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Dear Editors:

Thank you very much for your kind decision letter on our paper entitled "**Improving $PM_{2.5}$ forecast over China by the joint adjustment of initial conditions and source emissions with an ensemble Kalman**" (acp-2016-732). We are grateful for the helpful comments from you and the reviewer. We have changed the manuscript according to his suggestions. We are looking forward to hearing from you soon.

Sincerely Yours,

Zhen Peng

Response to Reviewer #3's comments:

We thank Reviewer # 3 for his thoughtful comments and suggestions that have helped to improve this manuscript. Our responses to comments (in bold style) and the corresponding changes to the manuscript are detailed below.

I gave went through the manuscript and the authors made a decent effort to try to address the issues I raised in my comments. I still have one comment though and that is that the authors should be more honest with their results. I'm not saying they're hiding information or twisting it around but they should acknowledge that for some cases the assimilation just didn't improve things, I'm talking about the performance of the assimilation in JJJ. There is some improvement in the first 24 hrs but then there is no difference between the control run and the forecasts of both experiments. Considering they're focusing on the 48hr forecast this is not a minor issue. The authors do mention this in section 5.4 but then in the discussions and conclusions they state "Large improvements were achieved for almost all the 48-h forecasts, particularly in the YRD and PRD. However, relatively smaller improvements were achieved in the first 24-h forecast in the JJJ region, which …". They focus on the 48hrs in the analysis of the results and then switch to the 24 hr forecast in the conclusions because it did have some improvement. I can understand that the authors want to highlight the things that worked in their assimilation, especially after all the work they have done, which is really good. All I'm saying is that the authors should be honest with the results and if something doesn't work just say so, it doesn't make the work less valuable. Based on the results the authors provide, the assimilating simply doesn't work for the 48 hr forecast in some regions.

We realized that DA did work for some regions in certain periods although it cannot work perfectly for everywhere at all the time and it shows that there is no difference between the control run and the forecasts of both experiments for the second day forecast in JJJ. So I have replaced "relatively smaller improvements were achieved in the first 24-h forecast in the JJJ region" with "it did show some improvements in the first 24-h but then there is no difference between the control run

and the forecasts in the JJJ region afterwards" in Lines 764-766.

Also I have added this in the abstract in Line 28 and in section 5.4 in Lines 739.

What I also find interesting and the authors didn't address is that based on the analysis runs (table 1) there is not much difference in assimilating IC or IC+emissions (the authors themselves claim the statistics are the same, I would say they are even better for expC in some cases). Anyway, considering that and some of the results in figure 10 (in JJJ and YRD during the night), the improvement of assimilating emissions in addition to IC seems to be not so big or even negligible. I would have expected the authors say something about that as well. Is there really a benefit of assimilating both IC and emissions everywhere? my answer would be no, but the authors suggest that it is the case, at least imply it.

No. Actually the performances of assimilating emission are better in some regions.

From Figure 10 we can see that sometimes the performance of expJ was much better than expC in PRD and YRD. We addressed this in Lines 29-32, 722-725, 769-771.

Last thing, one argument used to explain the performance of the system there was the sparsity of the network, but in PRD the network is even sparser and the system performs well so I don't see the argument. The authors should better explain why in JJJ it is a problem but not in PRD.

In this work, our interested regions were JJJ (36-40 °N, 110-120 °E), YRD (27-35 °N, 116-122 °E) and PRD (22-24 °N, 112-115 °E). There were 12, 24 and 9 stations used for the $PM_{2.5}$ assimilation experiment in JJJ, YRD and PRD respectively. From figure 1 we can see that the measurement coverage in JJJ was much sparser than that in the YRD or PRD. So we think that the sparser coverage of measurements in JJJ may be one of the reasons.

Some specific comments (based on the version with the modifications highlighted):

Line 705: it says P25, i suppose it should be PM25

In this work, we used P_{25} (the unspectiated aerosol contributions) to calculate $\lambda^{f}_{PM2.5}$, so it was right in Line 325. We have no special reasons for this. It is also fine to use PM_{25} to calculate $\lambda^{f}_{PM2.5}$.

Line 748-749: What do they mean with "Since we did not know the exact station type,….", Do they mean urban or rural? please reformulate or clarify. In that same sentence the "We" after the , should be "we".

We have changed this sentence as "Since we did not know the exact observation environment of the sites, …." in Lines 367-368. Also we have replaced "We" with "we".

Lines 1061-1066: As far as I understand the reasons they are providing go in the different direction as what they try to justify. They are addressing the similar performance of the control run and both forecast experiments in YRD during the night. The claim that the prior emissions are larger than the optimized emission during the night and therefore the control run performs worse during night and better during day. What I see in figure 10 is the opposite, the control run performs better during the night than during day. The RMS and bias is smaller during the night than during the day. I would strongly suggest the authors to revise this part.

In this work, the 48-h forecasts were performed at each 0000 UTC from 6 to 16 October 2014 with the hourly forecast output for both assimilation experiments. So in Figure 10, daytime starts from 1~11 UTC and 25~35 UTC (corresponding to 9-19 Local Time). So it was right in the text and we added the forecasts time in Line 1233. Also we labeled the daytime in Figure 10.

Lines 1099-1102: I would ask the authors to reformulate these lines, I'm sure it can be improved.

We have changed these sentences in Lines 727-736.

Line 1104: replace “RRD” with "PRD"

We have replaced “RRD” with “PRD” in Lines 738.

Improving $PM_{2.5}$ forecast over China by the joint adjustment of initial conditions and source emissions with an ensemble Kalman filter

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Abstract. In an attempt to improve the forecasting of atmospheric aerosols, the ensemble square root filter algorithm was extended to simultaneously optimize the chemical initial conditions and emission input. The forecast model, which was expanded by combining the Weather Research and Forecasting with Chemistry (WRF-Chem) model and a forecast model of emission scaling factors, generated both chemical concentration fields and emission scaling factors. The forecast model of emission scaling factors was developed by using the ensemble concentration ratios of the WRF-Chem forecast chemical concentrations and also the time smoothing operator. Hourly surface fine particulate matter ($PM_{2.5}$) observations were assimilated in this system over China from 5 to 16 October 2014. A series of 48-h forecasts were then carried out with the optimized initial conditions and emissions on each day at 0000 UTC and a control experiment was performed without data assimilation. Besides, we also performed an experiment of pure assimilation chemical ICs and the corresponding 48-h forecasts experiment for comparison. The results showed that the forecasts with the optimized initial conditions and emissions typically outperformed those from the control experiment. In the Yangtze River delta (YRD) and the Pearl River delta (PRD) regions, large reduction of the Root Mean Square Errors (RMSEs) was obtained for almost the entire 48-h forecast range attributed to assimilation. Especially, the relative reduction in RMSE due to assimilation was about 37.5% at nighttime when WRF-Chem performed comparatively worse. In the Beijing–Tianjin–Hebei (JJJ) region, relatively smaller improvements were achieved in the first 24-h forecast but then no improvements were achieved afterwards. Comparing to the forecasts with only the optimized ICs, the forecasts with the joint adjustment were always much better during the night in the PRD and YRD regions. However, they

were very similar during daytime in both regions. Also, they performed similarly for almost the entire 48-h forecast range in the JJJ region.

1. Introduction

Aerosol prediction by regional air quality model in heavy polluted regions is challenging due to many factors. In addition to the deficiency of chemistries, the uncertainties of primary and precursor emissions and the initial conditions (ICs) also limit the forecast accuracy. Data assimilation (DA), which is used to improve the ICs of aerosols and to optimize data on aerosol emissions, has been shown to be one of the most effective ways to improve the forecasting of aerosol pollution.

From the perspective of reducing the uncertainties in the ICs for aerosols, recent efforts have focused on assimilating aerosol observations using optimal interpolation (Collins et al., 2001; Yu et al., 2003; Adhikary et al., 2008; Tombette et al., 2009; Lee et al., 2013) or variational (Kahnert, 2008; Zhang et al., 2008; Benedetti et al., 2009; Pagowski et al., 2010; Liu et al., 2011; Schwartz et al., 2012; Li et al., 2013; Jiang et al., 2013; Saide et al., 2013) DA algorithms. Ensemble-based DA algorithms, such as the ensemble Kalman filter (EnKF) (Sekiyama et al., 2010; Schutgens et al., 2010a, 2010b; Pagowski and Grell, 2012; Dai et al., 2014; Rubin et al., 2016; Ying, X.M., et al., 2016; Yumimoto et al., 2016) and the hybrid variational-ensemble DA approach (Schwartz et al., 2014) have also been applied to aerosol predictions. All these studies have shown that DA is one of the most effective ways of improving aerosol forecasting through assimilating aerosol observations from multiple sources (e.g. ground-based observations and satellite measurements) to update the chemical ICs.

Numerous studies have used DA approaches to estimate or improve source emissions. The EnKF is one of the most popular DA algorithms used to improve estimates of aerosols and gas-phase emissions, such as NO_x, volatile organic compounds, and SO_2 (van Loon et al., 2000; Heemink and Segers, 2002; Zhang et al., 2005; Barbu et al., 2009; Sekiyama et al., 2010; Huneeus et al., 2012; Schutgens et al., 2012; Huneeus et al., 2012, 2013; Miyazaki et al., 2014). Variational DA algorithms have also been applied to constrain emissions of air pollution, such as black carbon,

organic carbon, dust, NH_3, SO_x and NO_x (Hakami et al., 2005; Elbern et al., 2007; Henze et al., 2007, 2009; Yumimoto et al., 2007, 2008; Dubovik et al., 2008; Wang et al., 2012; Guerrette and Henze, 2015). These studies have indicated that DA can efficiently reduce the uncertainty in the emission inventories and lead to improvements in the forecasting of air quality (Mijling and van der A, 2012).

The optimization of chemical ICs and pollution emissions can improve aerosol forecasts and therefore further improvements are likely to be achieved by simultaneously optimizing the chemical ICs and emissions. Tang et al. (2011) reported that the simultaneous adjustment of the ICs of O_3, NO_x and volatile organic compounds and the emissions of NO_x and volatile organic compounds produced overall better performance in both the 1-h and 24-h ozone forecasts than the adjustment of pure ICs or emissions. Miyazaki et al. (2012) reported that the simultaneous adjustment of emissions and concentrations is a powerful approach to correcting the tropospheric ozone budget and profile analyses.

We developed a system to adjust the chemical ICs and source emissions jointly within an EnKF system coupled to the Weather Research and Forecasting with Chemistry (WRF-Chem) model (Grell et al., 2005). We then applied this system to assimilate hourly surface $PM_{2.5}$ measurements over China in early October 2014.

The remainder of the paper is organized as follows. Section 2 describes this DA system in detail and Section 3 describes the $PM_{2.5}$ observations. Then the experimental designs are introduced in Section 4. Finally, the surface $PM_{2.5}$ observations assimilation results are presented in section 5 before concluding in section 6.

2. Methodology

2.1 Forecast model

For a chemical model like WRF-Chem, the emissions are the model forcing (or boundary condition), rather than model states. Therefore, a forecasting model, **M**, was developed to forecast the emission scaling factors (representing emissions) as well as the aerosol concentrations. This model combines the WRF-Chem model and

the forecast model of emission scaling factors.

2.1.1 WRF-Chem model

Version 3.6.1 of the WRF-Chem model (Grell et al., 2005) was used to forecast the aerosol and chemical species. WRF-Chem is an online model with the fully coupled chemical and meteorological components.

Most of the WRF-Chem settings were the same as those reported in Liu et al. (2011): the Goddard Chemistry Aerosol Radiation and Transport (GOCART) aerosol scheme coupled with the Regional Atmospheric Chemistry Mechanism for gaseous chemical mechanisms; the WRF single-moment five-class microphysics scheme; the Rapid Radiative Transfer Model longwave and Goddard shortwave radiation schemes; the Yonsei University (YSU) boundary layer scheme; the Noah land surface model; and the Grell-3D cumulus parameterization. For the GOCART aerosol scheme, the aerosol species include 14 defined aerosol species and a 15^{th} variable representing unspectiated aerosol contributions (P_{25}). The 14 defined aerosol species are sulfate, hydrophobic and hydrophilic organic carbon (OC_1 and OC_2, respectively), hydrophobic and hydrophilic black carbon (BC_1 and BC_2, respectively), dust in five particle size bins (effective radii of 0.5, 1.4, 2.4, 4.5 and 8.0 μm; referred to as D_1, D_2, D_3, D_4 and D_5, respectively) and sea salt in four particle size bins (effective radii of 0.3, 1.0, 3.25 and 7.5 μm for dry air; referred to as S_1, S_2, S_3 and S_4, respectively).

Figure 1 illustrates the model computational domain. It has 120*120 horizontal grid scales at a 40.5 km spacing by the lambert conform map projection centered at (35°N, 105°E). There are 57 vertical levels with the model top at 10 hPa, about 12 layers within the planetary boundary layer (among them the lowest 8 layers were under 500 m), and the first layer centered at ~12 m.

With respect to the emissions, the hourly prior anthropogenic emissions were based on the monthly regional emission inventory in Asia (Zhang et al., 2009) for the year 2006 interpolated to the model grid. The power generator emissions were interpolated for the lowest eight vertical levels (Woo et al., 2003; de meij et al., 2006;

Wang et al., 2010). Other anthropogenic emissions were assigned totally to the 1st level. Emissions are very small above 500 m for all pollutants. In order to keep objective for the prior anthropogenic emissions, no time variation was added. Thus, the hourly prior anthropogenic emissions were constant. The biogenic (Guenther et al., 1995), dust (Ginoux et al., 2001), dimethylsulfide and sea salt emissions (Chin et al., 2000, 2002) were calculated online.

2.1.2 Forecast model of scaling factors

As no suitable dynamic model was available to forecast the emission scaling factors, a persistence forecasting operator served as the forecast model for the scaling factors, similar to the method used by Peng et al. (2015) for CO_2 emission inversion. Figure 2a shows the flowchart for the persistence forecasting operator $\mathbf{M}_{\mathrm{SF}}$.

If the ensemble members of the updated chemical fields $\mathbf{C}_{i,t-1}^{\mathrm{a}}$ (the subscript i refers to the ith ensemble member, the superscript a refers to the analysis, and t refers to the time) and the forecast emissions $\mathbf{E}_{i,t-1}^{\mathrm{f}}$ (the superscript f refers to the forecast) in the previous assimilation cycle are known, then the chemical fields $\mathbf{C}_{i,t}^{\mathrm{f}}$ at time t can be generated via WRF-Chem (Figure 2b). In the actual process, $\mathbf{C}_{i,t}^{\mathrm{f}}$ were available in the previous assimilation cycle, so we did not need to perform the ensemble forecasts again. A dotted box was used in Figure 2a to indicate that the ensemble forecasts were not performed in real process. The ensemble concentration ratios $\boldsymbol{\kappa}_{i,t}, (i = 1, \dots, N)$ are then calculated using

$$\boldsymbol{\kappa}_{i,t} = \frac{\mathbf{C}_{i,t}^{\mathrm{f}}}{\overline{\mathbf{C}_{t}^{\mathrm{f}}}}, (i = 1, \dots, N), \quad (1)$$

where $\overline{\mathbf{C}_{t}^{\mathrm{f}}} = \frac{1}{N}\sum_{i=1}^{N} \mathbf{C}_{i,t}^{\mathrm{f}}$ is the ensemble mean of the forecast. The ensemble mean of $\boldsymbol{\kappa}_{i,t}$ is,

$$\overline{\boldsymbol{\kappa}_{t}} = \frac{1}{N}\sum_{i=1}^{N} \boldsymbol{\kappa}_{i,t} = \frac{1}{N}\sum_{i=1}^{N} \mathbf{C}_{i,t}^{\mathrm{f}} / \overline{\mathbf{C}_{t}^{\mathrm{f}}} = 1, \quad (2)$$

so $\boldsymbol{\kappa}_{i,t}$ are numbers distributed around 1 and with ensemble mean values of 1.

The ensemble spreads of $\boldsymbol{\kappa}_{i,t}, (i = 1, \dots, N)$ may be small and therefore

263 covariance inflation is used to maintain them at a certain level:

264 $$(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}} = \beta\left(\boldsymbol{\kappa}_{i,t} - \overline{\boldsymbol{\kappa}_t}\right) + \overline{\boldsymbol{\kappa}_t}, (i = 1, \dots, N)\ , (3)$$

265 In Peng et al. (2015), the CO_2 DA system worked comparatively well when the
266 ensemble spread of $\lambda^{\mathrm{a}}_{\mathrm{i,t}}$ ranged from 0.05 to 1.25 for β =60, 70, 75, 80. The
267 assimilated CO2 fluxes deviated markedly from the "true" CO2 fluxes when the
268 ensemble spread of $\lambda^{\mathrm{a}}_{\mathrm{i,t}}$ were too small for β =10, 50 or when the ensemble spread
269 of $\lambda^{\mathrm{a}}_{\mathrm{i,t}}$ were too large for β =100. Therefore, in this work, $\beta = 1.5$ was chosen to
270 make ensure the ensemble spread of $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ ranged from 0.1 to 1.25. Same as $\boldsymbol{\kappa}_{i,t}$,
271 the ensemble mean values of $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ are 1. It is noted that perhaps there are very
272 few negative values for $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ after inflation. A quality control procedure is
273 performed for $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ before further appliance. All these negative data were set as
274 0.001 in this work. There was no special reason to set them as 0.001. It is also fine to
275 set them as 0. Then $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ were re-centered to ensure the ensemble mean values of
276 $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ were all 1.

277 As the concentrations were closely related to the emissions both locally and in
278 the upwind regions and there is no suitable dynamic model available to forecast the
279 emission scaling factors, the inflated concentration ratios $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ serve as the prior
280 emission scaling factors $\boldsymbol{\lambda}^{\mathrm{p}}_{i,t}$:

281 $$\boldsymbol{\lambda}^{\mathrm{p}}_{i,t} = (\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}, (i = 1, \dots, N), (4)$$

282 The above equation is not supported according to the mass conservation equation
283 but just for the purpose to generate the ensemble emissions. Same as $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$, $\boldsymbol{\lambda}^{\mathrm{p}}_{i,t}$
284 are numbers distributed around 1. From the perspective of generating the ensemble
285 emissions, they can play the same role as other data, such as the random numbers
286 created by using the standard normal distribution function. However, there are
287 correlations among the grid-points of $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ because $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$ are calculated
288 through a short-term forecast of WRF-Chem. Thus, $\boldsymbol{\lambda}^{\mathrm{p}}_{i,t}$ have the same correlations
289 as $(\boldsymbol{\kappa}_{i,t})_{\mathrm{inf}}$. While, the random numbers are totally different. There are no

correlations unless they are generated under certain correlations.

To incorporate the useful information from the previous times, the previous DA cycles' analysis scaling factors, $\boldsymbol{\lambda}_{i,t-M+1}^{\mathrm{a}}$, ⋯, $\boldsymbol{\lambda}_{i,t-2}^{\mathrm{a}}$, $\boldsymbol{\lambda}_{i,t-1}^{\mathrm{a}}$ and the prior scaling factor $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$ were used to estimate $\boldsymbol{\lambda}_{i,t}^{\mathrm{f}}$ by the time smooth operator; namely,

$$\boldsymbol{\lambda}_{i,t}^{\mathrm{f}} = \frac{1}{M}\left(\sum_{j=t-M+1}^{t-1} \boldsymbol{\lambda}_{i,j}^{\mathrm{a}} + \boldsymbol{\lambda}_{i,t}^{\mathrm{p}}\right), (i = 1, \dots, N, j = t - M + 1, \dots, t - 1), (5)$$

Here, M is the time window of the smooth operator. In this study, a value of $M = 4$ (hours) was chosen. According to the smooth operator, the ensemble mean values of $\boldsymbol{\lambda}_{i,t}^{\mathrm{f}}$ depend on the ensemble mean of $\boldsymbol{\lambda}_{i,t-M+1}^{\mathrm{a}}$, ⋯, $\boldsymbol{\lambda}_{i,t-2}^{\mathrm{a}}$, $\boldsymbol{\lambda}_{i,t-1}^{\mathrm{a}}$, $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$, where the ensemble means of $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$ are all 1. After multiple iterations, the smooth operator can give comparatively good estimation for $\boldsymbol{\lambda}_{i,t}^{\mathrm{f}}$ since anthropogenic emissions are stable at a certain time scale (Mijling et al., 2012). It is a compromise between prescribed prior emissions and letting the system propagate all observation information from one step to the next without any guidance (Peters et al., 2007), for the case $M = 4$.

The ensemble members of the emissions were calculated according to

$$\mathbf{E}_{i,t} = \boldsymbol{\lambda}_{i,t}\mathbf{E}_t^{\mathrm{p}}, (i = 1, \dots, N), (6)$$

where $\mathbf{E}_{i,t}$ is the ith ensemble member of the emissions for each grid at time t, $\boldsymbol{\lambda}_{i,t}$ represents the scaling factors and $\mathbf{E}_t^{\mathrm{p}}$ is the prescribed emission, which can be obtained from the emission inventories. It is noted that the correlations among the grid-points of the prior emissions depend on $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$. These correlations may deviate far from the truth but we have no other suitable substitute. However, the correlations among the grid-points of the forecast emissions should be more or less close to the truth due to the appliance of the smooth operator after multiple iterations.

It is noted although the method is very similar to that used by Peters et al. (2007) and Peng et al. (2015) for CO_2 emission inversion, it is still of novelty for applications in aerosol anthropogenic emissions. In Peters et al. (2007), $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$ were all 1. And only natural CO_2 emissions (i.e., biospheric and oceanic emissions) were assimilated at the ecological scale due to the 'signal-to-noise' problem. Thus, the uncertainty of

anthropogenic and other CO_2 emissions were ignored. Besides, the framework is more advanced compared to our previous work. In Peng et al. (2015), in order to generate $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$, a set of ensemble forecasts were performed from time *t* to *t*+1 to produce the CO_2 concentration fields, forced by the prescribed net CO_2 surface fluxes with the previous assimilated concentration fields as initial conditions. That means that the ensemble forecast were performed twice in that DA system and it was time consuming. However, in order to save computing time, we used the chemical fields $\mathbf{C}_{i,t}^{\mathrm{f}}$ available in the previous assimilation cycle to calculate $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$ in this work. Thus, WRF-Chem runs to forecast only once during a DA cycle.

2.2 Ensemble square root filter

The ensemble square root filter (EnSRF) algorithm was introduced by Whitaker and Hamill (2002) and its expansion to analyzing aerosol ICs was described by Schwartz et al. (2014). The traditional EnKF with perturbed observations (Evensen 1994) introduces sampling errors by perturbing the observations. In contrast to the traditional EnKF, the EnSRF (Whitaker and Hamill, 2002) and the Ensemble Adjustment Kalman Filter (EAKF, developed by Anderson, 2001) obviate the need to perturb the observations. The local ensemble Kalman filtering (LEKF), a kind of EnSRF, was presented by Ott et al. (2002, 2004). It was computationally more efficient compared to the traditional EnKF, since it simultaneously assimilates the observations within a spatially local volume independently. The local Ensemble Transform Kalman Filter (LETKF, Hunt, 2007) integrates the advantages of the Ensemble Transform Kalman Filter (ETKF, developed by Bishop et al., 2001) and the LEKF. The computational cost of LETKF is much lower than that of the original LEKF because the former does not require an orthogonal basis. Though LETKF has more advantages, we still chose the same EnSRF as Schwartz et al. (2014) because we did not need to extend it to analyzing aerosol ICs, very similar to Schwartz et al. (2014).

Following the notation of Ide et al. (1997), given an *m*-dimensional background

model forecast vector $\mathbf{x}^{\mathrm{b}}$, a p-dimensional observation vector $\mathbf{y}^{\mathrm{o}}$ and an operator $\mathbf{H}$ that converts the model state to the observation states, we expressed the variables as an ensemble mean (denoted by an over-bar) and a deviation from the mean (denoted by a prime). Thus, the ensemble mean $\bar{\mathbf{x}}^{\mathrm{a}}$ of the analyzed state $\mathbf{x}^{\mathrm{a}}$ and the deviations $\mathbf{x}'^{\mathrm{a}}$ from the ensemble mean are updated separately by

$$\bar{\mathbf{x}}^{\mathrm{a}} = \bar{\mathbf{x}}^{\mathrm{b}} + \mathbf{K}(\mathbf{y}^{\mathrm{o}} - \mathbf{H}\bar{\mathbf{x}}^{\mathrm{b}}), \quad (7)$$

$$\mathbf{x}'^{\mathrm{a}} = \mathbf{x}'^{\mathrm{b}} + \widetilde{\mathbf{K}}(\mathbf{y}'^{\mathrm{o}} - \mathbf{H}\mathbf{x}'^{\mathrm{b}}), \quad (8)$$

where $\mathbf{K}$ is the traditional Kalman gain matrix and $\widetilde{\mathbf{K}}$ is the gain used to update the deviations from the ensemble mean. These are given by

$$\mathbf{K} = \mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}}(\mathbf{H}\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}} + \mathbf{R})^{-1}, \quad (9)$$

$$\widetilde{\mathbf{K}} = \mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}}\left[\left(\sqrt{\mathbf{H}\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}} + \mathbf{R}}\right)^{-1}\right]^{\mathrm{T}}\left(\sqrt{\mathbf{H}\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}} + \mathbf{R}} + \sqrt{\mathbf{R}}\right)^{-1}$$

$$= \left(\mathbf{1} + \sqrt{\mathbf{R}/(\mathbf{H}\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}} + \mathbf{R})}\right)^{-1}\mathbf{K}, \quad (10)$$

where $\mathbf{P}^{\mathrm{b}} = \frac{1}{N-1}\sum_{i=1}^{N}\mathbf{x}'^{\mathrm{b}}(\mathbf{x}'^{\mathrm{b}})^{T}$ is the $m * m$ -dimensional background error covariance matrix and $\mathbf{R}$ is the $p * p$-dimensional diagonal observation error covariance matrix. In real applications, $\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}}$and $\mathbf{H}\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}}$ will be approximated using the background ensemble; namely,

$$\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}} = \frac{1}{N-1}\sum_{i=1}^{N}\mathbf{x}'^{\mathrm{b}}(\mathbf{H}\mathbf{x}'^{\mathrm{b}})^{T} \quad (11)$$

$$\mathbf{H}\mathbf{P}^{\mathrm{b}}\mathbf{H}^{\mathrm{T}} = \frac{1}{N-1}\sum_{i=1}^{N}\mathbf{H}\mathbf{x}'^{\mathrm{b}}(\mathbf{H}\mathbf{x}'^{\mathrm{b}})^{T}. \quad (12)$$

In equations (11) and (12), N is the ensemble size.

Note that for the joint analysis of ICs and emissions, the state vector $\mathbf{x}$ is the joint vector of the mass concentration $\mathbf{C}$ and the emission scaling factor $\boldsymbol{\lambda}$, i.e. $\mathbf{x} = [\mathbf{C}, \boldsymbol{\lambda}]^{\mathrm{T}}$. In this study, the state variables of the analysis of the ICs were the 15 WRF-Chem/GOCART aerosol variables, same as that reported by Schwartz et al. (2012). The state variables of the emission scaling factors include $\boldsymbol{\lambda}_{\mathrm{PM2.5}}$, $\boldsymbol{\lambda}_{\mathrm{SO2}}$, $\boldsymbol{\lambda}_{\mathrm{NO}}$ and $\boldsymbol{\lambda}_{\mathrm{NH3}}$ and are described in section 2.3.1. After each ensemble analysis, the ensemble forecasts were performed with the corresponding models to advance $\mathbf{C}$ and $\boldsymbol{\lambda}$ to the next analysis time.

In this work, a 50-member ensemble was chosen, following Schwartz et al.

(2012) and Whitaker and Hamill (2002). Covariance localization forced EnSRF analysis increments to zero 1280 km from an observation in the horizontal and one scale height to reduce spurious correlations due to sampling error for all control variables, similar to Pagowski et al., (2012) and Schwartz et al., (2012, 2014). In addition, posterior (after assimilation) multiplicative inflation following Whitaker and Hamill (2012) was applied aiming to maintain ensemble spread for only the concentration analysis. The inflation factor $\alpha = 1.2$ was chosen as Pagowski et al., (2012) and Schwartz et al., (2012, 2014). Additive or prior inflation was not employed. As for the emission scaling factor $\boldsymbol{\lambda}$, the inflation was not used at this step.

2.3 Data assimilation system

2.3.1 State variables

As stated in section. 2.2, the state variables of the analysis of the ICs were the 15 WRF-Chem/GOCART aerosol variables. The $PM_{2.5}$ observation operator was the same as that described by Schwartz et al. (2012) and expressed as

$$\mathbf{y}^{\mathrm{f}} = \boldsymbol{\rho}_{\mathrm{d}}[\mathbf{P_{25}} + 1.375\mathbf{S} + 1.8(\mathbf{OC_1} + \mathbf{OC_2}) + \mathbf{BC_1} + \mathbf{BC_2} + \mathbf{D_1} + 0.286\mathbf{D_2} + \mathbf{S_1} + 0.942\mathbf{S_2}], \quad (13)$$

where $\boldsymbol{\rho}_{\mathrm{d}}$ represents the dry air density, which is multiplied by the mixing ratios of aerosol species (in $\mu g \cdot kg^{-1}$) to convert the units to $\mu g\ m^{-3}$ for consistency with the observations.

From the perspective of the optimization of emissions, four species of emission scaling factors ($\boldsymbol{\lambda}_{\mathrm{PM2.5}}$, $\boldsymbol{\lambda}_{\mathrm{SO2}}$, $\boldsymbol{\lambda}_{\mathrm{NO}}$ and $\boldsymbol{\lambda}_{\mathrm{NH3}}$) were also considered as the state variables of the DA system. Atmospheric inorganic aerosols are not only from the primary emissions, but also from secondary processes- chemical and thermodynamic transformations from the gas-phase precursors. Therefore, not only the primary sources of $PM_{2.5}$, but also the sources of the gas-phase precursors, need to be optimized. In this study, the sources of SO_2, NO_x and NH_3 ($\mathbf{E}_{\mathrm{SO2}}$, $\mathbf{E}_{\mathrm{NO}}$ and $\mathbf{E}_{\mathrm{NH3}}$), which have a large impact on the distribution of $PM_{2.5}$, were also optimized in addition to the primary sources of $PM_{2.5}$. It is noted that for the optimization of the emission scaling factors, $\mathbf{M}_{\mathrm{SF}}$ serves as the forecast model and the observation

operator reflects the combined information of emissions (in the format of $\boldsymbol{\lambda}$ in equation (6)), the physics and chemistry processes in WRF-Chem simulations and the transformation $PM_{2.5}$ from model space to observation space (equation (13)).

The direct sources of $PM_{2.5}$ include the unspeciated primary sources of $PM_{2.5}$ $\mathbf{E}_{\mathrm{PM2.5}}$, sulfate $\mathbf{E}_{\mathrm{SO4}}$, nitrate $\mathbf{E}_{\mathrm{NO3}}$, organic compounds E_{org} and elemental compounds E_{BC}; all of them are given in two modes (the nuclei and accumulation modes, represented as i and j in the subscripts respectively). The ratios between the nuclei and accumulation modes were the same as in the suggested emission process for National Emission Inventory in WRF-Chem (Freitas et al., 2011). The formula of sulfate and nitrate emissions in the model are as below:

$$\mathbf{E}_{\mathrm{PM2.5i}}:\mathbf{E}_{\mathrm{PM2.5j}} = 1:4, \quad (14)$$

$$\mathbf{E}_{\mathrm{SO4i}}:\mathbf{E}_{\mathrm{SO4j}} = 1:4, \quad (15)$$

$$\mathbf{E}_{\mathrm{NO3i}}:\mathbf{E}_{\mathrm{NO3j}} = 1:4, \quad (16)$$

$$\mathbf{E}_{\mathrm{SO4i}} + \mathbf{E}_{\mathrm{SO4j}} = a * (\mathbf{E}_{\mathrm{PM2.5i}} + \mathbf{E}_{\mathrm{PM2.5j}} - \mathbf{E}_{\mathrm{EC}} - \mathbf{E}_{\mathrm{ORG}}), \quad (17)$$

$$\mathbf{E}_{\mathrm{NO3i}} + \mathbf{E}_{\mathrm{NO3j}} = b * \left(\mathbf{E}_{\mathrm{PM2.5i}} + \mathbf{E}_{\mathrm{PM2.5j}} - \mathbf{E}_{\mathrm{EC}} - \mathbf{E}_{\mathrm{ORG}}\right), \quad (18)$$

where $\mathbf{E}_{\mathrm{EC}}$ represents elemental carbon and $\mathbf{E}_{\mathrm{ORG}}$ organic compounds, and $a = 0.074$ and $b = 0.038$ were chosen based on the internal emissions and observational data. In the DA process, the first 6 species of direct sources of emissions ($\mathbf{E}_{\mathrm{PM2.5i}}$, $\mathbf{E}_{\mathrm{PM2.5j}}$, $\mathbf{E}_{\mathrm{SO4i}}$, $\mathbf{E}_{\mathrm{SO4j}}$, $\mathbf{E}_{\mathrm{NO3i}}$, and $\mathbf{E}_{\mathrm{NO3j}}$), which may have larger uncertainties in heavy polluted events, were updated according to the variation of $\boldsymbol{\lambda}_{\mathrm{PM2.5}}$. $\mathbf{E}_{\mathrm{PM2.5i}}$ and $\mathbf{E}_{\mathrm{PM2.5j}}$ were directly updated according to the variation in $\boldsymbol{\lambda}_{\mathrm{PM2.5}}$. The emissions ($\mathbf{E}_{\mathrm{SO4i}}$, $\mathbf{E}_{\mathrm{SO4j}}$, $\mathbf{E}_{\mathrm{NO3i}}$ and $\mathbf{E}_{\mathrm{NO3j}}$) were also updated according to the variations in $\mathbf{E}_{\mathrm{PM2.5i}}$ and $\mathbf{E}_{\mathrm{PM2.5j}}$.

$\mathbf{E}_{\mathrm{EC}}$ and $\mathbf{E}_{\mathrm{ORG}}$ of the anthropogenic emissions were not assimilated, which is a limitation in this work. Besides, emissions of dust and sea salt were not assimilated. It is true that these emissions are also important for the atmosphere aerosol. The reason we did not assimilate $\mathbf{E}_{\mathrm{EC}}$ and $\mathbf{E}_{\mathrm{ORG}}$ is that only the $PM_{2.5}$ measurements are used in this DA experiment. However, the sources of the aerosols (especially organic aerosols)

are so complex that our knowledge of their formation mechanisms is far from clear. Though it is technically possible to have all emissions assimilated, with such limited observations adding more control variables would cause much more uncertainties in the system which might lead to unreasonable analysis.

2.3.2 Procedure for the DA system

Figure 2 (b) shows the workflow of the DA system. The steps in this workflow are as follows.

(1) The persistence forecasting operator $\mathbf{M}_{\mathrm{SF}}$ is applied to forecast the background fields of the emission scaling factors $\boldsymbol{\lambda}^{\mathrm{f}}_{\mathrm{PM2.5}}$, $\boldsymbol{\lambda}^{\mathrm{f}}_{\mathrm{SO2}}$, $\boldsymbol{\lambda}^{\mathrm{f}}_{\mathrm{NO}}$ and $\boldsymbol{\lambda}^{\mathrm{f}}_{\mathrm{NH3}}$. The forecast chemical fields of P_{25}, SO_2, NO and NH_3 of the previous assimilation cycle are used to create the prior emission scaling factors $\boldsymbol{\lambda}^{\mathrm{p}}_{\mathrm{PM2.5}}$, $\boldsymbol{\lambda}^{\mathrm{p}}_{\mathrm{SO2}}$, $\boldsymbol{\lambda}^{\mathrm{p}}_{\mathrm{NO}}$ and $\boldsymbol{\lambda}^{\mathrm{p}}_{\mathrm{NH3}}$. The background scaling factors are then generated using equation (5).

(2) The ensemble members of the emissions, $\mathbf{E}^{\mathrm{f}}_{\mathrm{PM2.5i}}$, $\mathbf{E}^{\mathrm{f}}_{\mathrm{PM2.5j}}$, $\mathbf{E}^{\mathrm{f}}_{\mathrm{SO2}}$, $\mathbf{E}^{\mathrm{f}}_{\mathrm{NO}}$ and $\mathbf{E}^{\mathrm{f}}_{\mathrm{NH3}}$, are prepared according to equation (6). The corresponding emissions of $\mathbf{E}^{\mathrm{f}}_{\mathrm{SO4i}}$, $\mathbf{E}^{\mathrm{f}}_{\mathrm{SO4j}}$, $\mathbf{E}^{\mathrm{f}}_{\mathrm{NO3i}}$ and $\mathbf{E}^{\mathrm{f}}_{\mathrm{NO3j}}$ are obtained based on equations (15–18). Other inorganic species of the anthropogenic emission, such as $\mathbf{E}_{\mathrm{EC}}$ and $\mathbf{E}_{\mathrm{ORG}}$, are not perturbed for WRF-Chem. However, other anthropogenic emissions, such as $\mathbf{E}_{\mathrm{PM2.5}}$, $\mathbf{E}_{\mathrm{SO4}}$ and $\mathbf{E}_{\mathrm{NO3}}$, are much larger than $\mathbf{E}_{\mathrm{EC}}$ and $\mathbf{E}_{\mathrm{ORG}}$ in most area of China, and the ensemble spreads of the aerosol concentrate largely dependent on the uncertainties of those anthropogenic emissions. Besides, model errors raised from the meteorology, the emission and the chemical model itself are compensated to some extent through the use of multiplicative inflation. In other words, the ensemble spread of the concentrations can be kept at a certain level though $\mathbf{E}_{\mathrm{EC}}$ and $\mathbf{E}_{\mathrm{ORG}}$, are not perturbed.

Natural emissions, such as dust and sea salt emissions were not perturbed explicitly when the forecast emissions were generated. However, emissions of dust and sea salt were parameterized within the GOCART model (Chin et al., 2002). Within the DA system, varying meteorology across the members implicitly perturbed

dust and sea salt emissions.

(3) Forced by the changed emissions ($\mathbf{E}_{PM2.5i}$, $\mathbf{E}_{PM2.5j}$, $\mathbf{E}_{SO2}$, $\mathbf{E}_{NO}$, $\mathbf{E}_{NH3}$, $\mathbf{E}_{SO4i}$, $\mathbf{E}_{SO4j}$, $\mathbf{E}_{NO3i}$ and $\mathbf{E}_{NO3j}$ were substituted by $\mathbf{E}^{f}_{PM2.5i}$, $\mathbf{E}^{f}_{PM2.5j}$, $\mathbf{E}^{f}_{SO2}$, $\mathbf{E}^{f}_{NO}$, $\mathbf{E}^{f}_{NH3}$, $\mathbf{E}^{f}_{SO4i}$, $\mathbf{E}^{f}_{SO4j}$, $\mathbf{E}^{f}_{NO3i}$ and $\mathbf{E}^{f}_{NO3j}$; the other emissions such as $\mathbf{E}_{EC}$ and $\mathbf{E}_{ORG}$ remained unchanged), WRF-Chem is run again to forecast the chemical fields $\boldsymbol{\rho}^{f}$ with the updated chemical fields of the previous assimilation cycle as the ICs. The state variables, i.e., 15 aerosol species and four scaling factors, are then prepared.

(4) The model-simulated $PM_{2.5}$ concentration at the observation space is then calculated via equation (13). At this time, the state vector $\mathbf{x}^{f} = [\mathbf{C}^{f}, \boldsymbol{\lambda}^{f}]^{T}$ was prepared.

(5) In the assimilation step, the state variables, the concentrations of 14 defined aerosol species and a 15th unspeciated aerosol, and the four species of emission scaling factors $\boldsymbol{\lambda}^{f}_{PM2.5}$, $\boldsymbol{\lambda}^{f}_{SO2}$, $\boldsymbol{\lambda}^{f}_{NO}$ and $\boldsymbol{\lambda}^{f}_{NH3}$, were optimized through EnSRF.

(6) After the assimilation step, the optimized emissions ($\mathbf{E}^{a}_{PM2.5i}$, $\mathbf{E}^{a}_{PM2.5j}$, $\mathbf{E}^{a}_{SO2}$, $\mathbf{E}^{a}_{NO}$, $\mathbf{E}^{a}_{NH3}$, $\mathbf{E}^{a}_{SO4i}$, $\mathbf{E}^{a}_{SO4j}$, $\mathbf{E}^{a}_{NO3i}$ and $\mathbf{E}^{a}_{NO3j}$) were calculated according to equations (6, 15–18) using the optimized scaling factors ($\boldsymbol{\lambda}^{a}_{PM2.5}$, $\boldsymbol{\lambda}^{a}_{SO2}$, $\boldsymbol{\lambda}^{a}_{NO}$ and $\boldsymbol{\lambda}^{a}_{NH3}$).

3. $PM_{2.5}$ observation data and errors

Hourly averaged surface $PM_{2.5}$ observations from the Ministry of Environmental Protection of China were assimilated. There were altogether 876 national control measurement sites over China. The $PM_{2.5}$ observation sites spanned most of central and eastern China but were primarily located in urban and suburban areas. So it always happened that there were more than one observation sites in certain city, which were fall into the same model grid. Since we did not know the exact observation environment of the sites, we randomly selected one observation site in a city for assimilation experiment and one for verification purposes to ensure that there was at most one assimilated measurements for one model grid. Altogether 77 stations were selected for the $PM_{2.5}$ assimilation experiment and another 77 independent

stations were selected for verification. Figure 1 shows the locations of 77 measurement sites used for the $PM_{2.5}$ assimilation experiment and 77 independent sites used for forecast verification.

The observation error covariance matrix **R** in equation (9) includes contributions from measurement and representation errors. Similar to the work of Schwartz et al. (2012), the measurement error ε_0 is defined as $\varepsilon_0 = 1.5 + 0.0075 * \Pi_0$, where Π_0 denotes the observational values for $PM_{2.5}$ ($\mu g\ m^{-3}$). Thus, higher $PM_{2.5}$ values were associated with larger measurement errors. Following Elbern et al. (2007) and Pagowski et al. (2010), Schwartz et al. (2012), the representativeness error ε_r depends on the resolution of the model and the characteristics of the observation locations and is calculated as $\varepsilon_r = r\varepsilon_0\sqrt{\Delta x/\mathrm{L}}$, where r is an adjustable parameter (here, $r = 0.5$), Δx is the grid spacing (here, 40.5 km), and L is the radius of influence of an observation (here, L was set to 3 km following Elbern et al. (2007), since we do not know the station type that used in this work). The total $PM_{2.5}$ error (ε_t) is defined as $\varepsilon_t = \sqrt{{\varepsilon_0}^2 + {\varepsilon_r}^2}$. The observation errors are assumed to be uncorrelated so that **R** is a diagonal matrix.

The $PM_{2.5}$ observations were subject to quality control to ensure data reliability before DA. Considering that China has had intense pollution events, $PM_{2.5}$ values larger than 800 $\mu g\ m^{-3}$ were classified as unrealistic and were not assimilated; observations with the ensemble mean of the first guess departure exceeding 100 $\mu g\ m^{-3}$ were also omitted, following Schwartz et al. (2012). The numbers of the observations were about 17700. Among them 8 observations were discarded because they were larger than 800 $\mu g\ m^{-3}$ and 243 (around 1.5%) were discarded due to the latter reasons.

4. Experimental design

Two parallel experiments were performed to evaluate the impact of $PM_{2.5}$ DA on the analyses and forecasts of aerosols over China: an assimilation experiment and a control experiment. Both experiments used identical WRF-Chem settings and

physical parameterizations.

4.1 Spin-up ensemble forecast with perturbed Initial and boundary conditions

The initialization and spin-up procedures were identical to those reported by Schwartz et al. (2014). The ICs and lateral boundary conditions (LBCs) for the meteorological fields were provided by the National Centers for Environmental Prediction Global Forecast System (GFS).

The initial meteorological fields were created at 0000 UTC 1 October 2014 by interpolating the GFS analyses onto the model domain. The 50 ensemble members were then generated by adding Gaussian random noise with a zero mean and static background error covariances (Torn et al., 2006) to the temperature, water vapor, velocity, geopotential height and dry surface pressure fields. The ICs of each member were zero in the initial aerosol fields, representing clean conditions as described by Liu et al. (2011).

The LBCs for the meteorological fields were then interpolated from the GFS analyses from 0000 UTC 1 October 2014 to 0000 UTC 16 October 2014 and perturbed similarly to the initial fields at 0000 UTC 1 October 2014. The aerosol LBCs of each member for all experiments were idealized profiles embedded within the WRF/Chem model.

Fifty-member emissions were created by adding random noise to the anthropogenic emissions, same as reported by Schwartz et al. (2014),

$$\mathbf{E}_{ip}^{*}(\eta,t) = \mathbf{E}_{p}(\eta,t) + \boldsymbol{W}_{ip}\boldsymbol{\sigma}_{p}^{\mathrm{E}}(\eta,t)$$

where $\mathbf{E}_{ip}^{*}(\eta,t)$ is the *i*th ensemble member for the *p*th emissions variable at the ηth grid point and the *t*th hour, $\mathbf{E}_{p}$ is the unperturbed emissions. The term $\boldsymbol{\sigma}_{p}^{\mathrm{E}}$ is the standard deviation of all $\mathbf{E}_{p}$ values and in the horizontally adjacent points of grid box η at and within 2 h of t. $\boldsymbol{W}$ is a weight that was randomly drawn from a standard Gaussian distribution and varied for each ensemble member and variable but was spatially and temporally constant. No correlations between emissions variables were considered, which was a limitation of this approach. For possible negative

perturbed emissions, they were set as $\mathbf{E}_{ip}^{*}(\eta,t) = 0.001 * \mathbf{E}_{p}(\eta,t)$. This will increase the prescribed emissions more or less. However, only very few data were negative. So, this influence can be negligible.

Before the first DA cycle, a 50-member ensemble of four-day WRF-Chem forecasts was performed from 0000 UTC 1 October to 2300 UTC 4 October 2014 using the perturbed ICs at 0000 UTC 1 October 2014, the corresponding perturbed LBCs and the emissions. Then a 50-member ensemble aerosol forecasts at 0000 UTC 5 October 2014 were produced.

4.2 Assimilation experiments

Two DA experiments were performed. One was the pure assimilation of chemical ICs (hereafter expC), the others was the joint adjustment of chemical ICs and source emissions (hereafter expJ). Both DA experiments had same settings except for the emissions. They were conducted from 0000 UTC 5 October 2014 to 0000 UTC 16 October 2014. The assimilation cycle interval was 1 h.

In the first DA cycle in expJ, the first 50 ensemble chemical fields were drawn from the WRF-Chem ensemble forecasts valid at 0000 UTC 5 October 2014, as described in section 4.1. Using the ensemble aerosol forecasts, the prior emission scaling factors $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$ at 2300 UTC 4 October 2014 were calculated. $\boldsymbol{\lambda}_{i,t}^{\mathrm{p}}$ were used directly as $\boldsymbol{\lambda}_{i,t}^{\mathrm{f}}$ for the first 5 assimilation cycles (after 5 assimilation cycles, the system has been initialized, all future scaling factors could be created using the persistence forecasting operator $\mathbf{M}_{\mathrm{SF}}$). Then, the state vector $\mathbf{x}^{\mathrm{f}} = [\mathbf{C}^{\mathrm{f}}, \boldsymbol{\lambda}^{\mathrm{f}}]^{\mathrm{T}}$ was prepared. And after that, the DA cycle started.

In expC, the first chemical fields were also drawn from the WRF-Chem ensemble forecasts valid at 0000 UTC 5 October 2014. Then, the state vector $\mathbf{x}^{\mathrm{f}} = [\mathbf{C}^{\mathrm{f}}]^{\mathrm{T}}$ was prepared and the DA cycle started.

At the WRF-Chem forecast step of the subsequent assimilation cycles for both experiments, the ICs for the chemical variables of each member were drawn from the updated chemical fields of the previous cycle. The aerosol LBCs of each member for

all experiments were idealized profiles embedded within the WRF/Chem model. As for the meteorological ensemble fields, the LBCs were prepared in advance as depicted in section 4.1; the ICs of each member of the meteorological fields were drawn from the forecast meteorological fields of the previous cycle before re-centering with the GFS analysis because we do not do meteorological analysis:

$$\boldsymbol{\pi}_{i_{\mathrm{new}}} = \boldsymbol{\pi}_i + (\boldsymbol{\pi}_{\mathrm{GFS}} - \overline{\boldsymbol{\pi}}), \quad (18)$$

where $\boldsymbol{\pi}_i$ is the *i*th member of the forecast meteorological fields of the previous cycle, $\overline{\boldsymbol{\pi}}$ is the ensemble mean of the forecast meteorological fields of the previous cycle, $\boldsymbol{\pi}_{\mathrm{GFS}}$ is the meteorological field interpolated from the GFS analyses and $\boldsymbol{\pi}_{i_{\mathrm{new}}}$ is the new meteorological field used as the IC in WRF-Chem in the next cycle.

As stated in the first paragraph in this section, the settings of expC were the same as those in expJ except for the emissions. In expJ, the ensemble anthropogenic emissions were generated by using emission scaling factors. While in expC, the ensemble anthropogenic emissions were prepared by adding random noise, as stated in 4.1.

4.3 Control experiment

The control experiment was conducted for the same period as the assimilation experiment and the simulation cycle period was 1 h, as in the assimilation experiment. The first initial chemical fields were extracted from the ensemble mean valid at 0000 UTC 5 October 2014. In the subsequent simulation process, the ICs for the chemical fields were from the previous cycle's 1-h forecast. The LBCs and ICs for the meteorological fields were updated by interpolating the GFS analyses. The emissions were the prescribed emissions $\mathbf{E}_t^{\mathrm{p}}$ without any perturbation.

5. Results

Statistics for both expJ and expC were computed using the ensemble mean prior (background) and posterior (analysis) fields (average of the 50-member ensemble). The ensemble performances were first examined. Output from the first day of the

602 cycling DA configurations was excluded from all verification statistics to allow the
603 ensemble fields to ‘‘spin up’’ from the initial ensemble.
604 As the measurement coverage is an important factor that may determine the
605 performance in DA, we primarily focused our attention on the results from three
606 sub-regions with comparatively dense observational coverage (Figure 1): the Beijing–
607 Tianjin–Hebei region (JJJ, 12 stations for assimilation and 12 stations for verification);
608 the Yangtze River delta (YRD, 24 stations for assimilation and 24 stations for
609 verification); and the Pearl River delta (PRD, 9 stations for assimilation and 9 stations
610 for verification).
611
612 5.1 Ensemble performance
613 It is important to assess the ensemble performance for an ensemble-based DA system.
614 In a well-calibrated system, a comparison of the prior ensemble mean
615 root-mean-square error (RMSE) with respect to the observations should equal the
616 prior “total spread” (square root of the sum of ensemble variance and observation
617 error variance) (Houtekamer et al., 2005). Figure 3 shows the time series for the prior
618 ensemble mean RMSE and the total spread for $PM_{2.5}$ aggregated over all observations
619 in the three sub-regions for expJ. It indicates that the magnitudes of both the total
620 spread and the RMSE were influenced by the diurnal cycle and heavy air pollution.
621 Almost all the total spreads were smaller than the RMSE, showing an insufficient
622 spread of $PM_{2.5}$ ensemble forecasts, which is especially evident for heavy polluted
623 period with much larger RMSEs. For expC, the characteristics of the prior ensemble
624 mean RMSE and the total spread for $PM_{2.5}$ were very similar to that for the joint DA
625 experiment.
626 The magnitudes of the ensemble spread of the emission scaling factors of the
627 joint DA experiment were important for emission inversion. They were very stable
628 throughout the ~10 day experiment period, which indicates that $\mathbf{M}_{\mathrm{SF}}$ can generate
629 stable artificial data to generate the ensemble emissions. For $\boldsymbol{\lambda}^{\mathrm{f}}_{\mathrm{PM2.5}}$, they ranged
630 from 0.25 to 1 in most model area. Figure 3d shows the area-averaged time series

extracted from the ensemble spread of $\boldsymbol{\lambda}^{\mathrm{f}}_{\mathrm{PM2.5}}$. It shows that the ensemble spread was stably distributed around 0.5, which indicates that the uncertainty of the ensemble emissions was about 50%.

5.2 Impact on aerosol ICs

To evaluate quantitatively the impact of the ensemble assimilation system on the ICs, the mean errors (bias), RMSEs and correlation coefficient (CORR) of the assimilation experiment and the control run were first analyzed. These statistics were calculated against independent observations over all the analyses from 6 to 16 October 2014. Table 1 shows that the bias magnitudes of the control run were 15.9 and 20.6 μg m^{-3} for the YRD and the PRD, respectively, suggesting a significant overestimation of the WRF-Chem aerosol mass in these two sub-regions. However, a significant underestimation of the aerosol mass occurred in the JJJ region, where the model bias was −18.0 μg m^{-3}. The RMSEs of the control run were 81.6, 30.6 and 31.8 μg m^{-3} for the JJJ, YRD and PRD regions, respectively. After assimilation, the statistics showed an apparent improvement and the magnitude of the bias and the RMSE decreased for both DA experiment. For expJ, both the maximum bias and the RMSE were obtained in the JJJ region, and were -10.3 and 66.9 μg m^{-3}, respectively. The CORR increased from 0.79, 0.60, and 0.62 to 0.83, 0.85, and 0.80 for the JJJ, YRD and PRD, respectively. The statistics of expC were very similar to those of expJ. The bias and the RMSE in the JJJ region were -12.2 and 64.0 μg m^{-3}, respectively. And the CORR were 0.85, 0.80, and 0.80 for the JJJ, YRD and PRD, respectively. These results indicate that the initial $PM_{2.5}$ fields can be adjusted efficiently by the EnSRF.

It is interesting to note that expC has better RMSE and CORR than expJ but poor bias in JJJ. And expC has better bias and RMSE than expJ but poor CORR in PRD. Maybe small number of samples caused the uncertainties of the statics. However, the differences were very small. The analysis of both experiments were very similar.

Then the analysis increments (i.e. $\bar{\mathbf{x}}^{\mathrm{a}} - \bar{\mathbf{x}}^{\mathrm{b}}$) were investigated to show the direct impact of $PM_{2.5}$ DA. They are determined by both the observation increments and the relative magnitudes of the forecast error and the observation error, based on Equation

(7). From Figure 4(a), (e) and (f), the increments of both assimilation experiments were distributed around the observations as expected. However, the impact of assimilating $PM_{2.5}$ observations was not limited to the areas where observations were located, observations information was also transported to other areas through the WRF-Chem forecast. Besides, the ensemble forecasts also partly contributed to the spatial distribution of the $PM_{2.5}$ mass. Therefore, the spatial distributions of the $PM_{2.5}$ mass in both assimilation experiments were significantly different from the control run (see Figure 4(b), (c) and(d)), which suggest that assimilation $PM_{2.5}$ observations impacts greatly on the aerosol ICs. The $PM_{2.5}$ mass magnitude of both assimilation experiments were smaller than that of the control run at the lowest model level in the YRD, the PRD and in central China. Conversely, positive differences (analysis minus control) were gained in the JJJ region and in northeast China. These indicated the reduction of the overestimation or underestimation of the WRF-Chem simulation over these regions with data assimilation.

5.3 Impact on emissions

To determine the impact of assimilating $PM_{2.5}$ observations on the chemical emissions, we analyzed the area-averaged time series extracted from the forecast emission scaling factors, the optimized emission scaling factors, the prior emissions and the optimized emissions. Figure 5 shows that $\boldsymbol{\lambda}^{\mathrm{f}}_{\mathrm{PM2.5}}$ were changed along with $\boldsymbol{\lambda}^{\mathrm{a}}_{\mathrm{PM2.5}}$. This indicates that observation information ingested from the previous observations was incorporated through the usage of the time smooth operator.

Figure 5 also shows that although the prior emissions $\mathbf{E}^{\mathrm{p}}_{\mathrm{PM2.5}}$ had no diurnal variation when the experiments were designed, the optimized $PM_{2.5}$ scaling factor, $\boldsymbol{\lambda}^{\mathrm{a}}_{\mathrm{PM2.5}}$, showed an obvious variation with time, as did the optimized unspeciated primary sources of $PM_{2.5}$, $\mathbf{E}^{\mathrm{a}}_{\mathrm{PM2.5}}$. Moreover, the values of $\boldsymbol{\lambda}^{\mathrm{a}}_{\mathrm{PM2.5}}$ were <1 at almost all times in the YRD and PRD, which resulted that the analyzed emission $\mathbf{E}^{\mathrm{a}}_{\mathrm{PM2.5}}$ were lower than the prior $PM_{2.5}$ emissions $\mathbf{E}^{\mathrm{p}}_{\mathrm{PM2.5}}$. In the YRD, the prior $\mathbf{E}^{\mathrm{p}}_{\mathrm{PM2.5}}$ was about 0.127 $\mu g\ m^{-2}\ s^{-1}$ over all hours. After assimilation, the time-averaged optimized

690 $\mathbf{E}^{a}_{PM2.5}$ decreased to 0.107 μg m^{-2} s^{-1}, about 15.6% lower than the prior value. In the
691 PRD, the prior $\mathbf{E}^{p}_{PM2.5}$ was about 0.10 μg m^{-2} s^{-1}. The time-averaged optimized
692 $\mathbf{E}^{a}_{PM2.5}$ decreased to 0.066 μg·m^{-2} s^{-1}, leading to a decrease of 35.0%. However,
693 larger values for the optimized $\mathbf{E}^{a}_{PM2.5}$ were obtained in the JJJ region in three
694 periods, from 1600 UTC 6 October to 0000 UTC 8 October, from 1600 UTC 9
695 October to 0000 UTC 10 October, and from 1600 UTC 13 October to 0000 UTC 15
696 October as a result of the increased optimized scaling factor $\boldsymbol{\lambda}^{a}_{PM2.5}$. This may have
697 been caused by the burning of crop residues during harvesting in this region (Li et al.,
698 2016), which was not taken into account in the prior emissions. However, the $PM_{2.5}$
699 measurements network was still spatially sparse and heterogeneous in this work.
700 Almost all measurements were located in the city and no data available in the rural.
701 Meanwhile, the crop residues burning always occur in the rural region. Therefore, the
702 $PM_{2.5}$ measurements network can only capture the burning information a few hours
703 later. Hence, although the system is able to detect the emission changes caused by
704 burning events, the time that the system started to show increased scaling factors
705 might be not accurate enough (may shift a few hours later). Maybe a Kalman
706 smoother would have been a better system to solve this problem.

707 The NO, SO_2 and NH_3 emissions were all adjusted to some extent by our DA
708 approach (see Figure 6). The NO emissions increased by 41.3, 43.7 and 20.3% in the
709 JJJ, YRD and PRD regions, respectively. The SO_2 emissions increased by 16.3, 10.0
710 and 18.3% and the NH_3 emissions increased by 16.7, 7.8 and 7.5% in the JJJ, YRD
711 and PRD regions, respectively.

712 Figure 7 shows the spatial distribution of the time-averaged scaling factors
713 $\boldsymbol{\lambda}^{a}_{PM2.5}$ at the lowest model level over all hours from 6 to 16 October 2014, since the
714 emissions at higher levels were so small that the impact of assimilating $PM_{2.5}$
715 observations was negligible. Figure 8 shows the distribution of $\mathbf{E}^{p}_{PM2.5}$ and the
716 time-averaged differences between the ensemble mean of the assimilation and the
717 prior values.

718 These patterns are consistent with those in Figure 5. Negative differences were

obtained in most areas of the YRD and PRD, indicating that the $PM_{2.5}$ DA primarily decreased the $PM_{2.5}$ emissions. Conversely, positive differences were obtained in South Hebei, North Henan and Southeast Shanxi provinces, indicating that DA increased the $PM_{2.5}$ emissions.

As the economy in China has developed, the spatiotemporal distribution of emissions has changed as a result of changes in energy consumption, the structure of the energy market and advances in technology. Therefore although this inventory of emissions may have correctly described anthropogenic emissions in 2006 when it was constructed, it is not representative of the anthropogenic emissions in 2014. Theoretically, the assimilated emissions should reduce the uncertainty in the prior emissions as a result of the application of observations. Different from the situations that standard national emission inventories were reported by government in USA, European or other countries, the rapid economic development and complexity of emission sources in China lead to large uncertainties in the current emission inventories even for the latest version. Thus it's impossible for us to conduct the direct evaluation on emissions.

Although we had no direct emission observations to evaluate the analyzing emissions, which was a challenging to many emission inversion research teams (e.g. Tang et al, 2011; Miyazaki et al., 2012; Ding et al., 2015; Mclinden et al., 2016; etc.), the improvement of emissions can be verified in terms of two aspect, the diurnal variation and the location of increased emissions. The diurnal variation in the assimilated emissions verified this statement to some extent. Especially in the PRD and YRD, $\mathbf{E}^{a}_{PM2.5}$ in the daytime were always larger than those in the night, which agreed well with Olivier et al. (2003), the WRAP (2006) and Wang et al. (2010). In addition, the locations of the larger values for the optimized $E^{a}_{PM2.5}$ in the JJJ region was in good agreement with the place of the crop residues burning *traced by the* environmental satellite of China. There were 10, 231, 37 and 3 crop residue burning spots in Hebei, Henan, Shandong and Shanxi province respectively from 5 to 11 October 2014 and 7, 20, 5 and 21 respectively from 12 to 18 October 2014 (Weekly Crop Residue Burning Monitoring Report traced by

749 Environmental Satellite, 2015a, 2015b).

750 However, the analysis emissions are only a mathematical optimum. They are
751 influenced greatly by the model errors and the observation errors. In addition, only
752 surface $PM_{2.5}$ observations were applied in this work, which may lack abundant
753 constraint on the sources of the secondary aerosol precursors. More observations are
754 needed to obtain reliable emissions for the sources of the gas-phase precursors.

755

756 5.4 Verification of aerosol forecasting

757 For the assimilation experiment, 48-h forecasts were performed at each 0000
758 UTC from 6 to 16 October 2014 with the hourly forecast output for both assimilation
759 experiments. For the verification forecasting experiment for expJ (hereafter fcJ), the
760 ensemble mean of the analyzed ICs and emissions of expJ were used in this
761 longer-range model forecast. For the verification forecasting experiment for expC
762 (hereafter fcC), the ensemble mean of the analyzed ICs of expC and the prescribed
763 anthropogenic emissions were used.

764 In order to get a more visualized picture of the impact of DA for both
765 assimilation experiments, time series of the hourly $PM_{2.5}$ extracted from the analysis
766 (AN), the control run (CT) and the hourly output of 48-h forecast (fc24 for the first
767 day forecast and fc48 for the second day forecast) were compared with the
768 observations (OBS) for three megacities Beijing, Shanghai and Guangzhou,
769 respectively (Figure 9). As expected, the time series of the analysis (also the
770 background) were consistent with the observations. The control run showed large
771 deviations from the observations, especially in Shanghai and Guangzhou. Benefit
772 from DA on both the first day and the second day forecasts can be clearly seen.

773 The bias and the RMSE of the surface $PM_{2.5}$ forecasts as a function of forecast
774 range was then calculated against the independent observations for the three
775 sub-regions (Figure 10). Both the bias and the RMSEs of the control run were
776 characterized by the diurnal cycle in the YRD and PRD. The largest errors were seen
777 at 2100 UTC in the YRD (about 29 $\mu g \cdot m^{-3}$ for bias and 37 $\mu g \cdot m^{-3}$ for RMSEs) and at
778 2300 UTC in the PRD (about 36 $\mu g \cdot m^{-3}$ for bias and 41 $\mu g \cdot m^{-3}$ for RMSEs), likely

779 indicating significant systematic forecast errors at these times. From 0300 to 0900
780 UTC, the bias (about 1 μg·m^{-3} in the YRD and -5 μg·m^{-3} in the PRD) and the RMSE
781 values (about 14 μg·m^{-3} in the YRD and 16 μg·m^{-3} in the PRD) were much smaller
782 than at other times in both the YRD and PRD, showing that WRF-Chem performed
783 well during this period. However, in the JJJ region, the bias (about -20 μg·m^{-3}) and
784 the RMSEs (about 50 μg·m^{-3}) were always large as a result of a heavy pollution event.
785 After assimilation, both the magnitude of the bias and the RMSEs decreased sharply.
786 Especially in in YRD and PRD, most bias ranged from -5 to 5 μg·m^{-3} and most
787 RMSEs ranged from 11 to 14 μg·m^{-3}, further indicating that DA greatly affected the
788 ICs.

789 The improvements in the surface $PM_{2.5}$ forecasts by the joint adjustment of the
790 ICs and emissions were very large in the YRD and PRD for expJ. Large reduction of
791 the magnitude of the bias and the RMSEs due to assimilation can be seen for almost
792 the entire 48-h forecast range. From 10- to 23-h and from 34- to 47-h, in particular,
793 the relative reduction in RMSE was about 37.5%. However, the DA impact was much
794 smaller for 3- to 9-h forecast ranges, which are at daytime of the first day forecast. In
795 addition, the improvements were nearly negligible in PRD from 27- to 33-h, the
796 daytime of the second day forecast, suggesting that the benefit gained from adjusting
797 the ICs decreased progressively and eventually disappeared with model integration.
798 And the performance was actually deteriorated in YRD during the same time. One of
799 the possible reasons was that chemical model performed sufficiently well during
800 daytime when the boundary layer was unstable and therefore the further improvement
801 was more difficult. And there were always large errors during the night when the
802 boundary layer was stable, so that large improvements could be obtained. The other
803 possible reason can be attributed to the a priori constant emissions. The differences
804 between the optimized $PM_{2.5}$ emissions and the prior emissions were comparatively
805 small during the day, but the optimized $PM_{2.5}$ emissions were much smaller than the a
806 prior emissions during the night. So that the control run could performed worse during
807 the night and it could performed well during the day. Given the a priori variable

808 emissions provided, the control run will perform better during the night. Nevertheless，
809 attributed greatly to the large adjustment of chemical emissions, substantial
810 improvements were still achieved from 34- to 47- h. These results revealed that joint
811 adjustment of the ICs and emissions can improve surface $PM_{2.5}$ forecasts up to 48 h in
812 the YRD and PRD.

813 As for expC, it seemed that large improvements in the surface $PM_{2.5}$ forecasts
814 were gained through the adjustment of the ICs in PRD from 10- to 23-h and from 34-
815 to 47-h. Large reduction of the magnitude of the bias and the RMSEs due to
816 assimilation can be seen during this period. The relative reduction in RMSE ranged
817 from 25% to 37.5%. However, the forecasts deviated much from the observations for
818 3- to 9-h and 27- to 33-h forecast ranges. One of the reason may be that the
819 adjustment of the ICs decreased the analysis field too much on the whole since the
820 WRF-Chem forecast aerosol mass was systematically overestimated in PRD (see
821 Figure 4, Figure 9f and Figure 10e). While this aerosol mass overestimation might be
822 also due to the possibly overestimated emissions in some time periods (not all-day
823 long) which are not corrected in the simulation. So the over-adjusted ICs compensated
824 the unadjusted emissions in some period but also lead to the negative biases for the
825 periods when emission is not overestimated or underestimated. The other factor was
826 the diurnal variation. It is very clear that $PM_{2.5}$ mass gradually decreased with time
827 from 0000 UTC to 0008 UTC and then obtained the smallest value. After that it
828 increased with time from 0009 UTC to 0023 UTC obtained the largest value at about
829 0000 UTC. Both reasons led to the systematically underestimation of $PM_{2.5}$ mass of
830 fcC from 3- to 9-h and from 27- to 33-h, though maybe the aerosol ICs were very
831 close to the observations. Therefore, both the magnitude of the bias and the RMSEs of
832 the fcC were larger than those of the control run. In addition, $PM_{2.5}$ forecasts of the
833 fcC were benefit much from the diurnal variation and the adjustment of the ICs from
834 10- to 23-h and from 34- to 47-h. As a consequence, the magnitude of the
835 corresponding bias and the RMSEs of the fcC were smaller than those of the control
836 run. Similar statics characteristics were also gained in YRD. But the improvements
837 were comparatively small from 10- to 23-h and from 34- to 47-h. However, the

838 performance of fcJ was much better than that of fcC during the night in PRD and
839 YRD. While in the daytime, the improvement of expJ seems to be not so big or even
840 negligible. This could be attributed much to the emissions since the ICs of both
841 forecasts were very similar. In the forecast experiment of expC, the emissions were
842 the default monthly anthropogenic emissions. While in the forecast experiment of
843 expJ, the assimilated emissions were much smaller than the default anthropogenic
844 emissions in almost all the night in both regions indicating that the prior emission
845 uncertainties might be the dominating reasons that cause biases between observed and
846 model simulated concentrations in these cases. In the daytime in PRD, the assimilated
847 emissions were a little smaller than the default anthropogenic emissions. But in the
848 daytime in YRD, the assimilated emissions were a little larger than the default
849 anthropogenic emissions for most time (see Figure 5). While those changes between
850 assimilated emissions and prior emissions in the daytime are not as significant as that
851 in the nighttime.

852 Both DA systems did not perform as well in the JJJ region as in the YRD and
853 PRD. Relatively smaller improvements were achieved in the first 24-h forecast but
854 then no improvements were achieved afterwards in JJJ. One possible reason for this
855 result may be systematic errors due to chemistry mechanism in WRF-Chem. The
856 sources of the aerosols are so complex that our knowledge of their formation
857 mechanisms is far from clear and large uncertainties still exist in the model
858 simulations. Chemical transport models have a tendency to underestimate PM
859 concentrations, especially during episodes of heavy pollution (Denby et al., 2007) due
860 to some missing reactions (Wang et al., 2014; Zhang et al., 2015, Zheng et al., 2015;
861 Chen et al., 2016). Another reason can be attributed to the forecast meteorological
862 fields. There were still large uncertainties, especially when boundary layer was stable
863 and the wind speed was very small during episodes of heavy pollution. As a result, a
864 large bias may be obtained in forecasts of heavy pollution given the ICs and emission
865 inventories achieved from the joint assimilation. Another reason may be the sparse
866 coverage of measurements. There were only 12 sites in the JJJ region (Figure 1) and
867 the measurement coverage was much sparser than in the YRD or PRD.

6. Summary and Discussion

The EnSRF algorithm was extended to adjust the chemical ICs and the primary and precursor emissions to improve forecasts for surface $PM_{2.5}$. This system was applied to assimilate hourly surface $PM_{2.5}$ measurements from 5 to 16 October 2014 over China. To evaluate the effectiveness of DA, 48-h forecasts were performed using the optimized ICs and emissions, together with a control experiment without DA. Besides, the experiment of pure assimilation chemical ICs and the corresponding 48-h forecasts experiment were also performed for comparison. The results indicated that the forecasts with the optimized ICs and emissions performed much better than the control simulations. Large improvements were achieved for almost all the 48-h forecasts, particularly in the YRD and PRD. However, it did show some improvements in the first 24-h but then there is no difference between the control run and the forecasts in the JJJ region afterwards, which may be attributed to the sparse measurement coverage and the deficiencies in the model system for forecasting heavy pollution. Comparing to the forecasts with only the optimized ICs, the forecasts with the joint adjustment were always much better during the night in the PRD and YRD regions. However, they were very similar during daytime in both regions. And in the JJJ region, they performed similarly for almost the entire 48-h forecast range.

There are still some limitations in this study. Firstly, we use the default monthly anthropogenic emissions as the prior emissions and no time variation was added to keep objective, since no resolution of temporal allocations at shorter but critical (e.g.,day-of-week, diurnal) scales is available. As shown in earlier work, the constant emissions will worsen the chemical forecasts (de Meij et al., 2006; Wang et al, 2009). For the joint DA system itself, it cannot benefit from the constant prior anthropogenic emissions. But the normalized RMSE in Figure 10g decreased due to the poor forecasts of control run. The control run will perform better when variable emissions within the day are allowed, especially during the night. As a result, the relative reduction in RMSE could not be so large during the night. Secondly, no correlations between emissions variables were considered when perturbing the emissions, which

will lead to the reduction of the correlations between the variables. Thus, the chemical forecast will deviate from the truth to some degree. Fortunately, the perturbed emissions were only used in the initialization and spin-up experiment and expC. Therefore, there were no impacts on expJ and the control run except for expC. Thirdly, $\mathbf{E}_{\mathrm{EC}}$ and $\mathbf{E}_{\mathrm{ORG}}$ are not perturbed in expJ. However, as stated in Sect. 2.3.2, the ensemble spread of $\mathbf{OC_1}$ and $\mathbf{OC_2}$ can be kept at a certain level. As a result, $\mathbf{OC_1}$ and $\mathbf{OC_2}$ changed much contributed to the $PM_{2.5}$ assimilation in expJ, which suggests that the influence of not perturbing $\mathbf{E}_{\mathrm{EC}}$ and $\mathbf{E}_{\mathrm{ORG}}$ could be negligible. But, because of the too small magnitudes of $\mathbf{BC_1}$ and $\mathbf{BC_2}$, the differences (assimilation minus control) of $\mathbf{BC_1}$ and $\mathbf{BC_2}$ were nearly close to zero. Fourthly, the experiment (expE) where only emissions were assimilated was not included here. But it was still worth to simultaneously assimilate the chemical ICs and emission. For one thing, in expE, the chemical concentrations can be updated by the WRF-Chem model simulations with the assimilated emissions as the initial field in each DA cycle. That means that the 50-member ensemble forecasts were performed twice and it was time consuming. For another, better concentration analysis could be obtained in expJ due to the simultaneous assimilation of ICs and emissions. While in expE, there may be larger uncertainties for the updated chemical concentrations through WRF-Chem due to the deficiency of chemistries and the uncertainties of the ICs. This will lead to larger uncertainties for the emission inversion. Also the improvement of $PM_{2.5}$ forecasts will be limited due to the comparatively poor chemical ICs.

This study represents the first step in the simultaneous optimization of chemical ICs and emissions and only surface $PM_{2.5}$ measurements were assimilated. In future work, gas-phase observations of SO_2, NO_2 and CO will be used to further improve the performance of this DA system.

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List of Figures and Table

Figure 1. Locations of 77 PM2.5 assimilation observation stations (black dot) and the 77 independent observation stations (red triangle) in the model domain. The three colored boxes mark sub-regions with relatively dense coverage for the Beijing–Tianjin–Hebei region (JJJ, 12 assimilation stations and 12 independent stations, red box), the Yangtze River delta (YRD, 24 assimilation stations and 24 independent stations, blue box) and the Pearl River delta (PRD, 9 stations and 9 independent stations, green box).

Figure 2. (a) Framework of M_{SF} and (b) flow chart of the data assimilation system that simultaneously optimizes the chemical initial conditions and emissions.

Figure 3. Time series of prior ensemble mean RMSE and total spread for PM2.5 concentrations aggregated over all observations over the three sub-regions: (a) Beijing–Tianjin–Hebei region; (b) Yangtze River delta; (c) Pearl River delta; and (d) time series of the area mean ensemble spread for $\lambda_{PM2.5}$ over the three sub-regions.

Table 1. Comparison of the surface PM2.5 mass concentrations from the control and assimilation experiments to observations over all analysis times from 6 to 16 October 2014.

Figure 4. Spatial distribution of the PM2.5 mass (μg·m−3) of the (a) observations; (b) simulation of the control run; (c) analysis of expJ; (d) analysis of expC; (e) increments of expJ; (f) increments of expC; at the lowest model level averaged over all hours from 6 to 16 October 2014.

Figure 5. Hourly area-averaged time series of emission scaling factors (black) extracted from the ensemble mean of the analyzed $\lambda^{a}_{PM2.5}$ and the corresponding analyzed unspeciated primary PM2.5 emissions $E^{a}_{PM2.5}$ (blue) over the three sub-regions: (a) Beijing–Tianjin–Hebei region; (b) Yangtze River delta; and (c) Pearl River delta.

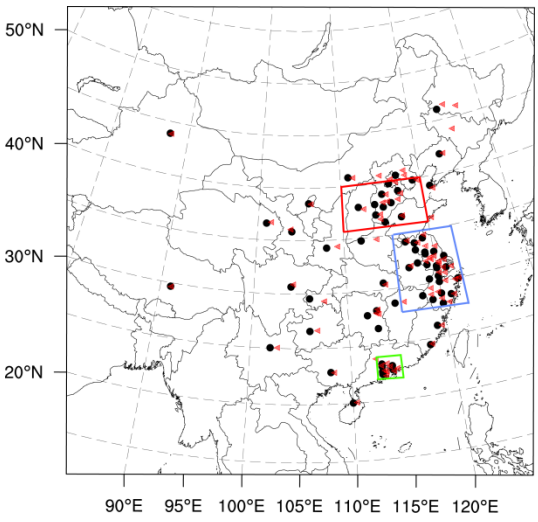
Figure 6. Hourly area-averaged time series of emission scaling factors extracted from the ensemble mean of the analyzed (a) $\boldsymbol{\lambda}^{a}_{NO}$; (a) $\boldsymbol{\lambda}^{a}_{SO2}$; (a) $\boldsymbol{\lambda}^{a}_{NH3}$ over the three sub-regions: Beijing–Tianjin–Hebei region (JJJ, black), Yangtze River delta (YRD, green), and Pearl River delta (PRD, blue).

Figure 7. Spatial distribution of $\lambda_{PM2.5}$ at the lowest model level averaged over all hours from 6 to 16 October 2014.

Figure 8. Spatial distribution of (a) the prior unspeciated primary sources of PM2.5 (μg·m−2 s−1) and (b) the time-averaged differences between the ensemble mean analysis and the prior values (μg· m−2 s−1) at the lowest model level averaged over all hours from 6 to 16 October 2014.

Figure 9. Time series of the hourly PM2.5 obtained from observations (circle), analysis (blue line), control run (black line) and hourly output of 48-h forecast in three megacities: (a) Beijing; (c) Shanghai; and (e) Guangzhou in expJ and (b) Beijing; (d) Shanghai; and (f) Guangzhou in expC. See text in section 5.4.

Figure 10. Bias of surface PM2.5 as a function of forecast range calculated against independent observations over the three sub-regions: (a) Beijing–Tianjin–Hebei region; (c) Yangtze River delta; (e) Pearl River delta and RMSE over (b) Beijing–Tianjin–Hebei region; (d) Yangtze River delta; (f) Pearl River delta; (g) Normalized RMSE (assimilation divided by control) for expJ and (h) (g) Normalized RMSE for expC.



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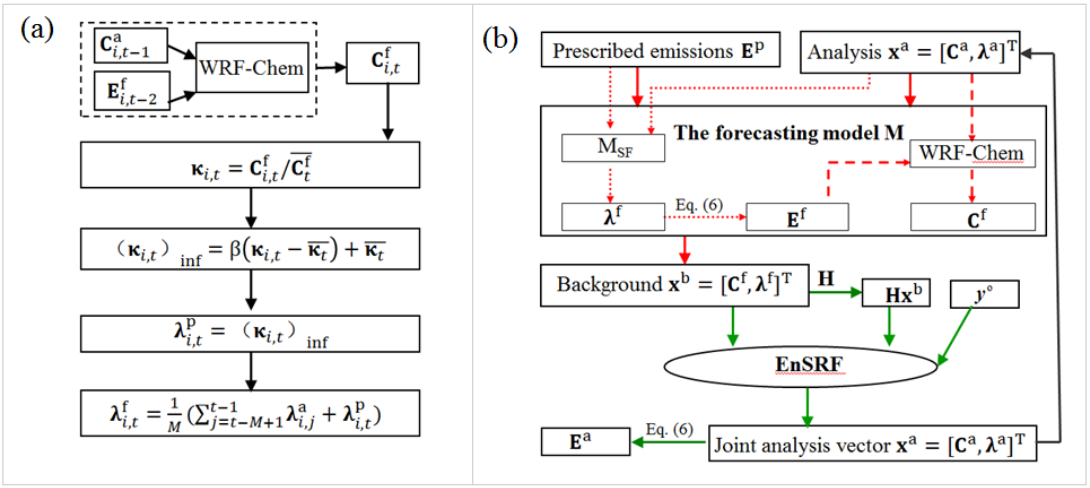

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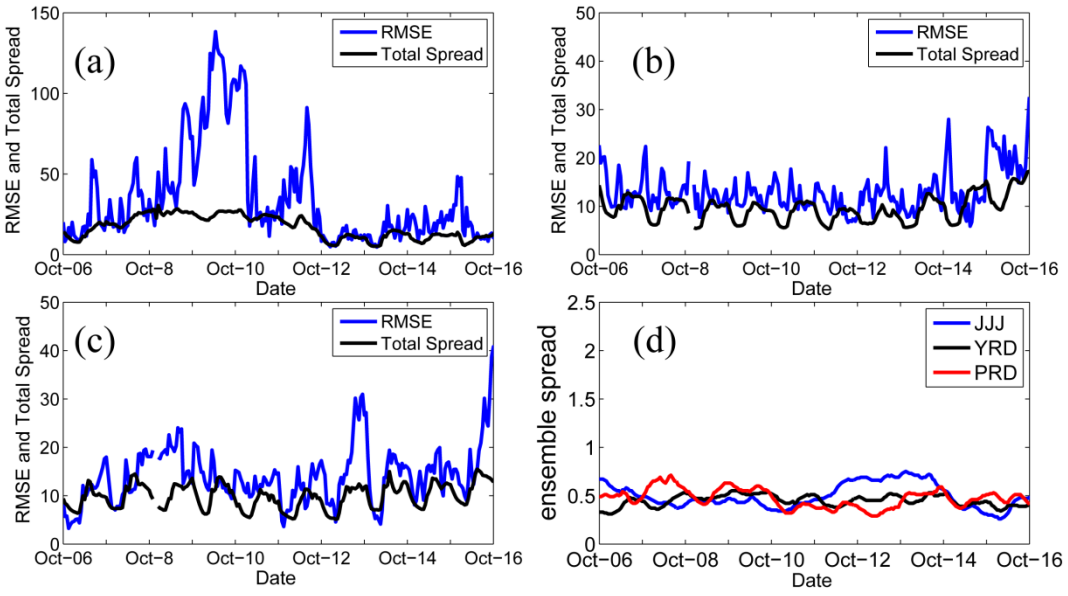

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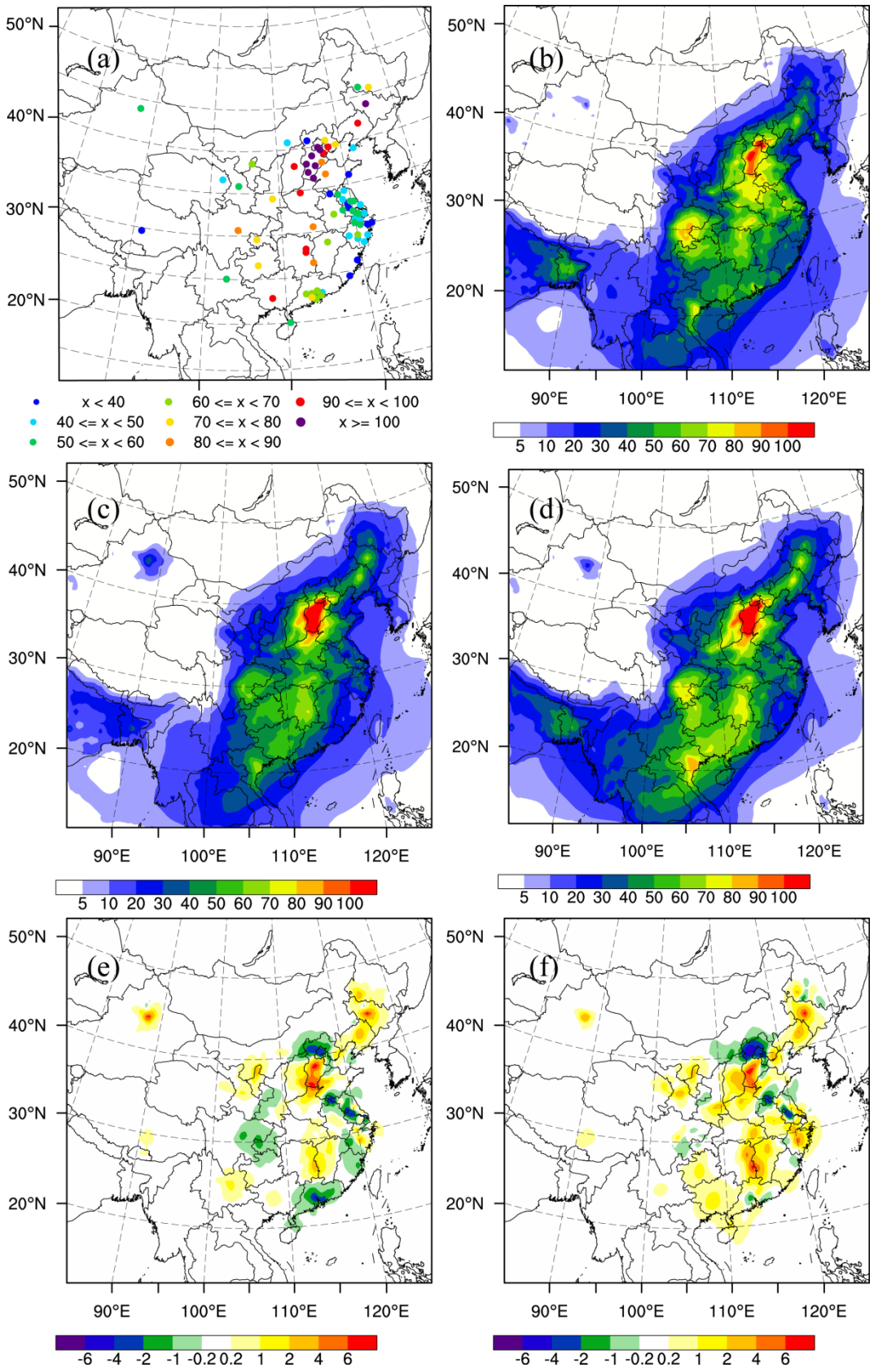
Region	Experiment	Mean observed value	Mean simulated value	BIAS	RMSE	CORR
Beijing–Tianjin–Hebei	Control	116.3	98.3	−18.0	81.6	0.790
	expJ		106.0	−10.3	66.9	0.827
	expC		104.1	-12.2	64.0	0.845
Yangtze River delta	Control	48.5	64.4	15.9	30.6	0.593
	expJ		46.9	-1.6	15.3	0.846
	expC		46.1	-2.4	17.3	0.803
Pearl River delta	Control	61.8	82.4	20.6	31.8	0.624
	expJ		66.5	4.7	16.1	0.800
	expC		64.1	-2.3	15.6	0.797



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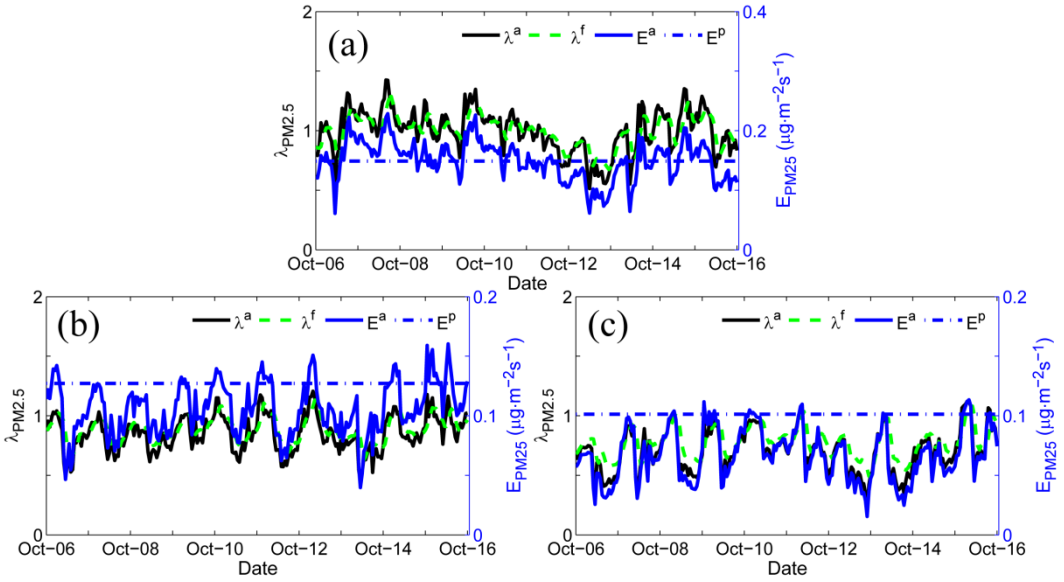

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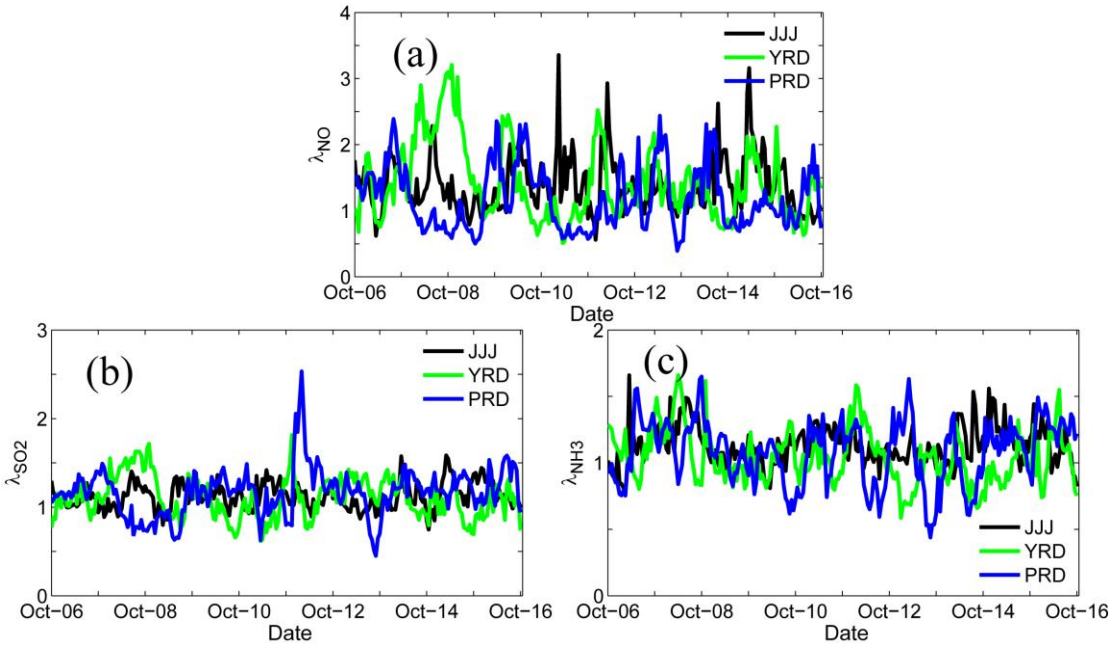

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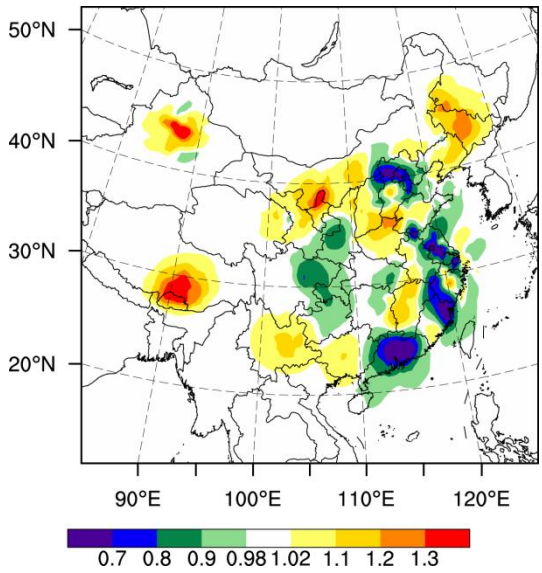

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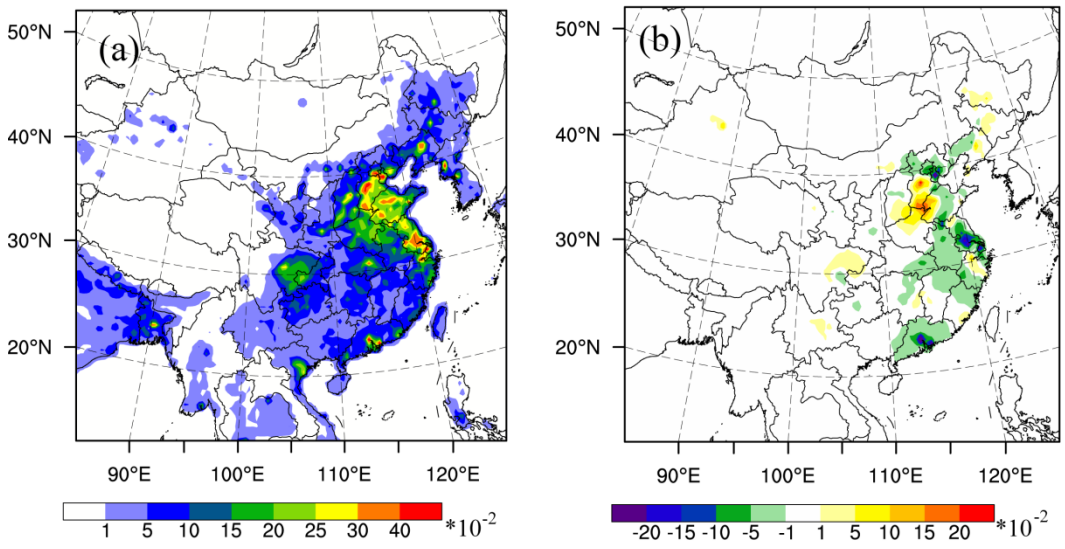

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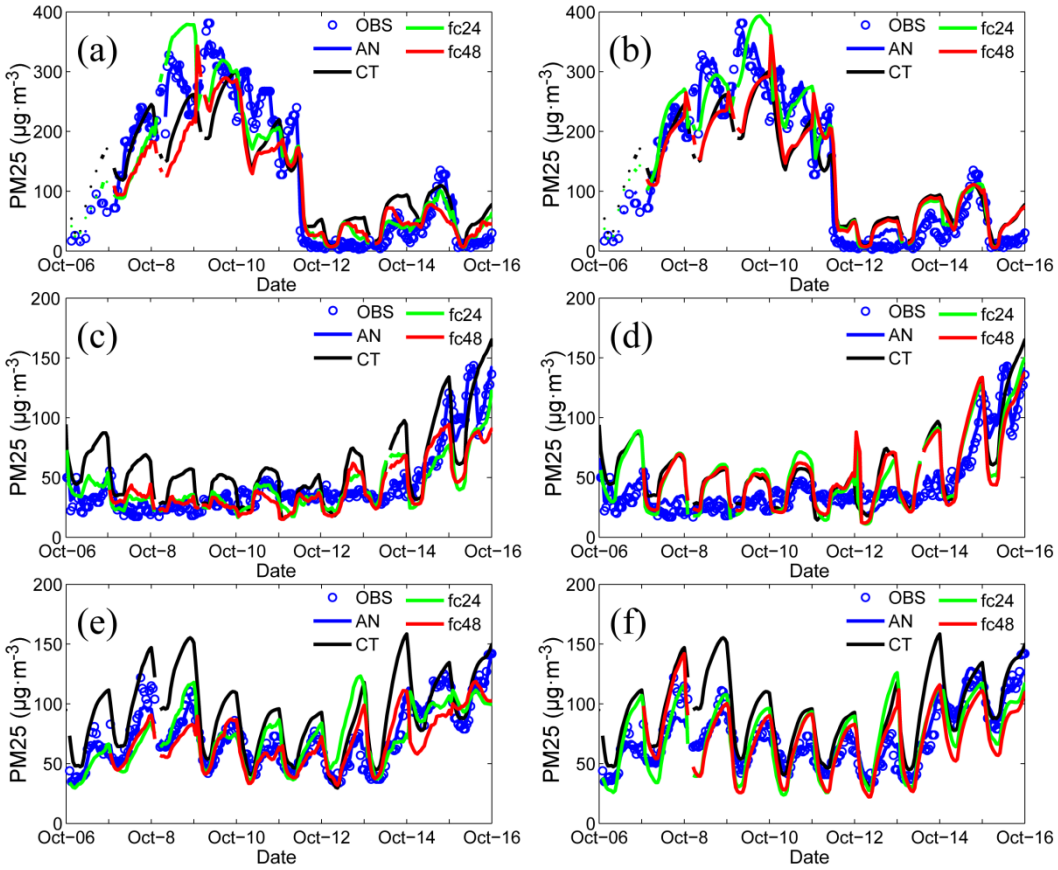

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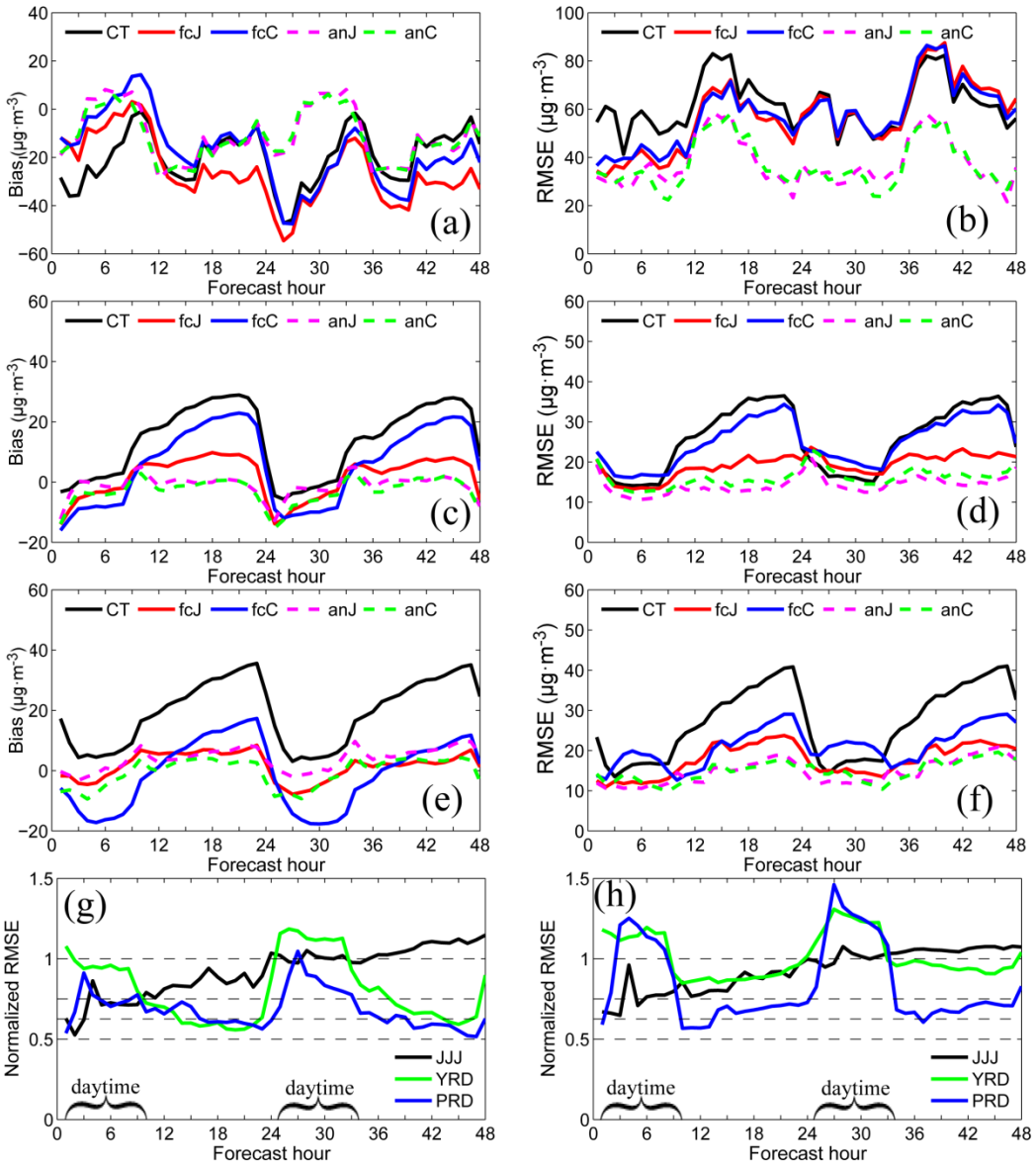

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