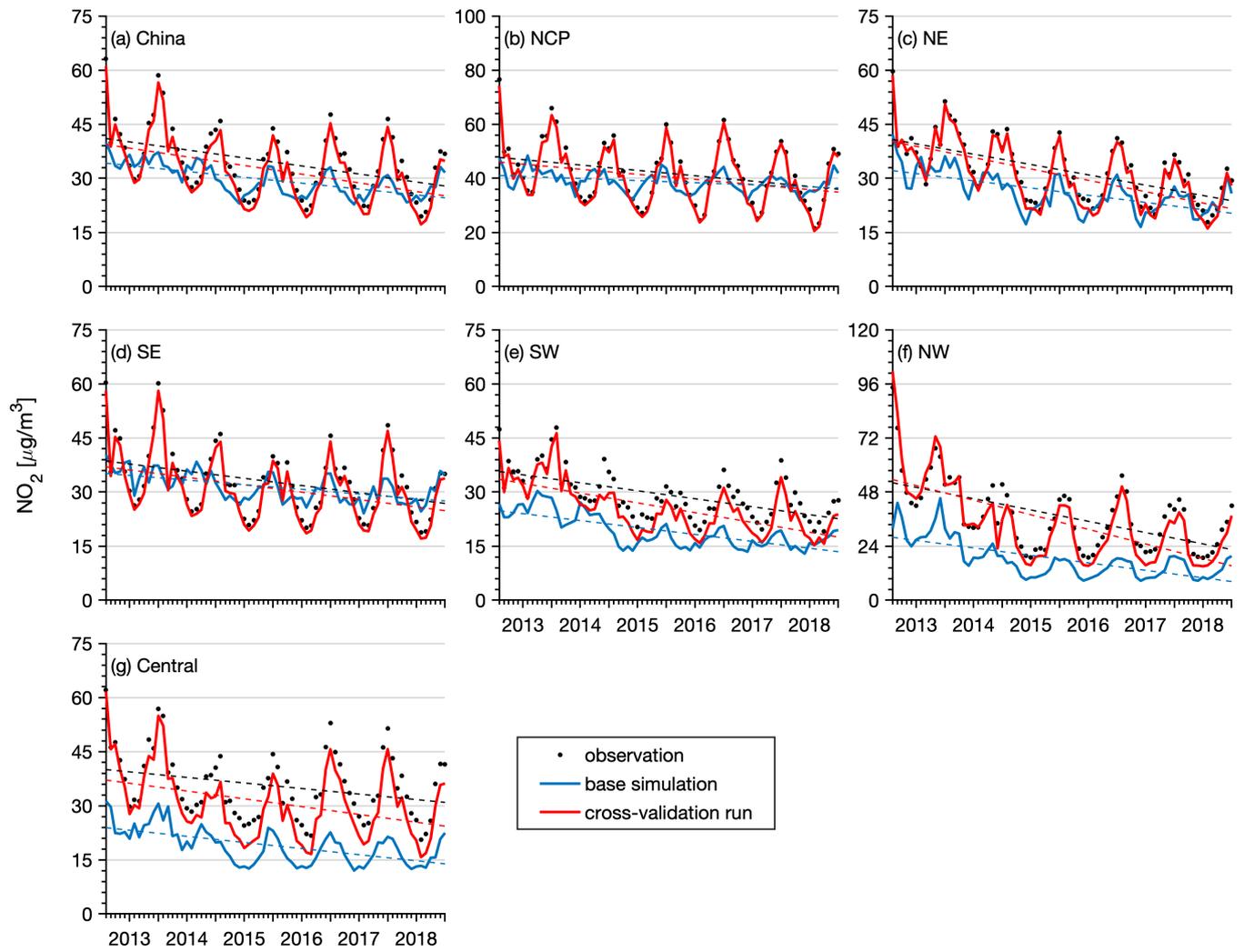
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956 **Figure 10: Same as Fig. 5 but for NO_2 and O_3 .**



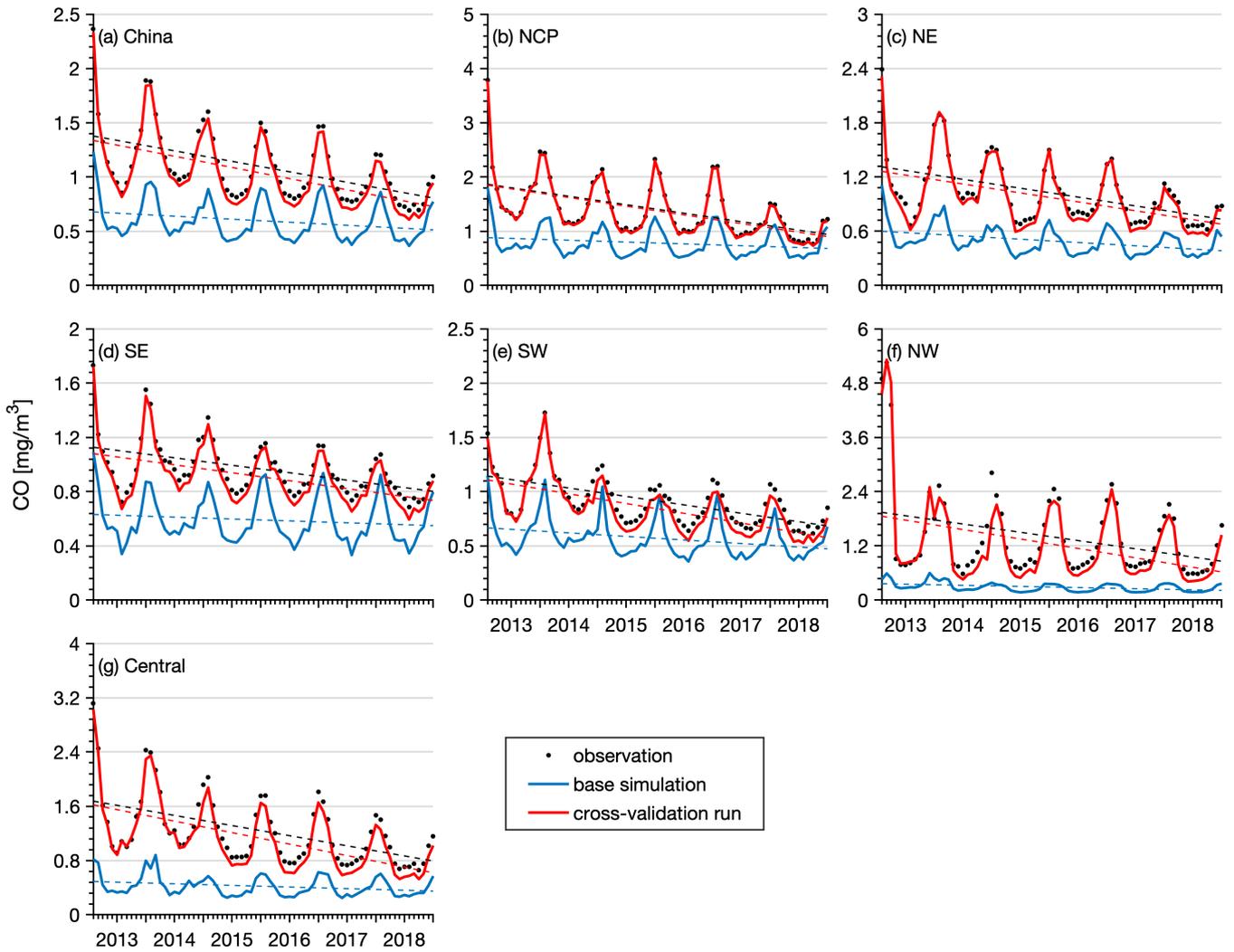
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958 **Figure 11: Same as Fig. 7 but for the SO₂ concentration.**



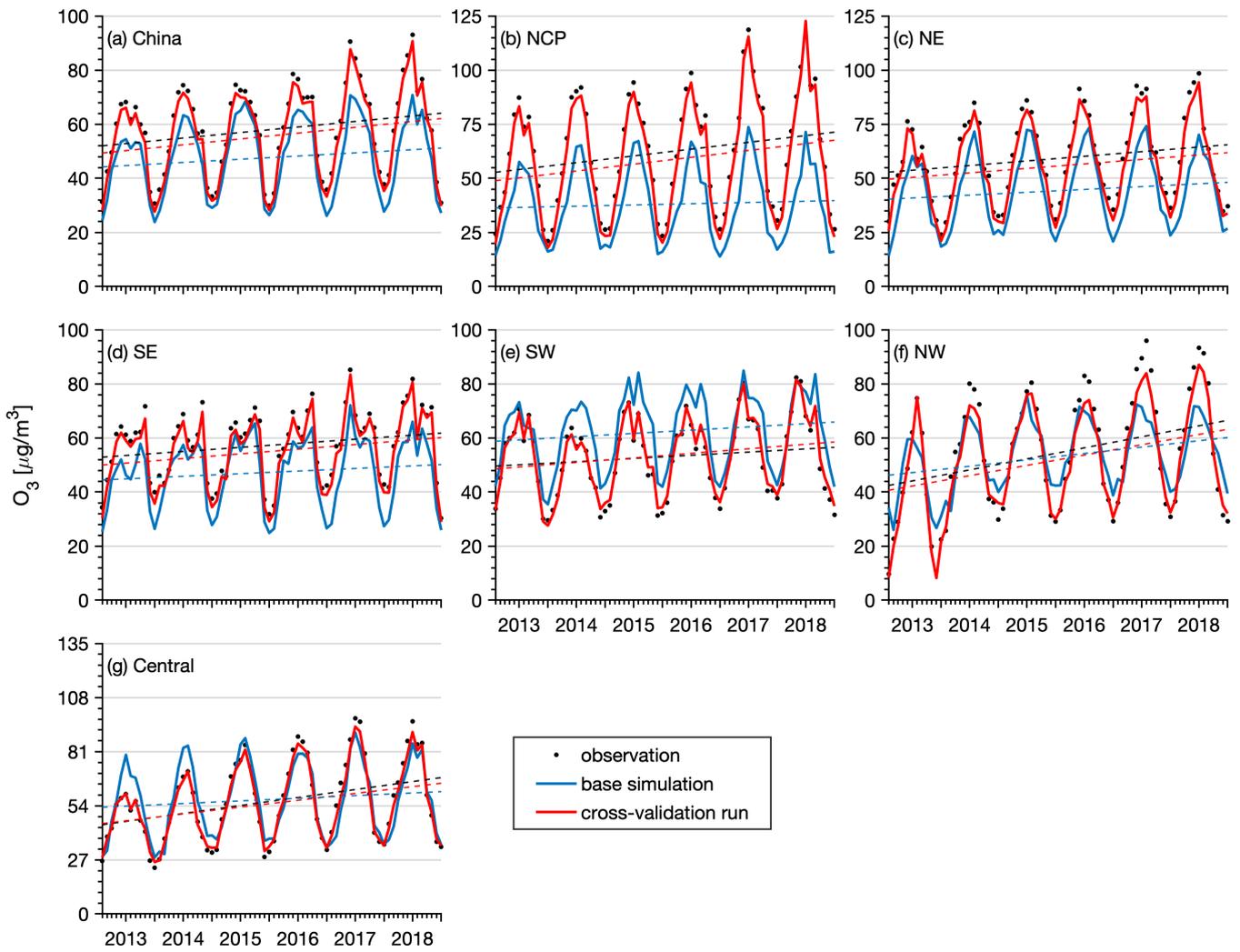
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960 **Figure 12: Same as Fig. 7 but for the NO₂ concentration.**



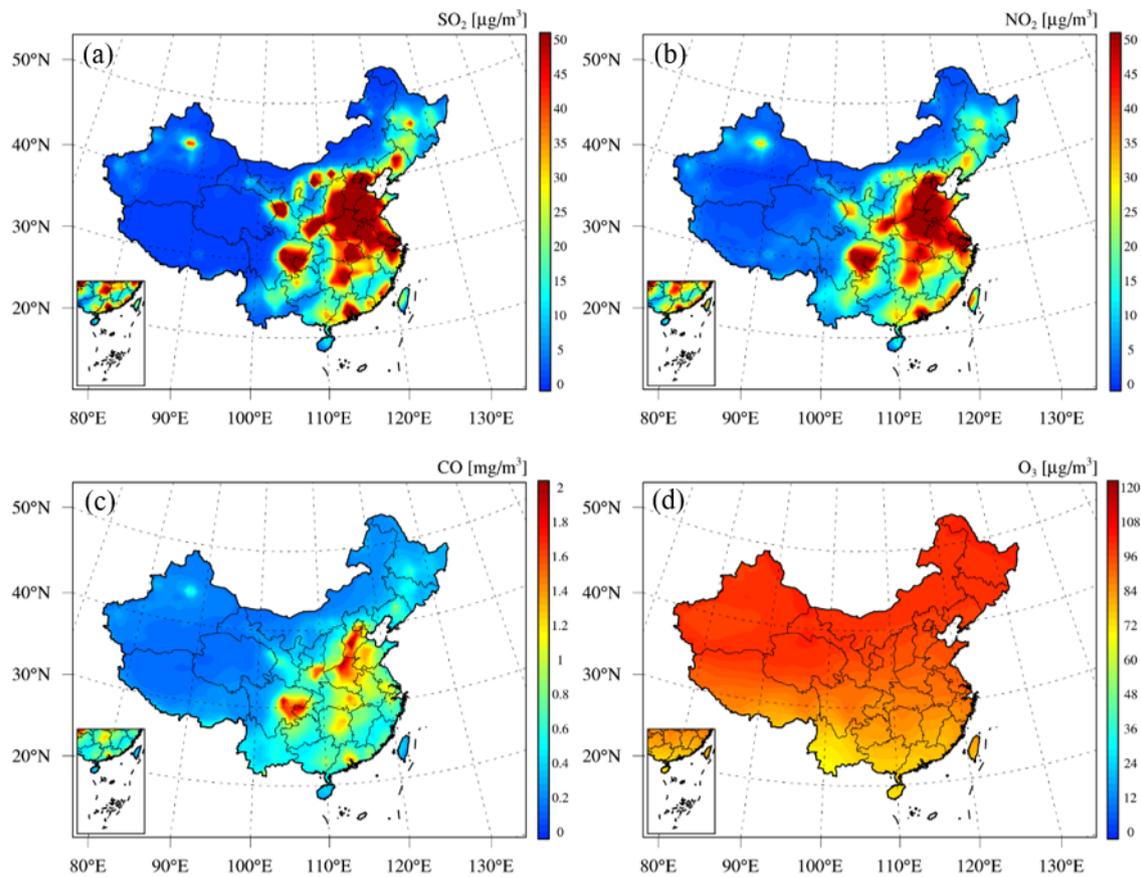
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962 **Figure 13: Same as Fig. 7 but for the CO concentration.**



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964 **Figure 14: Same as Fig. 7 but for the O₃ concentration.**



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966 **Figure 15: Spatial distributions of the multiyear average concentrations of (a) SO_2 , (b) NO_2 , (c) CO and (d) O_3 from**
 967 **2013 to 2018 obtained from CAMSRA.**

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