Implementation and application of Ensemble Optimal Interpolation on an operational chemistry weather model for improving PM$_{2.5}$ and visibility predictions

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Abstract. The data assimilation technique is one of the important ways to reduce the uncertainty of atmospheric chemistry model input and improve the model forecast accuracy. In this paper, an ensemble optimal interpolation assimilation (EnOI) system for a regional online chemical weather numerical forecasting system (GRAPES_Meso5.1/CUACE) is developed for operational use and efficient updating of the initial fields of chemical components. A heavy haze episode in eastern China was selected, and the key factors affecting the EnOI, such as localization length-scale, ensemble size, and assimilation moment, were calibrated by sensitivity experiments. The impacts of assimilating ground-based PM$_{2.5}$ observations on the model chemical initial field and PM$_{2.5}$, visibility forecasts were investigated. The results show that assimilation of PM$_{2.5}$ ~~significantly~~ reduces the uncertainty of the initial PM$_{2.5}$ field considerably. ~~The mean error and root mean square error (RMSE) of initial PM$_{2.5}$ for mainland China have all decreased by more than 75%, and the correlation coefficient could be improved to more than 0.95. Even greater improvements appear in North China~~. Using only 50% observations to do the assimilation, the root mean square error (RMSE) of initial PM$_{2.5}$ for independent verification sites in mainland China decreases from 73.7 to 46.4 μg m^{-3}, and the correlation coefficient increases from 0.58 to 0.84. An even larger improvement appears in North China. For the forecast fields, assimilation of PM$_{2.5}$ improves PM$_{2.5}$ and visibility forecasts throughout the ~~lead~~ time window of 24 h. The PM$_{2.5}$ RMSE can be reduced by 10%-21% within 24 h, ~~but~~ and the assimilation effect is most ~~obvious~~ remarkable in the first 12 h. ~~The assimilation moment chosen at 1200 UTC is more effective than that at 0000 UTC for improving the forecast, because the discrepancy between simulation and observation at 1200 UTC is larger than that at 0000UTC, indicating the assimilation efficiency will be higher when the bias of the model is higher.~~ Within the same assimilation time, the assimilation efficiency varies with the discrepancy between model forecasts and observations at the moment of assimilation, and the larger the deviation, the higher the efficiency. ~~Assimilation of PM$_{2.5}$ also improves visibility forecast accuracy significantly.~~ The assimilation of PM$_{2.5}$ further contributes improvement of visibility forecast. When the PM$_{2.5}$ increment is negative, it corresponds to an increase in visibility, and when the PM$_{2.5}$ analysis increment is positive, visibility decreases. It is worth noting that the improvement of visibility forecasting by assimilating PM$_{2.5}$ is more obvious in the light pollution period than in the heavy pollution period, since visibility is much more affected by humidity during the heavy pollution period

accompanied by low or extreme low visibility. To get further visibility improvement, especially for extreme low visibility during severe haze pollution, not only $PM_{2.5}$ but also relative humidity should be simultaneously assimilated as well. The results of this study shows that the EnOI may provide a practical and cost-effective alternative to the EnKF for the applications where computational cost is a main limiting factor, especially for real-time operational forecast.

1 Introduction

Air pollution is an intractable problem that all developing countries with high population in the world are facing at present. ~~The rapid development of industrialization and urbanization has led to different degrees of air pollution in many countries and regions (Perez et al., 2020; Xiao et al., 2020; Sahu and Kota, 2017). The energy-intensive and coal-fired economy in China has led to a dramatic deterioration in air quality over the past three decades(Piovani, 2017).~~ $PM_{2.5}$ plays an important role in air pollution, and its concentration will directly affect air quality. From the health perspective, long-term exposure to high concentrations of $PM_{2.5}$ has adverse effects on the human body, including the respiratory system, cardiovascular disease and other chronic diseases(Ghorani-Azam et al., 2016)~~;.~~ From the meteorological perspective, aerosol particles can effectively absorb and scatter solar radiation, change the intensity and direction of sunlight, reduce atmospheric horizontal visibility(Liu et al., 2019; Yadav et al., 2022; Ting et al., 2022), leading to ~~low visibility events occur frequently during the~~ haze ~~period~~episodes which~~.~~ are characterized by significant growth in the concentration of aerosol particles and sharp reduction of visibility. ~~Aerosol particles can effectively absorb and scatter solar radiation, changing the intensity and direction of sunlight, resulting in reduced atmospheric horizontal visibility(Liu et al., 2019; Yadav et al., 2022; Ting et al., 2022), which will affect people's daily travel and traffic safety, etc.~~

Accurate $PM_{2.5}$ and visibility forecasts are critical for human health, air quality assessment and public transportation safety issues(Zhang et al., 2013). Chemistry Transport Model (CTM) or Coupled chemistry meteorology models (CCMM) are~~is a~~ ~~key~~important tool~~s~~ for $PM_{2.5}$ and visibility forecasting, and is pivotal in air quality and atmospheric chemistry research. However, various uncertainties exist in the simulation of atmospheric components in CTM or CCMM, especially for aerosols (Lee et al., 2016). The complexity of atmospheric pollution formation mechanisms and model structure, the uncertainty of chemical initial conditions (ICs) and the lag in emission inventories lead to a deviation of air quality forecast results from observed comparisons. ~~, which can reach 30-50% under heavy pollution conditions(Zheng et al., 2015). The forecast accuracy of air quality forecasting still needs to be improved~~ (Wang et al., 2016; Peng et al., 2021b)~~.~~

Data assimilation (DA) is one of the most effective ways to improve model predictions. ~~It combines observational information with numerical models to provide an estimate of the state of the system which is widely used in atmospheric, oceanic, and land surface surveys. DA were earliest applied to numerical weather prediction(Navon, 2009). There are many DA methods for example, Polynomial Interpolation(Panofsky, 1949; Gilchrist and Cressman, 1954), the methods of successive corrections(Bergthórsson and Döös, 1955), and the Kalman Filter(KF)(Kalman, 1960), the optimal interpolation (OI) (Gandin,~~

1963), the variational assimilation methods (e.g., the three-dimensional variational assimilation (3D-VAR), the four dimensional variational assimilation (4D-Var)) (Talagrand and Courtier, 1987; Derber, 1989), Ensemble Kalman filter method (EnKF) (Evensen, 1994), Ensemble Optimal Interpolation (EnOI) (Oke et al., 2002; Evensen, 2003) , etc. The use of data assimilation in atmospheric chemistry models to improve air quality forecasting is more recent. For example, by assimilating aerosol observations with OI(Zheng et al., 2018; Tombette et al., 2009; Carnevale et al., 2021; Tang et al., 2015; Mauricio Agudelo et al., 2015); 3D-Var(Fu and Zhu, 2011; Feng et al., 2018; Wang et al., 2020; Li et al., 2013; Liu et al., 2011; Ye et al., 2021); 4D-Var(Cao et al., 2022; Wang et al., 2021; Liu et al., 2021; Skachko et al., 2016; Zhang et al., 2011); EnKF(Lin et al., 2007; Tang et al., 2011; Pagowski and Grell, 2012; Tang et al., 2016; Lopez-Restrepo et al., 2020; Park et al., 2022); EnOI(Wang et al., 2016); Four-dimensional ensemble Kalman filter (4DEnKF)(Cheng et al., 2019) ; Four Dimensional Local Ensemble Transform Kalman Filter (4D-LETKF)(Dai et al., 2019). Weather prediction had relied on data assimilation for many decades (Kalnay, 2003; Navon 2009). In comparison, the use of data assimilation in atmospheric chemistry models to improve air quality forecasting is more recent, but important advances have been made. Tombette et al. (2009) presented an experiment on PM_{10} data assimilation using OI method to improve PM_{10} forecasting. Tang et al. (2015) used the same DA method to assimilate ozone/ $PM_{2.5}$ and MODIS aerosol optical depth data into the Community Multi-scale Air Quality model to improve the ozone and total aerosol concentration for the CMAQ simulation over the contiguous United States. Liu et al. (2011) assimilated AOD from Terra and Aqua satellites using the GSI 3D-Var assimilation system, showing that AOD data assimilation system can serve as a tool to improve simulations of dust storms. Li et al. (2013) and Feng et al. (2018) assimilated ground-based observations of $PM_{2.5}$ using 3D-Var to improve $PM_{2.5}$ forecasting. 4D-Var has been successfully implemented on CTMs and has improved the $PM_{2.5}$ forecasting capability. Zhang et al. (2016) constructed a GEOS-Chem adjoint model suitable for $PM_{2.5}$ pollution diffusion based on 4DVAR algorithm, which was verified by the monitoring data of APEC in Beijing in 2014. Zhang et al. (2021) built a $PM_{2.5}$ data assimilation system based on the 4DVar algorithm and the WRF-CMAQ model, which can assimilate synchronous observations simultaneously to improve aerosol prediction accuracy. Wang et al. (2021) established a 4D-VAR assimilation system based on GRAPES_CUACE to optimize black carbon (BC) daily emissions in northern China on 4 July 2016. EnKF also plays a significant role in improving the accuracy of atmospheric chemistry model forecasts. Lin et al. (2007) developed a EnKF system for a regional dust transport model. Tang et al. (2016) investigated a cross-variable ozone DA method based on an EnKF, for improving ozone forecasts over Beijing and surrounding areas. Park et al. (2022) developed a DA system for the CTM using the EnKF technique, where $PM_{2.5}$ observations from ground stations are assimilated to ICs every 6 hours to improve $PM_{2.5}$ forecasting in the Korean region. Peng et al. (2017) used EnKF to optimize ICs and emission input, resulting in significant improvements in $PM_{2.5}$ forecast. Overviews of these achievements were provided in several literatures (Bocquet et al., 2015; Benedetti et al., 2018; Zhu et al., 2018; Sokhi et al., 2022).

Although the previous studies have reported DA method using ground-based or satellite-retrieved observation led to improvement of atmospheric composition prediction, each of these DA methods has its own limitation. In OI and 3D-Var, the background error covariance (BEC) matrix is estimated at once and the prediction error is statistically stationary. 4D-Var and

EnKF are advanced data assimilation methods that provide the evolution of the forecast error covariance, but when they are employed in the operation use, each of them is facing their own challenge. 4D-Var requires coding the adjoint model, which is difficult to perform for complex systems. The CTM and CCMM are complex systems with rapid updates, and the implementation of 4D-Var requires a large workload of adjoint models coding (Ha, 2022). EnKF obtains a flow-dependent BEC using ensemble forecast by integrating the model multiple times, and that makes it approximately 100 times more computationally expensive than the forward model when applied to nonlinear systems (Counillon and Bertino, 2009). Notably, the problem of BEC forecasting in OI and 3D-Var is solved by EnKF using a Monte Carlo approach. EnKF obtains the forecast ensemble by integrating the model multiple times and uses the empirical covariance of the forecast ensemble to update all ensemble members to obtain the analysis ensemble. When the number of ensemble members tends to infinity, the empirical covariance of the forecast ensemble approximates the true value of the forecast error covariance. Compared with variational assimilation, EnKF has a flow-dependent BEC and does not require an adjoint model, which is an advantageous data assimilation method. Several studies applied EnKF to assimilate surface or satellite observations to improve the CTM model forecast accuracy. For example, Lopez-Restrepo et al. (2020) calibrated the spatial length scale of the covariance localization and the temporal length scale of the stochastic model for the emission uncertainty of EnKF and assimilated ground-level PM$_{2.5}$ and PM$_{10}$ data with an optimized assimilation system to improve the simulation and forecasting of PM$_{2.5}$ and PM$_{10}$ in a densely populated urban valley of the tropical Andes. Park et al. (2022) developed a DA system for the CTM using the EnKF technique, where PM$_{2.5}$ observations from ground stations are assimilated to ICs every 6 hours to improve PM$_{2.5}$ forecasting in the Korean region. Peng et al. (2017) used EnKF to optimize ICs and emission input, resulting in significant improvements in PM$_{2.5}$ forecast. However, EnKF also requires an appropriately sized ensemble. A small ensemble introduces significant sampling error, while an excessive ensemble consumes considerable computational resources.

For low-dimensional linear dynamical systems, standard techniques are used to achieve DA, such as KF, or the adjoint methods. However, the CTMs are strongly nonlinear systems, and the assumptions of Gaussian variables and non-biased do not apply. Advanced DA techniques such as 4D-Var, EnKF are approximately 100 times more computationally expensive than the forward model when applied to nonlinear systems(Counillon and Bertino, 2009). Moreover, the Coupled chemistry meteorology models (CCMM) are CTMs that simulate meteorological processes and chemical transformations jointly, with model computations far exceeding those of equivalent weather models, and it will be a great challenge to establish a real-time and operational DA system. Compared to EnKF, EnOI is a suboptimal method for ensemble-based assimilation (Evensen, 2003). EnOI uses a stationary ensemble to estimate the BEC and only one analysis field (AF) is updated at a time, which makes the computation time greatly reduced. EnOI is robust, flexible, portable, and inexpensive, and is not burdened with the technical difficulties that some other methods carry. EnOI can be used in conjunction with other DA methods and may be an appropriate choice for coupled forecast systems(Oke et al., 2010). EnOI has been widely used in ocean models with significant improvements to model forecast (Counillon and Bertino, 2016; Castruccio et al., 2020; Xie and Zhu, 2010; Belyaev et al., 2021), but not in the CTM or CCMM. actual operational use in CTMs is still relatively rare. To our knowledge, there are only several papers involved researches of EnOI in atmospheric chemistry models so far. Zhang et al. (2014) implement the EnOI

on an air quality numerical modelling MM5-STEM for the Pearl River Delta region in China. They found that EnOI produced the initial condition closer to the true situation, but they didn't investigate the effect of EnOI on forecast. Wang et al. (2016) used EnOI to investigate the possibility of optimally recovering the spatially resolved emissions bias of black carbon aerosol. Kong et al. (2021) applied EnOI to assimilate hourly surface observations of CO concentrations at 1107 sites over China in January 2015. They found that simulations with the updated emissions revealed a decrease bias of average CO concentrations at 349 independent validation sites from 0.74 mg m^{-3} to 0.01 mg m^{-3} and a reduction of the RMSE by 18%. Results from these papers showed that EnOI is a useful and computation-free method to reduce the errors of initial chemical condition or emissions. Since development of CCMM are fairly recent, EnOI have not applied for real-time CCMM yet. The GRAPES-CUACE is an online CCMM system developed by the China Meteorological Administration (Gong and Zhang, 2008; Zhou et al., 2008; Wang et al., 2010a). This model not only plays an important role in the scientific research on air pollution (Wang et al., 2015a; Wang et al., 2015b), aerosol-cloud interaction (Wang et al., 2018; Peng et al., 2022); and aerosols' weather feedback (Wang et al., 2010b; Zhang et al., 2022), but is also applied for the operational forecasting of air quality, Fog-Haze weather and dust storm in China (Wang and Niu, 2013; Liu et al., 2017). Very recently, this model system has been updated to a new version (GRAPES_Meso5.1/CUACE) with many improvements (Wang et al., 2022). In this study, we established a real-time EnOI chemistry initial fields PM$_{2.5}$ assimilation system for this new version of model,GRAPES_Meso5.1/CUACE to with assimilatinge PM$_{2.5}$ data from nearly 1500 ground stations in China into the model chemical initial fields to improve the model forecasts of the concentrations of PM2.5PM2.5 forecast accuracy and discuss the impact of assimilating PM$_{2.5}$ on visibility.

2 Methods and Data

2.1 EnOI The ensemble optimal interpolation algorithm

DA methods are algorithms for optimal estimation. They combine observations and model results and their respective statistical characteristics of errors to obtain a statistically optimal analysis value by minimizing the analysis variance. Based on Kalman filter theory, the sequential assimilation methods use Eq. (1) to update the state variables.

$$x^a = x^b + K(y - H(x^b)) \hspace{3cm} (1)$$ Where the model forecast and analysis are denoted as x^a and x^b, respectively, and the measurements are contained in $\mathbf{y}$. The observation operator $\mathbf{H}$ is the spline or other interpolations from the initial fields PM$_{2.5}$ to the observational space. $\mathbf{K}$ is the Kalman gain matrix. $\psi_i = (i = 1, \cdots, N)$ is an n dimensional model state vector representing members of the ensemble. N ensemble samples are combined in an ensemble A.

$$A = (\psi_1, \psi_2, \cdots, \psi_N) \in \Re^{n \times N} \hspace{3cm} (2)$$

$$\overline{A} = A E_N \in \Re^{n \times N} \hspace{3cm} (3)$$

Where $\overline{A}$ is the ensemble mean, and the ensemble anomaly A' is then defined as

165 EnOI

170 algorithm used in this study is based on the work of Evensen (2003). A brief recall of the EnKF and EnOI is given in this section. DA methods are algorithms that combine observations and model results and their respective statistical characteristics of errors to obtain a statistically optimal analysis value by minimizing the analysis variance. Based on Kalman filter theory, the analysis state ψ^a is determined by a linear combination of the vector of measurements y and the forecasted model state vector or background ψ^f, which is given by the equation (1) and (2),

175 $\psi^a = \psi^f + K(y - H\psi^f)$ —————— (1)

$K = PH^T[HPH^T + R]^{-1}$ —————— (2)

where K is Kalman gain matrix, P is the background error covariance matrix, H is the observation operator that relates the model state to the observation, and R is the observation error covariance matrix.

Now we define A is the matrix holding the ensemble members ψ_i

180 $A = (\psi_1, \psi_2, \cdots, \psi_N) \in \Re^{ndim \times N}$ —————— (3)

where N is the number of ensemble members and $ndim$ is the size of the model state vector

let $\bar{A}$ be the ensemble mean of A, then the ensemble anomaly A' is defined as

$A' = A - \bar{A}$ —————— (4)

The ensemble covariance matrix P can be defined as

185 $P = \frac{A'A'^T}{N-1} \in \Re^{ndim \times ndim}$ —————— (5)

the vector of measurements y need to be perturbed with its error as the following

$d_j = y + \varepsilon_j, \; j = 1, \cdots, N$ —————— (6)

which can be stored in a matrix as

$D = (d_1, d_2, \cdots, d_n) \in \Re^{m \times N}$ —————— (7)

190   where m is the number of measurements.

The EnKF analysis equation will be expressed as the following

$$A^a = A + A'A'^T H^T [(HA'A'^T H^T + (N-1)R)]^{-1}(D - HA) \quad (8)$$

The analysis includes updating each ensemble and need to run the model N times in every forecast cycle to calculate P, therefore tends to be computationally demanding and has limited use when the computer time is the main affecter to be

195   considered, especially in real-time operational forecast.

The EnOI analysis is computed with the ensemble covariance matrix P spanned by a stationary ensemble of model states sampled from a long-time integration. It is computed by solving an equation written as the following

$$\psi^a = \psi^f + \alpha A'A'^T H^T [(\alpha H A'A'^T H^T + (N-1)R)]^{-1}(y - H\psi^f) \quad (9)$$

where the scalar $\alpha \in (0,1]$ is introduced to allow for different weights on the ensemble versus measurements. As Evensen

200   (2003) pointed out, an ensemble consisting of model states sampled over a long time period will have a climatological variance which is too large to represent the actual error in the model forecast, and α which mainly depends on how the model forecast behaviours is used to reduce the variance to a realistic level and can be tuned for optimal performance. In this study, it is taken as 0.9 based on our experience. Through ~~Eq.(~~Eq. (9) the EnOI analysis updates only one model state at a time, so the computer time can be reduced by one or two orders of magnitude.

205   **2.2 The EnOI data assimilation system design**

Using a set of ensemble forecasts with finite number to calculate the BEC will suffer from sample error and cause imperfect estimation or even filter divergence (Houtekamer and Mitchell, 1998). There are two sorts of techniques to possibly solve this problem. One is the distance-dependent covariance localization, which is done by updating the analysis at all grid points with the multiplication of the BEC by a correlation function (Hamill et al., 2001). The other is done by updating the analysis at each

210   grid point simultaneously using the state variables and the observations in the local region centred at that point (Ott et al.2004). In our EnOI DA system, we use the second technique. First, we define the localization length-scale as L. For each model grid point, we find the observations within L which are called active observations, and then calculate the corresponding innovation. This localization effect on the analysis is illustrated in section 3.1 (Fig.3).

The observation error covariance matrix R is assumed to be diagonal here, that is, the observation errors are not correlated.

215   The diagonal elements of R are thus given by the sum of the measurement error variance ε_o^2 and representativeness error variance ε_r^2, following Elbern et al. (2007). The measurement error ε_o is assigned as 7.5% of observed value, and representativeness errors ε_r is formulated as $\varepsilon_r = \varepsilon_o \cdot \sqrt{\frac{\Delta x}{L_r}}$, where Δx is model grid resolution (10km in this study) and L_r is the characteristic representativeness length of the observation, defined as 2 km for urban sites, 10km for rural sites and 20km for remote sites, respectively.

220 Based on the ~~EnOI~~ Eq. (~~6~~9), we built the EnOI initial field PM$_{2.5}$ assimilation system, as shown in Fig. 1. The main procedures can be divided into pre-processing, analysis, and post-processing. Pre-processing involves the acquisition of observed data and ensemble samples. Analysis is the revised main module of EnOI where the main computational processes are performed. Post-processing firstly verifies the assimilation results using the validation observations which are not used in EnOI and then processes the results obtained from assimilation into model-readable chemical ~~ICs to preliminarily verify the assimilation~~

225 ~~results~~initial conditions. Compared with the traditional EnOI, the time-continuous model historical forecast~~samples~~ before the assimilation moment are selected as the ensemble samples for this study. The ensemble design is set to be as following: suppose the assimilation will be done at time t, first we evolve the model from the spin-up run at t-NΔt and integrate the model to time t (in our operational set up, Δt is 1 hour), therefore we get a time series of N hourly model forecast outputs A_{t-N+1}, A_{t-N+2}, ..., A_{t-1}, A_t. These hourly outputs before the assimilation time t form the N-number ensemble A, which can be used to calculate

230 the average $\overline{A}$ and anomalies A' and then the background error covariance matrices P is calculated. The BEC is stationary for a particular analysis moment, but it changes with the assimilation moment during a long assimilation period. Because background error covariance statistics are derived directly from forecasts and the DA scheme does not need to modify the original CCMM, EnOI is very easy to apply and very cost-free in term of computation time.

235 **2.~~2~~3 GRAPES_Meso5.1/CUACE**

In this study, the DA method EnOI was established for the latest updated version of the regional atmospheric chemistry model GRAPES_Meso5.1/CUACE developed by the China Meteorological Administration (Wang et al., 2022). ~~The model has been~~ ~~widely used to study dust and haze prediction, aerosol radiation, and aerosol cloud interactions (Wang et al., 2008; Wang et~~ ~~al., 2010a; Wang et al., 2010b; Wang and Niu, 2013; Wang et al., 2015; Zhou et al., 2012; Wang et al., 2018; Peng et al., 2020;~~

240 ~~Peng et al., 2021a; Zhai et al., 2018; Zhang et al., 2022).~~ The model system consists of two main components, which are called GRAPES_Meso and CUACE, respectively. GRAPES_Meso refers to a real-time operational weather forecasting model used by China Meteorological Administration (Chen et al., 2008; Zhang and Shen, 2008). Now, the new version of it has been established with the resolutions ranging from 3 to 10 km for regional forecast (Shen et al., 2020). It uses fully compressible non-hydrostatic equations as its model core. The vertical coordinates adopt the height-based, terrain-following coordinates,

245 and the horizontal coordinates use the spherical coordinates of equal longitude–latitude grid points. The horizontal discretization adopts an Arakawa-C staggered grid arrangement and a central finite-difference scheme with second-order accuracy, while the vertical discretization adopts the vertically staggered variable arrangement. The time integration discretization uses a semi-implicit and semi-Lagrangian temporal advection scheme. The transport and advection processes for all gases and aerosols are calculated by the dynamic framework of it. The second component, CUACE, refers to the

250 atmospheric chemistry model (the Chinese Unified Atmospheric Chemistry Environment model), which mainly includes three modules: the aerosol module (CAM), the gaseous chemistry module (RADM2) and the thermodynamic equilibrium module

(ISOPIA).~~For the dynamic frame, the model uses a full compressible non-hydrostatic model core, an Arakawa C staggered grid, an improved material advection scheme of the Piecewise Rational function Method(Peng et al., 2005), and a height-based terrain following coordinate. The chemical module uses the CUACE, which consists of an emission inventory system, CAM aerosols module, Regional Acid Deposition Model (RADM2) and gases-particles transformation-related processes.~~ In the RADM2 module, 63 gas species through 21 photochemical reactions and 136 gas-phase reactions participate in the calculations. CAM module considers the dynamic, physical and chemical processes of aerosols including hygroscopic growth, dry and wet depositions, condensation, nucleation~~,~~ vertical mixing, cloud chemistry, and coagulation and activation of cloud condensation nodules from aerosols. ~~etc (Gong and Zhang, 2008)~~. Seven types of aerosols (sea salt, sand/dust, black carbon, organic carbon, sulfate, nitrate, and ammonium salt) are considered in the CAM. The aerosol size spectrum (except for ammonium salt) is divided into 12 bins with particles radium of 0.005–0.01, 0.01–0.02, 0.02–0.04, 0.04–0.08, 0.08–0.16, 0.16–0.32, 0.32–0.64, 0.64–1.28, 1.28–2.56, 2.56–5.12, 5.12–10.24, and 10.24–20.48 μm. The interface program that connects CUACE and GRAPES_Meso transmits the meteorological fields calculated in GRAPES_Meso and the emission data processed as needed to each module of CUACE. GRAPES_Meso and CUACE are online fully-coupled (Peng et al., 2021; Zhang et al., 2022).

2.~~3~~4 Data ~~used~~

Based "Ambient air quality standards" (GB 3095-2012) of China, the mass concentration limit of $PM_{2.5}$ and its corresponding air quality level and air pollution index (API) are shown in Table 1. Haze is defined as a weather phenomenon caused by air pollution when visibility is less than 10km, according to "Observation and forecasting levels of haze" (QX/T 113-2010) of China. Three pollution episodes occurred in China in December 2016, with the most severe haze episode occurring in China ~~A severe haze episode occurs in Northern China~~ from 1~~6~~5 to 2~~1~~3 December 2016 (see more details in Wang et al. 2022, Table 3). During this pollution episode, the highest daily $PM_{2.5}$ concentration peaks 600 μg m^{-3} in Shijiazhuang and some other cities, reaching the severely polluted level (250-500 μg m^{-3}). In this study, 15-23 December ~~this regional haze episode~~2016 was selected as the main study period, and both model input data and observation data used in this study are within this ~~period~~month. Model input data include anthropogenic emission data, model meteorological initial and boundary data. The emission inventory used in this study is from the Multi-resolution Emissions Inventory for China (MEIC) in December 2016 (http://www.meicmodel.org/). The emission inventory covers power plants, industry (cement, Iron and steel, industrial boilers, petroleum industry), residential, transportation, solvent use and agriculture, in-field crop residue burning~~etc~~. National Centers for Environmental Prediction (NCEP) Final analysis (FNLs) data (https://rda.ucar.edu/datasets/ds083.3/) are used for the model's initial and 6 h meteorological lateral boundary input fields. The observations include $PM_{2.5}$ and visibility. Nearly 1500 ground-based hourly $PM_{2.5}$ (μg m^{-3}) observations from the Chinese Ministry of Environmental Protection, with the detailed location and spatial distribution of the stations shown in Fig. ~~1~~2. The hourly meteorological automatic ground-based visibility data (km) were obtained from the China Meteorological Administration. The time format of these observations is processed to UTC and all the observational data are obtained after quality control and rechecked before use.

2.~~4~~5 Experimental Setup

285 The horizontal resolution, time step, forecast length and model domain of the GRAPES_Meso5.1/CUACE model are optional. In this study, the horizontal resolution of the model is 0.1°×0.1°, the time step is 100 s considering model integration stability and accuracy, and the model domain is15-60° E, 70-145° N and (grey dashed box in Figure 2). There are 49 model layers ascending vertically from the surface to 31km in height. The model warm restart time is 0000 UTC and 1200 UTC, and the forecast length is 24 hours. The ~~chemical initial field of the model warm restart uses the 24-hour forecast field of the day before~~

290 ~~the model, or the chemical initial field assimilated by EnOI, and the~~ model simulation results are output on an hourly basis. Three groups of experiments were performed in this study: one set of control experiments (CR), one set of sensitivity experiments and one set of cyclic DA experiments, as shown in Table 1. CR00 is the control experiment representing model run without DA begin at 0000 UTC every day and forecast 24 hours~~the model with a daily warm restart at 0000 UTC and without DA~~ (the initial field is the previous day's 24-hour forecast field), simulated from ~~9~~1 to ~~23~~31 December 2016. CR12 is

295 also model run without DA but begins at 1200 UTC every day and forecast 24 hours. The localisation length-scale L and the ensemble size N are the key parameters affecting EnOI. Based on CR00, two parallel sensitivity experiments were designed to study the impact of localisation length-scale and ensemble size on the assimilation effects. The chemical initial fields, ensemble samples for the sensitivity experiments were obtained from the CR00. The first group of sensitivity experiments is fixed with ensemble size N of 48, and length-scale is selected for 20, 40, 60, 80, and 100 km to investigate the impacts of

300 different localization length-scale choices on the optimized chemical initial field; the second group is fixed with length-scale L of 80 km, and the 24, 48, 72, 96, 120, and 144 simulations before the assimilation moment (0000 UTC) were selected as ensemble samples, respectively, and the effect of the number of ensemble samples on the assimilation effect was discussed. ~~Considering that the missing localization could break the model dynamical balance(Oke et al., 2007), localization was performed in selecting the optimal ensemble size.~~

305 To investigate the impact of the assimilation moment on the forecast fields, the optimal length-scale and ensemble size were selected based on the results of sensitivity experiments, and two sets of cyclic DA experiments, DA00 and DA12, were set up to represent the daily assimilation of the initial fields at 0000 UTC and 1200 UTC, respectively. The N hourly model forecasts before the assimilation moment were used as the ensemble samples to approximate the BEC, and the analysis increments are calculated by combining the model forecasts and PM$_{2.5}$ observations at 0000 UTC and 1200 UTC, and the ~~revised AF~~analysis

310 are used as the chemical initial fields for the next forecast to achieve cyclic DA. ~~To avoid errors caused by different warm restart times, the CR12 control experiment is set up to represent a daily warm restart at 1200 UTC but without assimilation to be used as a reference experiment for DA12.~~

3 Result and discussion

3.1 Localiszation length-scale sensitivity experiments

315 ~~The aspect of localisation needs to be adapted for specific applications~~The localization effect on the analysis is illustrated firstly and~~, so~~ two observation sites A (114.5° E, 38.0° N), and B (36.6° N, 116.9° E) were selected to perform a length-scale single-point experiment for the initial field at 0000 UTC on 15 December 2016, corresponding to the left and right columns of the analysis increments ($\psi^a - \psi^f$) shown in Fig. 3. The analysis increments are determined by both the observation increments and the BEC based on Eq. (~~6~~9). As shown in Fig. 3, the increments are positive in the left and negative in the right column,

320 which represent the underestimation of $PM_{2.5}$ concentration at site A and overestimation at site B before being assimilated. As the length-scale increases, the range of analysis increment expands, and the number of model grids that can be affected increases gradually. Due to the sparse distribution of $PM_{2.5}$ sites, if the localization length-scale is too small, most of the model grids cannot be updated, which reduces the assimilation efficiency; whereas if the localisation length-scale is too large, the analysis increments between distant sites will offset and superimpose, creating fake increments. With the experiments using

325 length-scale of L = 80 and 100 km, a small negative analysis increments are found at site A in the southeast direction. Compared to site A, a wide positive analysis increments that do not match the actual situation are found at site B in the west direction for experiments using L=60, 80, and 100km. ~~Clearly too large a localization radius can lead to error increments.~~ It is worth noting that there are differences in the shape of the analysis increment fields at sites A and B, which is related to the EnOI having a flow-dependent BEC, the details of BEC will be discussed in 3.2.

330 Ground-based $PM_{2.5}$ sites are established according to the population and economic development level of the region, and are not evenly distributed, such as Beijing, Shanghai, Guangzhou, and other economically developed and populous megacities, which have a high density of $PM_{2.5}$ sites, while the western and central regions of China are sparsely populated, and the sites are partially sparse. So, in order ~~T~~to obtain the statistically optimal localization length-scale, we performed assimilation experiments on the initial fields at 0000 UTC each day from 1 to 31 December. 50% of $PM_{2.5}$ sites were randomly selected as

335 DA sites, and the rest were used as verification sites (without DA), and the blue and brown sites shown in Fig. 2 represent the spatial distribution of verification and assimilation sites, respectively. ~~the initial fields from 15 to 23 December 2016 were performed for sensitivity experiments, respectively, and t~~The statistics results of verification sites against the observation are shown in ~~Fig. 4~~Table 3. ~~The more the scatter points are clustered on the diagonal line means that more the simulation is closer to the observation.~~ Compared to the CR, ~~the scatter distribution of the DA experiments is closer to the diagonal, with~~

340 Correlation Coefficient (CORR) of DA for verification sites increase from 0.65 to 0.77 at least, and the Root Mean Square Error (RMSE), Mean Bias (MB), and Mean Error (ME) of the DA experiment are smaller than those of the CR. The statistical data are different for different Localisation length-scale, indicating that localization can have an effect~~ively~~ on the assimilation. Compared with the CR, RMSE of DA decreased from 60.1 to 41.6 µg m^{-3}, MB decreased from 8.5 to 3.2 µg m^{-3} and ME decreased from 41.73 to 25.9 µg m^{-3} for the localization length-scale selection of 40 km,~~When localisation is used with a 40-~~

345 ~~member ensemble and length-scale of 40km, the RMSE is 18.472 µg m^{-3}, ME is 10.481 µg m^{-3}, and CORR is 0.977,~~ which is

the best among all the experiments on different length-scale. Localisation length-scale of 60km and 80km have similar statistics results, but the statistics of 20km and 100km are not very good. For experiments with length-scale of 20, 40, 60, 80 and 100 km MB is 2.656, 1.973, and1.928, 2.194, 1.985 µg m⁻³, respectively. Ground-based PM$_{2.5}$ sites are established according to the population and economic development level of the region, and are not evenly distributed, such as Beijing, Shanghai,

350 Guangzhou, and other economically developed and populous megacities, which have a high density of PM$_{2.5}$ sites, while the western and central regions of China are sparsely populated, and the sites are partially sparse. Using a localization length-scale of 20 km prevents most of the model data from being updated while using too large a length-scale allows remote sites to interact with each other and produce more spurious increments. In addition, from the meteorological conditions, heavy pollution weather is always characterized by small or static winds, pollutant transport over small distances, an observation site

355 represents a limited spatial extent, so a larger localized length-scale setting may also not produce an very realistic unreasonable initial field. In summary, we concluded that the best assimilation effect can be achieved From this sensitivity experiment, we find that when the localization length-scale is using from 40 km to 80km, the statistics are relatively good and the optimal assimilation effect can be achieved.

3.2 Ensemble size sensitivity experiments

360 We repeat the series of experiments presented in Fig. 3, but with a localising length-scale of 80 km and 24, 48, 72, 96 and 120 ensemble members. Figure 54 shows a map of BEC CORRcorrelation field between observation sites (A, B) and BECs for different ensemble size, overlaid with the 0000 UTC surface wind vector of 15 December 2016. Site A is controlled by strong north and northwest winds, which makes the rCORR field show a northeast-southwest trend; The wind speed at site B is less than 5 m.s⁻¹ in all directions with a steady state, so the CORR field is approximately distributed in concentric circles nearby

365 the center of the site. As the number of ensemble samples increases, the area of positive CORR greater than 0.7 gradually increases in A and B. the range of positive CORR at sites A and B gradually increases with the range of CORR greater than 0.7. The ensembles of size N =24 or N=48 can be considered small compared to the selection of other ensemble sizes in sensitivity experiments. In this case, the CORRs between the observation sites and the surrounding large-scale areas are all greater than 0.7, and an extremely strong negative correlation is found in the southwest, which exaggerates the correlation of

370 each area. The success of ensemble-based DA systems depends strongly on the number of samples. The smaller ensemble size fails to accurately estimate the BEC and is prone to sampling error, resulting in an overestimation or underestimation of the initial fieldunreasonable results, and Natvik and Evensen (2003) investigated the effect of the number of samples on assimilation and showed that an ensemble of fewer than 60 samples reduce the performance of assimilation. When the hourly model forecasts of over 5 days (N=120) before assimilation are selected as the ensemble samples, the correlations of both sites

375 A and B with the BECs in a wide area become positive.

Next, all PM$_{2.5}$ sites were used to assimilate the initial field at 0000 UTC per day for this pollution episode, and six different ensemble sizes were used to improve the initial field as shown in Table 2. Compared with the unassimilated initial field, the RMSE, CORR, MB, and ME of the initial field after assimilation changed significantly. With only 24 ensemble samples

assimilated, the RMSE rapidly decreased from 71.776 to 20.675 µg m^{-3}, and the CORR directly increased from 0.650 to 0.97, meanwhile the ME and MB were 1⁄4 and 1⁄9 of the original. When 120 samples were selected for assimilation, the analysis field PM$_{2.5}$ statistics were worse than those of fewer samples (N=24, 48, 72, 96, 144). The RMSEs for 24, 120, and 144 samples were 20.675, 23.919, and 23.416 µg m^{-3} respectively, and the CORRs were also less than 0.96, and the RMSE results for 48, 72, and 96 ensemble samples are 19.170, 18.908, and 18.849 µg m^{-3} respectively. The DA sites were used to assimilate the initial field at 0000 UTC per day for December 2016, and six different ensemble sizes were used to improve the initial field as shown in table 4. Compared with the initial field without data assimilation, the RMSE, CORR, MB, and ME of the initial field after assimilation had all been improved and the improvement were different depending on the ensemble size. The priori initial field is shown in table 4 "CR". With only 24 ensemble samples assimilated, the RMSE of verification sites decreased from 60.1 to 48.6 µg m^{-3}, the CORR increased from 0.56 to 0.76, and the MB and ME are decreased from 8.5 to 4.2 µg m^{-3} and 41.7 to 30.8 µg m^{-3}, respectively. As seen in table 3 the statistics of verification sites become progressively better as the ensemble members increases from 24 to 48, 72, and 96. The verification sites RMSEs for 48, 72, and 96 samples are 44.9, 42.4 and 40.70 µg m^{-3} respectively, the CORRs are 0.80, 0.81, 0.82, and the MEs are 27.0, 25.9, and 25.7. When 120 samples or 144 samples were selected for assimilation, the analysis field PM$_{2.5}$ DA and verification statistics were not better than those of 96 samples. The verification sites RMSEs for 120, and 144 samples were 44.1, and 45.8 µg m^{-3} respectively, and the CORRs also became smaller. The differences between the statistics also indicate there is an optimal ensemble size, the RMSE of the experiment using 96 samples is smaller than the RMSE when using the other ensemble sizes, and the remaining statistics are better than the results when other samples are selected, so we consider that in this sensitivity experiments the best assimilation is achieved when the number of ensemble size is 96. It is noted from the experimental results that not the larger the ensemble, the better the results in this study. It could be influenced by the following reasons that the atmospheric chemistry model used in the study is coupled online with the mesoscale regional weather model GRAPES_Mese5.1. The mesoscale regional weather model differs from the climate model and the global model in that the mesoscale model represents weather systems on time scales of one day to several days(Emanuel, 1986). In addition, atmospheric chemical processes are fast-varying processes with small time scales compared to climatic and oceanic processes, so using model results from long-time integrations as ensemble may average out the "error of the day" and will not be a very good assessment of model background errors.

3.3 Impact on initial fields

The optimal localization length-scale, ensemble size of 40 km and 96 were obtained by sensitivity experiments, respectively. In order to verify the assimilation effect and evaluate quantitatively the impact of the EnOI system on the initial fields, DA experiments with length-scale of L=40 and ensemble size of N= 96 was performed on the initial field at each 0000 UTC from 15 to 23 December 2016. The assimilated observations were obtained from the DA sites in Fig. 2, and the effect on both DA sites and verification sites are evaluated. Figure 5 shows the statistics for the two regions of the initial field, the China mainland, and North China. In China mainland, the CORRs of the verification sites and DA sites before assimilation were 0.60 and 0.58,

respectively, and the RMSEs were 73.9 and 73.4 µg m^{-3}, respectively. After the DA sites were assimilated, the CORR of assimilated sites increased to 0.99 and the RMSE decreased to 14.5 µg m^{-3}, and the CORR of verification sites increased to 0.84 and the RMSE decreased to 46.4 µg m^{-3}, meanwhile the ME changes from 49.7 to 27.3 µg m^{-3}. In North China, after the DA sites were assimilated, each statistic of the validation site also changed, with the CORR increasing from 0.53 to 0.87, RMSE decreasing from 105.5 to 65.7 µg m^{-3}. Only 50% of the ground-based observation are assimilated and the statistics of the validation sites also have been improved. This experimental results prove that the DA system can indeed yield more accurate initial field with over 40% increase of CORR and 37% reduction of RMSE, ~~firstly, an independence test with length scale of L=40 and ensemble size of N= 96 was performed on the initial field of 0000 UTC on 15 December 2016. The 50% of PM$_{2.5}$ sites were randomly selected as DA sites, and the rest were used as verification sites (without DA), and the blue and red sites shown in Figure 2 represent the spatial distribution of verification and assimilation sites, respectively. As shown in Figure 6, the CORRs of the verification sites and DA sites before assimilation were 0.441 and 0.470, respectively, and the RMSEs were 59.914 µg m^{-3} and 62.783 µg m^{-3}, respectively. After the DA sites were assimilated, the CORR of assimilated sites increased to 0.977 and the RMSE decreased to 14.140 µg m^{-3}, also the CORR of verification sites increased to 0.734 and the RMSE decreased to 46.041 µg m^{-3}. It is obvious that the assimilation corrected the initial PM$_{2.5}$ concentration significantly, especially in the region underestimate, and assimilating not only produces mainly localized increment structures concentrated around the measurement sites, but also affects other areas.~~

~~As in sensitivity experiments, all PM$_{2.5}$ sites were used as DA sites to assimilate the initial fields in the CR00 experiment separately, but with a localization length scale of 40 km and an ensemble size of 96.~~ To ~~understand~~ illustrate the assimilation effect of different pollution levels, we consider this episode from 15 to 23 December 2016, in which the first two days as the pollution start period, days 3 to 7 as the pollution period, and the last two days as the pollution dissipation period. We compared the PM$_{2.5}$ observations and initial conditions before and after DA within all the observations sites assimilated during this episode. This was done to understand the impact of DA for initial conditions in the system's actual operating situation. ~~In this section, we show in~~ Fig. ~~7~~6 shows the spatial distribution of PM$_{2.5}$ in the observation field (OB), background field (BF), analysis field (AF), and analysis field increments (AFI) for two days of light pollution (16 and 23 Dec.) and two days of heavy pollution (19 and 20 Dec.). The black boxed area in Fig. ~~7~~6 is the same as North China (NC) in Fig. 2, including Beijing, Tianjin, eastern Shanxi, southern Hebei, western Shandong, and northern Henan, which has the highest simulated PM$_{2.5}$ concentration. Table 5 summarizes the corresponding statistics of initial PM$_{2.5}$ concentrations for assimilation sites and verifications sites before and after EnOI. ~~Fig. 6 shows that, C~~compared with OBs, the model background ~~PM$_{2.5}$~~ without DA can capture the spatial pattern of distribution over China in general~~ly~~ which shows that the model performance is moderate good. However, there are still errors between the background and observations. PM$_{2.5}$ concentrations are overestimated in NC and eastern China during the pollution start and dissipation periods. During the heavy pollution period, the ~~BFs~~ background PM$_{2.5}$ concentrations are overestimated in northeast China and underestimated in NC. After assimilating the ~~BFs~~ ground-based PM$_{2.5}$, the PM$_{2.5}$ concentration increments were distributed around the observation sites as expected and ~~distribution changes from sheet-like to discrete,~~ were more closer to the observations distributions. ~~which is due to the update of the model data in~~

a length-scale of 40 km range with the distribution of observation sites, resulting in the adjustment of the PM$_{2.5}$ in the BFs. Negative values of the AFI demonstrate that assimilation reduces PM$_{2.5}$ concentrations, while positive values demonstrate that assimilation increases PM$_{2.5}$ concentrations. During the period before and after pollution, PM$_{2.5}$ concentrations decrease in eastern China and increase in western China and NC, indicating a reduction in over- or under-prediction of model PM$_{2.5}$

450 concentrations after assimilation. indicating that the reduction of the overestimation or underestimation of the model simulation over these regions with data assimilation. Table 5 shows that assimilating 50% of the ground-based observations improved the initial condition for other areas where have no assimilated sites. Take 19 Dec. 2016 as an example, the CORR for verification sites increased from 0.66 to 0.85, RMSE decreased from 79.2 to 56.1µg m^{-3}, and MB and ME also became smaller after EnOI. These results indicate that the initial PM$_{25}$ fields can be adjusted efficiently by EnOI. What is the impact of this innovation

455 through the EnOI system for PM$_{2.5}$ forecast is discussion in the next section.

To evaluate quantitatively the impact of the ensemble assimilation system on the initial fields, the RMSEs, MEs, MBs and CORRs of the assimilated initial fields and the BFs were first analysed. Table 3 shows the statistics for the two regions of the initial field, the China mainland (Total) and NC (Contains observation sites in NC). In China mainland, ME, and RMSE decreased by 75.53%, 72.33%, respectively, and CORR increased to 0.967. The MB changes from negative to positive,

460 meanwhile the MB in mainland China becomes larger. In NC, MB decreased by 109.63%, ME decreased by 79.59%, RMSE decreased to about 20 µg m^{-3}, and CORR increased by 148.59%. The results show that the correction effect of DA on the initial fields is evident.

3.4 Impact on forecast fields

3.4.1 Impact on PM$_{2.5}$ forecast fields

465 In this section, we will investigate the performance of assimilating the initial field at 0000 UTC per day (DA00) or 1200 UTC per day (DA12) on improving the PM$_{2.5}$ forecasts, with an example in North China (Figure 8), and the same study period as in section 3.3 was selected, DA00 (Figure 8 First and second rows) and DA12 (Figure 8 third and fourth rows) were performed in parallel. In NC, compared with the observations (brown line with circles), the forecast PM$_{2.5}$ concentrations (black line, grey line) are 20 to 100 µg m^{-3} higher in the pollution start period (16 December) and the pollution fading period (23 December),

470 lower in the pollution period (19 December), and relatively consistent in (20). The daily trend of PM$_{2.5}$ changes immediately one hour after 0000UTC in DA00 (blue line) or 1200UTC in DA12 (pink line), and the assimilation forecast value rapidly approaches the observations, after that the DA experiment gradually overlaps with the CR experiments in the context of daily changes of emission. It can be seen from Fig. 8 that the RMSEs of the DA experiments (blue and pink lines marked with squares) are always lower than that of the CR experiments (black and grey lines marked with squares), which proves that

475 assimilating the initial field improves the PM$_{2.5}$ forecast field throughout the assimilation time window, yet the assimilation impact is strongest in the first 12 hours. The comparison reveals that the assimilation effect is related to the choice of the assimilation moment, and the blank area between the RMSE line of the DA experiment and the RMSE line of the CR

In this section, we will discuss the impact of assimilation observations on PM$_{2.5}$ forecasts. As in Section 3.3, we assimilate the DA sites at 0000 UTC from 15 to 23 December 2016, and then analyse the following forecast of DA and the verification sites separately. The RMSE of the DA and verification sites in China mainland and North China for a complete pollution process obtained average over 15 to 23 December 2016 is shown in Fig. 7. For the DA sites in China mainland (Fig.7a), the model forecast RMSE without DA is about 75 µg m^{-3}, after the assimilation, the model forecasts RMSE is decreased rapidly from 75.4 to 40.1µg m^{-3}, which is an over 40% reduction. This implies that assimilation with EnOI can considerably improve the forecast accuracy. Meanwhile, it is notable that assimilation of DA sites also has an impact on the forecast at the verification sites. The trend of the RMSE series at the verification site is consistent with the DA site, but smaller in values. The RMSE of verification sites at 1h forecast hour dropped from 75.5 to 51.0 µg m^{-3}, about 32% reduction. For North China, which was shown in Figure7c and d, the model forecast RMSE without DA is about 115 µg m^{-3}. After the assimilation of PM$_{2.5}$ observation, the model forecasts RMSE of DA sites at 1h forecast time is decreased rapidly from 122 to 56.1µg m^{-3}, which is an over 54% reduction. For verification sites, the reduction amplitude is 33%, smaller than that of DA sites, but still a moderate improvement considering only 50% ground-based observations were used to be assimilated at 0000UTC. The results show that assimilation with EnOI not only improve the forecast for the DA sites but also the verification sites. The improvements are mainly within

the first 12~~24~~h forecasts with an RMSE greater than 10 µg m^{-3}. The improvement receded with forecast time, changing from 46% at 1h forecast hour to 7% at 24h forecast hour. These results are consistent with previous study, which either used 4DVAR or EnKF (Wu et al. 2008; Bocquet et al, 2015, Park et al.,2022). As Bocquet et al. (2015) pointed out, even with the improved analysis, the impact of initial state adjustment is generally limited to the first day of the forecast, for pollutant transport and transformation are strongly driven by uncertain external parameters, such as emissions, deposition, boundary conditions, and meteorological fields.

Now we use all ground-based observation sites as DA sites to investigate the performance of assimilating the initial field at 0000 UTC per day (DA00) or 1200 UTC per day (DA12) on improving the PM$_{2.5}$ forecasts. DA00 and DA12 were performed in parallel. The daily average of the 24-hour RMSE was obtained for the DA and CR experiments. Rate of improvement (ROI) by data assimilation in 1d (24h) predictions for 15 to 23 December 2016 for Mainland China and NC were calculated using the ratio of the reduced RMSE statistical metrics to those for the CR simulation and plotted in a daily time series histogram as shown in Fig. 8. In this episode, the improvement of China mainland PM$_{2.5}$ forecasts by DA00 and DA12 are minimum at 9% and 10% respectively on December 15 and maximum at 15% and 21% respectively on December 19. The minimum and maximum improvement of assimilation on PM$_{2.5}$ forecasts in NC both appear in DA12, which are 4% and 25%, respectively. The difference between DA12 and DA00 relative RMSEs is mostly positive, within 6% in China mainland, but in NC this difference can be up to 15%. The average RMSE improvement of 24h forecast for China mainland and the North China assimilation at 0000 UTC is 12.3% and 9.8%, respectively, while that at 1200 UTC is 14.4% and 14.0%, respectively. In terms of the average relative RMSE for this episode, assimilating the initial field at 1200 UTC improves the PM$_{2.5}$ forecast more than 0000 UTC, mainly because the model forecasts are not close to the observations at 1200 UTC in most cases, thus choosing this time for assimilation will have a significant impact. In addition, the DA effect varies for each day and the larger the error, the greater the improvement in RMSE from DA, which means that the larger the a priori error, the greater the improvement from DA. These results show that using EnOI to assimilate ground-based PM$_{2.5}$ observations for the model chemical initial field can reduce over 9.8% of RMSE for 24h forecast in average. Park et al. (2022) implemented an ensemble Kalman filter in the Community Multiscale Air Quality model (CMAQ model v5.1) for data assimilation of ground-level PM$_{2.5}$. They found using EnKF with 40 ensemble number can reduce 9.6% of RMSE for 24h forecast. Comparing their results with ours, we can find that, while EnOI is sub-optimal, it can give improvement of forecast that are comparable to those of the EnKF. Moreover, the computational cost of EnOI is typically about N times less than that of EnKF. Therefore, we suggest that EnOI may provide a practical and cost-effective alternative to the EnKF for the applications where computational cost is a main limiting factor, especially for real-time operational forecast.

To achieve better performance of assimilation, we update the initial field every 12h. Figure 9 gives time series of forecasts and observations in term of PM$_{2.5}$, together with RMSE of CR and DA for North China. Compared with the observations, the forecast PM$_{2.5}$ concentrations are 20 to 100 µg m^{-3} higher in the pollution start period (15-17 December) and the pollution fading period (21, 23 December), about 100 µg m^{-3} lower on December 19. The PM$_{2.5}$ concentrations changes immediately one hour after 0000 UTC or 1200 UTC. It can be seen from Fig. 9b, the RMSEs of the DA experiments are always lower than

545 that of the CR experiments, and the difference in RMSE between the CR and DA experiments receded with forecast time. This proves that assimilating the initial field can improve the PM$_{2.5}$ forecast. Note that the DA algorithm used here cannot produce an optimal solution when there are larger errors in the model. On 19 December 2016, even with DA the model still cannot retrieve the true variation very well for the first 12 hour forecast. This suggest that using DA on initial field can only partially remedy inherent model error. To improve the analysis capabilities and prolong the impact of DA on PM$_{2.5}$AQ forecasts, we

550 should extend the assimilation for adjusting emissions, meteorological field and other model uncertainty sources.

3.4.2 Impact on Visibility forecast fields

The occurrence of low visibility episodes is usually associated with aerosol pollution. The horizontal spatial distribution of the OBs, forecast fields without assimilation (CR), forecast fields with assimilation (DA), and incremental fields (DA-CR) for

555 visibility and PM$_{2.5}$ at 0100 UTC on 16 and 20 December are shown in Fig.10. During the pollution start period (16 December 0100 UTC) visibility is above 10km in most of China, and during the pollution period (20 December 0100 UTC) visibility is mostly below 7km in eastern China. After assimilating the ground-based PM$_{2.5}$, the visibility distribution of DAs becomes discrete more consistent with the observation compared to the CRs. A positive PM$_{2.5}$ concentration increment corresponds to a negative visibility increment, that means that when the PM$_{2.5}$ concentration increases, the visibility decreases at the same

560 moment. At 0100UTC On 16 December, the CR PM$_{2.5}$ concentration is underestimated in NC and overestimated in Southeast China, and after assimilating PM$_{2.5}$, the visibility is reduced in NC with increased PM$_{2.5}$ and increased in Southeast with reduced PM$_{2.5}$. In the period of light pollution, the absolute value of visibility increment is mostly in the range of 5-7 km when the PM$_{2.5}$ increment is from 30 to 110 µg m^{-3} or from -30 to -110 µg m^{-3} in NC, while in the pollution period (20 December 0100 UTC for example), under the same PM$_{2.5}$ analysis increment, the visibility increment in NC is between -3 and 3 km. It

565 proves that visibility is more correlated with PM$_{2.5}$ concentration when the pollution is lighter, while they are less correlated when the pollution is heavier, which is consistent with the findings of Yu et al. (2016) and Yadav et al. (2022).

Four stations, Beijing (BJ), Shijiazhuang (SJZ), Xingtai (XT), and Jinan (JN), were selected from the heavily polluted NC to study the effect of assimilating the initial field PM$_{2.5}$ on the visibility forecasts. Since the assimilation effect is most obvious in the first 12 hours, we focus on the improvement of visibility forecasts within 12 hours. Figure 11 shows the observation

570 (orange line), simulation (black dashed line) and assimilation (green line) of visibility and observation (grey line), simulation (black line) and assimilation (blue line) of PM$_{2.5}$ concentration for the above cities from 0100 to 1200 on 16 and 20 December 2016. On 16 December, when PM$_{2.5}$ concentration is less than 300 µg m^{-3} (December 16), visibility at all four stations is closer to the observed value by assimilating PM$_{2.5}$, among which BJ and JN have decreased PM$_{2.5}$ concentration after assimilation, and visibility has increased at the same time. SJZ and XT have increased PM$_{2.5}$ concentration and decreased visibility after

575 assimilation. In the period of low PM$_{2.5}$ concentration, about 100 µg m^{-3} PM$_{2.5}$ change makes visibility change 11km, 4km, 5km and 7km in BJ, SJZ, XT and JN respectively. In the period of heavy pollution, PM$_{2.5}$ concentration change 150 µg m^{-3} in Beijing and Shijiazhuang at 0100UTC, while visibility change 3.5km and 0.5km respectively. It is obvious These result show

that the improvement of visibility by assimilating $PM_{2.5}$ is limited during the heavy pollution period. It is worth noting that when the $PM_{2.5}$ concentration is greater than 350 μg m^{-3} at the JN site, although the decrease of $PM_{2.5}$ concentration corresponds
580 to the increase in visibility, the gap between the assimilated visibility and observation becomes larger at this time, which may be related to the inaccuracy of the humidity simulation here and inaccurate visibility parameterization scheme for the model. Visibility is not linearly related to $PM_{2.5}$, and visibility is also affected by humidity and other factors. Assimilation of the initial field $PM_{2.5}$ can improve the visibility forecast, but if we want to improve the visibility forecast significantly, ~~other objects of assimilation, such as PM_{10}, humidity, etc., need to be considered.~~ we need to improve not only the visibility parameterization
585 scheme, but also the humidity accuracy.

4 Conclusions

To improve the accuracy of $PM_{2.5}$ and visibility forecasting in China, a real-time and efficient EnOI assimilation system is established for the latest online operational chemistry weather model GRAPES_Meso5.1/CUACE of China Meteorological Administration. The ground-based $PM_{2.5}$ observation data nearly 1500 surface stations covering the whole country are used
590 for assimilation. $PM_{2.5}$ and visibility simulation-assimilation experiments were performed for a haze pollution episode from 15 to 23 December 2016. Parallel sensitivity experiments of localization length-scale and ensemble size were set up to determine two key parameters that influence the effectiveness of EnOI assimilation. Based on the results of sensitivity experiments, the initial fields were assimilated at 0000 UTC each day from 15 to 23 December 2016 to study the improvement of EnOI on the initial field $PM_{2.5}$. In addition to the analysis of the China mainland assimilation effect, the heavily polluted
595 North China was additionally divided to discuss the different impacts of assimilation on the overall and regional chemical initial fields. Cyclic assimilation experiments were performed at 0000 UTC (DA00) and 1200UTC(DA12) to investigate the impacts of assimilation on the forecast fields, taking NC as an example, to discuss the impacts of assimilation on $PM_{2.5}$ and visibility forecast fields.

The optimal localization length-scale and the number of ensemble samples are 40 km and 96, respectively, derived from
600 sensitivity experiments. ~~The DA can significantly improve the model initial field, the AFs $PM_{2.5}$ is more consistent with the observed results in both distribution and values. The AFs relative to the background fields (BFs) in China mainland, NC the ME decreased by almost 80%, the RMSE decreased by 72.33%, 75.53%, the CORR increased from 0.584 to 0.967, and 0.319 increased to 0.972.~~ Assimilating 50% of the ground-based observations improved the initial condition for other areas where have no assimilated sites. The DA can considerably improve the model $PM_{2.5}$ initial field, the CORR of verification sites in
605 mainland China improved from 0.58 to 0.84 and the RMSE decreased from 73.7 to 46.4 μg m^{-3}, respectively. The results of the DA00, DA12 assimilation experiments showed that the improved impacts of the DA worked throughout the forecast time window, but the assimilation impact was most pronounced in the first 12 hours and gradually decreased in the subsequent time. Within the 24-hour forecast time window, the average RMSE improvement for the China mainland $PM_{2.5}$ forecast field ranges from 9% to 21%, and between 4% and 25% in NC, and the comprehensive comparison shows that the initial field of 1200

610 UTC assimilation is superior to 0000 UTC. Therefore, in this study, it is considered that with limited computational resources, the E~~N~~nOI assimilation efficiency is highest with the largest distance between the model simulation and observation to assimilate according to the model characteristics. When it comes to operational use, the assimilation efficiency can be improved by shortening the assimilation time interval due to the small demand of EnOI computational resources.

The assimilation of $PM_{2.5}$ also has a positive impact~~significant improvement~~ on visibility forecasts~~, with different degrees of~~
615 ~~visibility improvement in different cities~~. When the $PM_{2.5}$ increment by assimilation is negative, it corresponds to an increase in visibility, and when the $PM_{2.5}$ analysis increment is positive, visibility decreases correspondingly. The greater the change in $PM_{2.5}$ concentration during periods of light pollution, the more pronounced the improvement in visibility~~, but this positive~~ ~~correlation is not particularly obvious during periods of heavy pollution. However,~~. iIt is worth noting that visibility is also related to a variety of factors and~~.~~ ~~Aa~~ssimilating only ground-based $PM_{2.5}$ sites has a limited effect on visibility, and we will
620 further consider assimilating PM_{10}, humidity and other meteorology factors to improve visibility forecasts in subsequent studies. ~~In addition, the number of ground-based PM~~$_{2.5}$~~ sites is not large enough in most region of China, and we will consider~~ ~~assimilating PM~~$_{2.5}$~~ and satellite AOD data simultaneously at a later stage to achieve more accurate PM~~$_{2.5}$~~ and visibility forecasts.~~

Code and data availability. The EnOI method and related processes written in Fortran language and observation data used in
625 this research are available at https://doi.org/10.5281/zenodo.7002847. The National Centers for Environmental Prediction Global Final Analysis (NCEP-FNL) data are available online (https://rda.ucar.edu/datasets/ds083.2/ and https://rda.ucar.edu/datasets/ds083.3/). The emission inventories are available online (http://www.meicmodel.org/).

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Table1. Mass concentration limit of PM$_{2.5}$ and its corresponding air quality level and air pollution index (API)

PM$_{2.5}$ concentration limit (ug m^{-3})	Air quality description	level	API
35	excellent	I	0-50
75	good	II	51-100
115	light pollution	III	101-150
150	moderate pollution	IV	151-200
250	heavy pollution	V	201-300
>250	hazardous pollution	VI	>300

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Table21. Experimental design.

Name	Experiment	Design
Control experiment	CR00	Warm restart (WS) at 0000 UTC and without DA
	CR12	WS at 1200 UTC and without DA
Sensitivity experiment	L20km N48 WS00	Fixed ensemble size N of 48, assimilation of the initial field at 0000 UTC per day, and localization length scale L of 20, 40, 60, 80, 100 km were selected for the assimilation experiment
	L40km N48 WS00	
	L60km N48 WS00	
	L80km N48 WS00	
	L100km N48 WS00	
	L80km N24 WS 00	Fixed localization length-scale L of 80 km, assimilation of the initial field at 0000 UTC per day, and ensemble size N of 24, 48, 72, 96, 120, 144 km were selected for the assimilation experiment
	L80km N48 WS 00	
	L80km N72 WS 00	
	L80km N96 WS 00	
	L80km N120 WS 00	
	L80km N144 WS 00	
	DA00	WS at 0000 UTC and with DA

	DA12	WS at 1200 UTC and with DA

Name	Experiment	Design
Control experiment	CR00	Model control run without DA begin at 0000 UTC every day and forecast 24 hours
	CR12	Model control run without DA begin at 1200 UTC every day and forecast 24 hours
Sensitivity experiment	Lxxkm-N48	assimilation at 0000 UTC with fixed ensemble size N of 48 and different localization length-scale Lxx of 20, 40, 60, 80, 100 km
	L80km-Nyy	assimilation at 0000 UTC with fixed localization length-scale L of 80 km and different ensemble size Nyy of 24, 48, 72, 96, 120, 144
Cycling assimilation experiment	DA00	Model forecast with assimilation at 0000 UTC every day
	DA12	Model forecast with assimilation at 1200 UTC every day
	DA00&12	Model forecast with assimilation at 0000 UTC and 1200UTC everyday
	DA00&12	Model forecast with assimilation at 0000 UTC and 1200UTC everyday

830 Table2. Statistics of the analysis fields PM$_{2.5}$ concentrations from 15 to 23 November 2016 at 0000 UTC for the sensitivity experiments with localization length-scale of 80km and ensemble size N of 24, 48, 72, 96, 120,144

	24	48	72	96	120	144
CORR	0.970	0.974	0.975	0.975	0.959	0.961
RMSE (ug m-3)	20.675	19.170	18.908	18.849	23.919	23.416
MB (ug m-3)	2.160	1.963	1.943	1.938	2.514	2.560
ME (ug m-3)	12.061	11.158	10.943	10.858	13.997	13.532

835 **Table3. Statistics of PM$_{2.5}$ concentrations for verifications sites of the initial field without (CR) and with assimilation at 0000 UTC each day from 1 to 31 December 2016. Assimilation sensitivity experiments were performed with 48 ensemble samples and length-scale L of 20, 40, 60, 80, 100km respectively and only assimilated the assimilation sites.**

	CORR	RMSE (µg m^{-3})	MB (µg m^{-3})	ME (µg m^{-3})
CR	0.56	60.1	8.5	41.7
20	0.77	48.2	4.1	30.2
40	0.82	41.6	3.2	25.9
60	0.81	43.1	3.5	26.8
80	0.80	44.9	3.8	27.0
100	0.79	46.0	4.0	28.2

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Table4. Statistics of PM$_{2.5}$ concentrations for verifications sites of the initial field withthout (CR) and with assimilation at 0000 UTC each day from 1 to 31 December 2016. Assimilation sensitivity experiments were performed with a localization length-scale L of 80 km and an ensemble size of 24, 48, 72, 96, 120, and 144, respectively and only assimilated the assimilation sites.

	CORR	RMSE (µg m^{-3})	MB (µg m^{-3})	ME (µg m^{-3})
CR	0.56	60.1	8.5	41.7
24	0.76	48.6	4.2	30.8
48	0.80	44.9	3.8	27.0
72	0.81	42.4	3.4	25.9
96	0.82	40.7	2.9	25.7
120	0.80	44.1	3.6	26.2
144	0.79	45.8	3.9	27.3

850 **Table5. Statistics of initial PM$_{2.5}$ concentrations for assimilation sites (DA) and verifications sites (Ve) before EnOI (BF) and after EnOI (AF) at 0000 UTC on 16, 19, 20 and 23 December 2016, respectively.**

Date	site	IC	CORR	RMSE (μg m^{-3})	ME (μg m^{-3})	MB (μg m^{-3})
16	DA	BF	0.50	50.8	9.1	38.2
		AF	0.98	11.0	0.9	7.1
	Ve	BF	0.48	56.5	9.7	42.8
		AF	0.73	39.1	2.5	25.2
19	Da	BF	0.65	81.5	-9.3	55.2
		AF	0.98	17.3	-1.2	10.4
	Ve	BF	0.66	79.2	-5.7	50.2
		AF	0.85	56.1	1.3	32.4
20	DA	BF	0.67	95.0	-24.2	64.5
		AF	0.99	19.1	-2.7	10.2
	Ve	BF	0.66	94.7	-20.5	60.5
		AF	0.87	60.2	-5.1	35.1
23	DA	BF	0.52	47.6	20.6	36.9
		AF	0.97	10.8	2.1	6.8
	Ve	BF	0.50	50.3	23.3	37.9
		AF	0.75	31.4	6.0	20.6

Date	Site	IC	CORR	RMSE (µg m^{-3})	MB (µg m^{-3})	ME (µg m^{-3})
16	DA	BF	0.50	50.8	9.1	38.2
		AF	0.98	11.0	0.9	7.1
	Ve	BF	0.48	56.5	9.7	42.8
		AF	0.73	39.1	2.5	25.2
19	DA	BF	0.65	81.5	-9.3	55.2
		AF	0.98	17.3	-1.2	10.4
	Ve	BF	0.66	79.2	-5.67	50.2
		AF	0.85	56.1	1.3	32.4
20	DA	BF	0.67	95.0	-24.2	64.5
		AF	0.99	19.1	-2.7	10.2
	Ve	BF	0.66	94.7	-20.5	60.5
		AF	0.87	60.2	-5.1	35.1
23	DA	BF	0.52	47.6	20.6	36.9
		AF	0.97	10.8	2.1	6.8
	Ve	BF	0.50	50.3	23.3	37.9
		AF	0.75	31.4	6.0	20.6

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Figure 1. Flow chart of the main ~~calculation~~ **procedures for EnOI initial field assimilation** system**. The Obs. PM$_{2.5}$ is ground-based observation of PM$_{2.5}$. The C**CM~~T~~**M is** coupled chemistry meteorology models~~chemistry transport model~~**.** SVD is singular value decomposition

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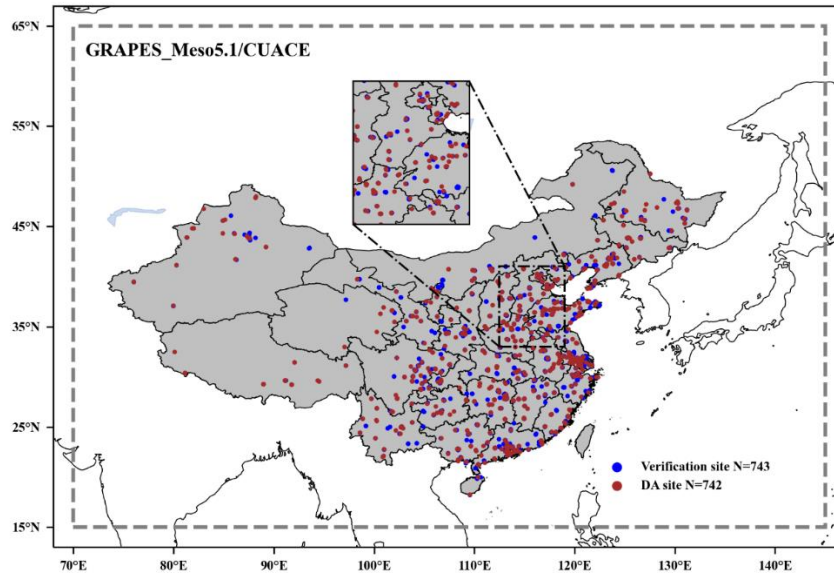
865 **Figure 2. Simulation domain of GRAPES_Meso5.1/CUACE. Minor region represents North China (NC). The locations of ground stations in China mainland are marked on the maps with blue and brown dots.** ~~Only when the independence test is performed, the brown assimilation sites and the blue verification sites are distinguished, otherwise, all sites are used as assimilation sites.~~ **The blue and brown dots represent verification sites and assimilation sites, respectively. "N=743" means there are 743 verification sites. "N=742" means there are 742 DA sites.**

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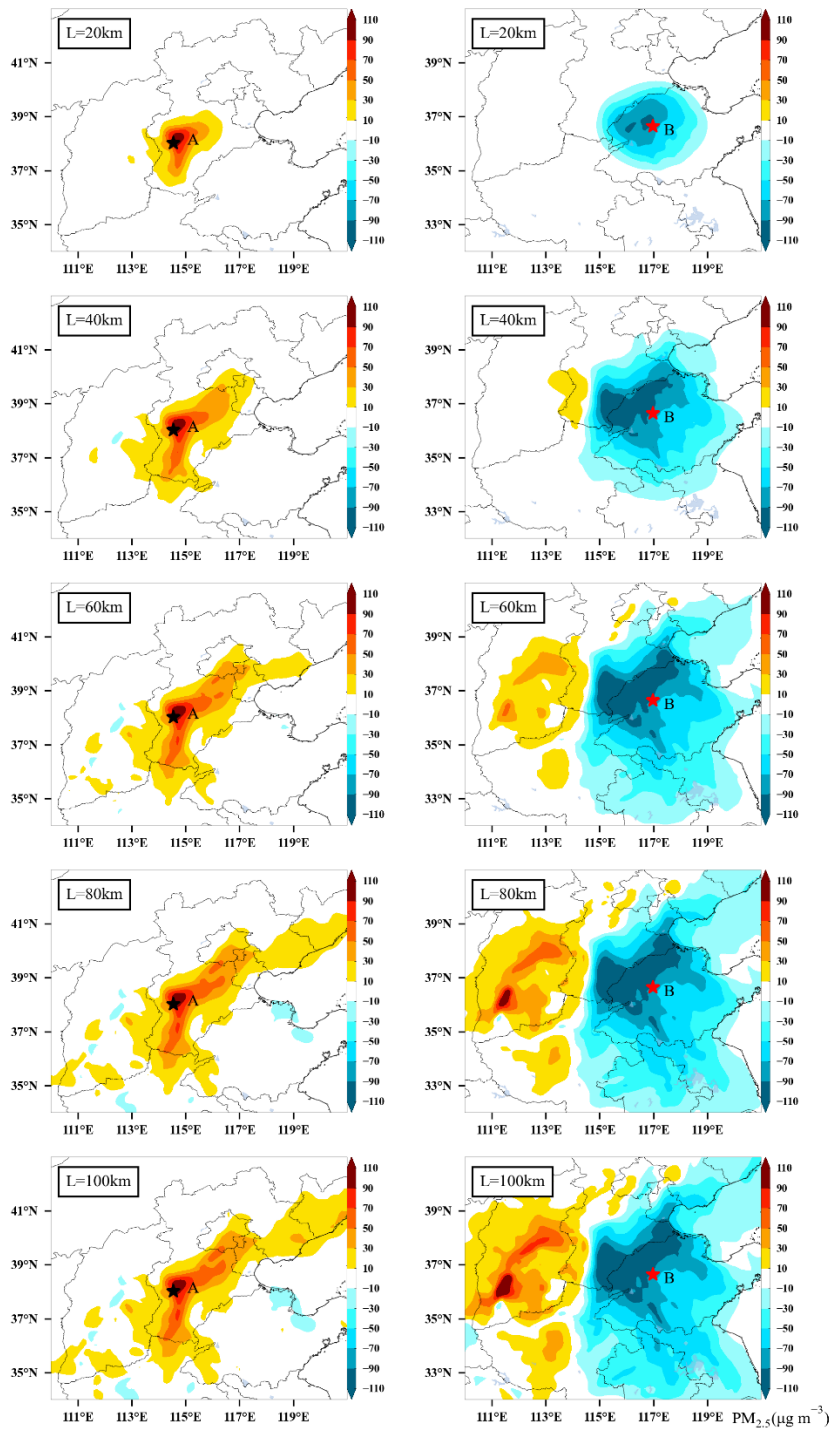


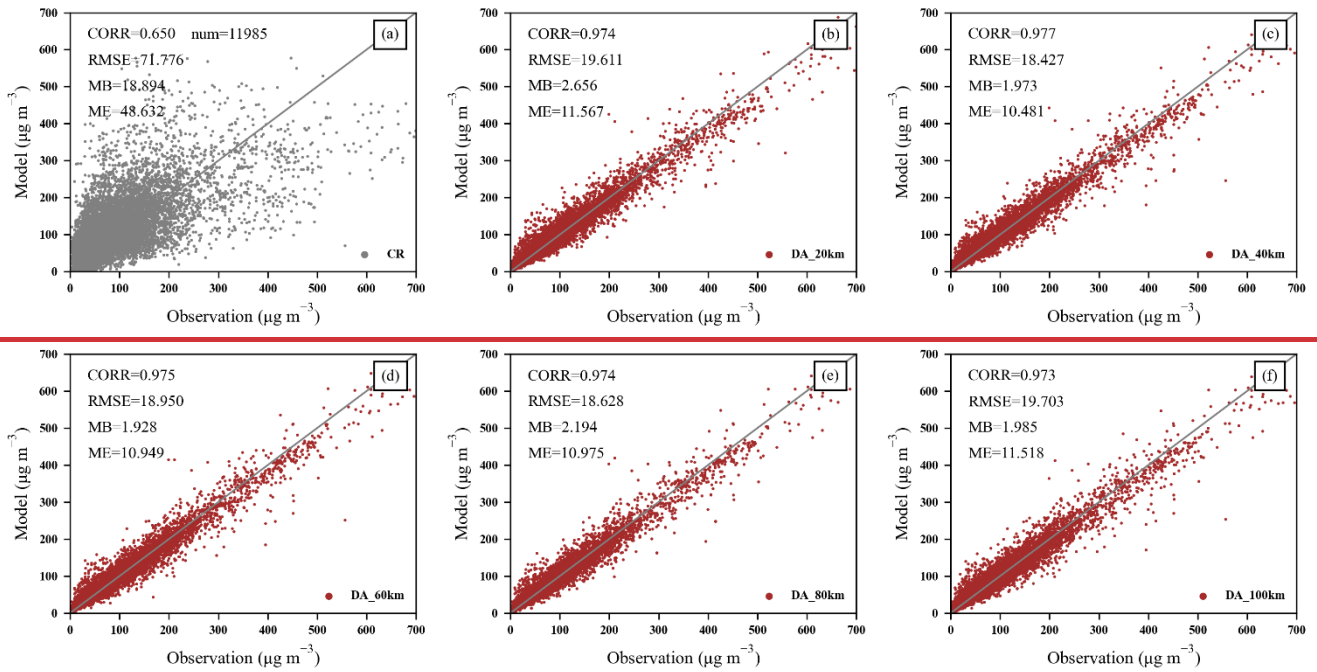
Figure 3. Spatial distribution of PM$_{2.5}$ analysis increments after assimilation of initial fields at 0000 UTC on 15 December 2016, for assimilation site A (38.0° N, 114.5° E) only (left column) and assimilation site B (36.6° N, 116.9° E) only (right column) with fixed ensemble size 48 and different localization length-scale of 20, 40, 60, 80, 100km.

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Figure 4. Scatter plot of PM$_{2.5}$ concentrations from the control experiment (CR) and the assimilation experiment with 40 ensemble samples and length-scale L of 20, 40, 60, 80, 100km (DA) with all observations analyses by 0000UTC during the experiment period. The num is the sum of all ground-based observations of PM$_{2.5}$ stations during the experiment period.

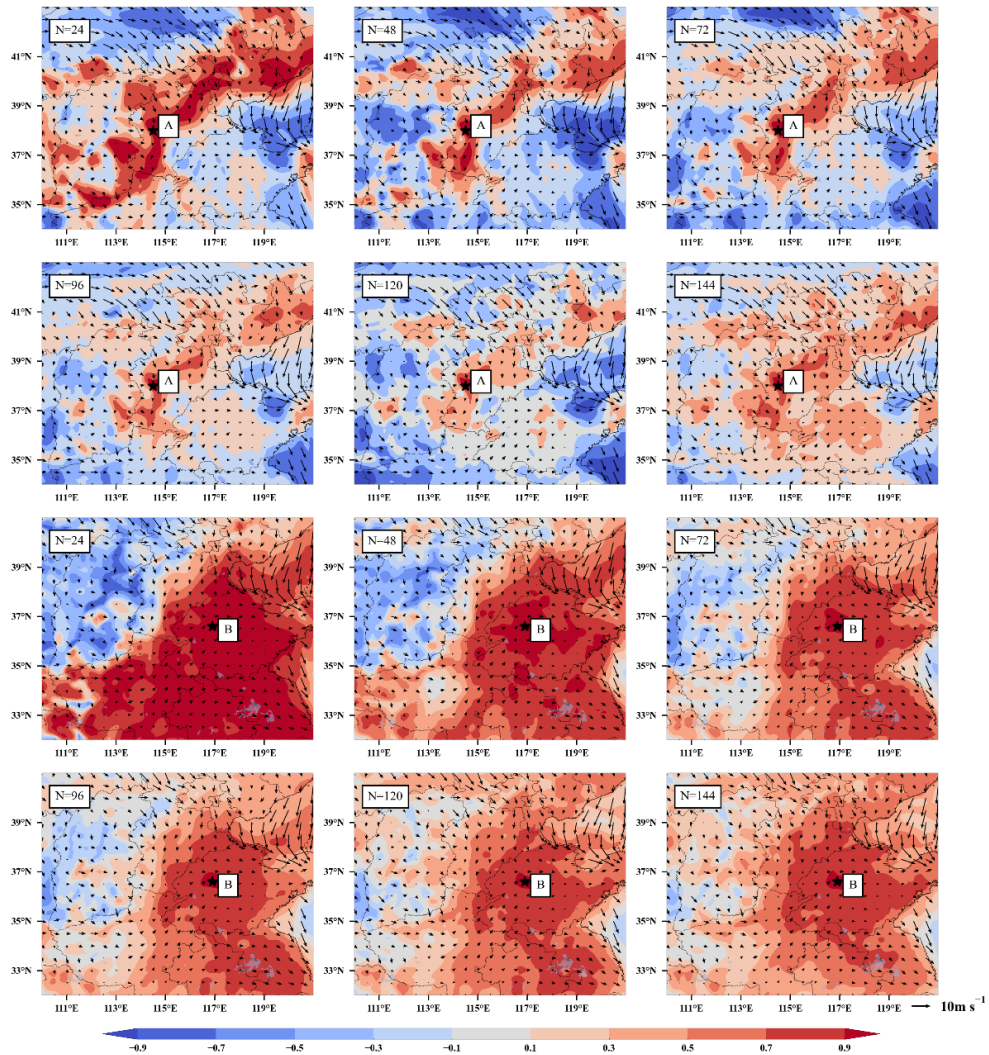



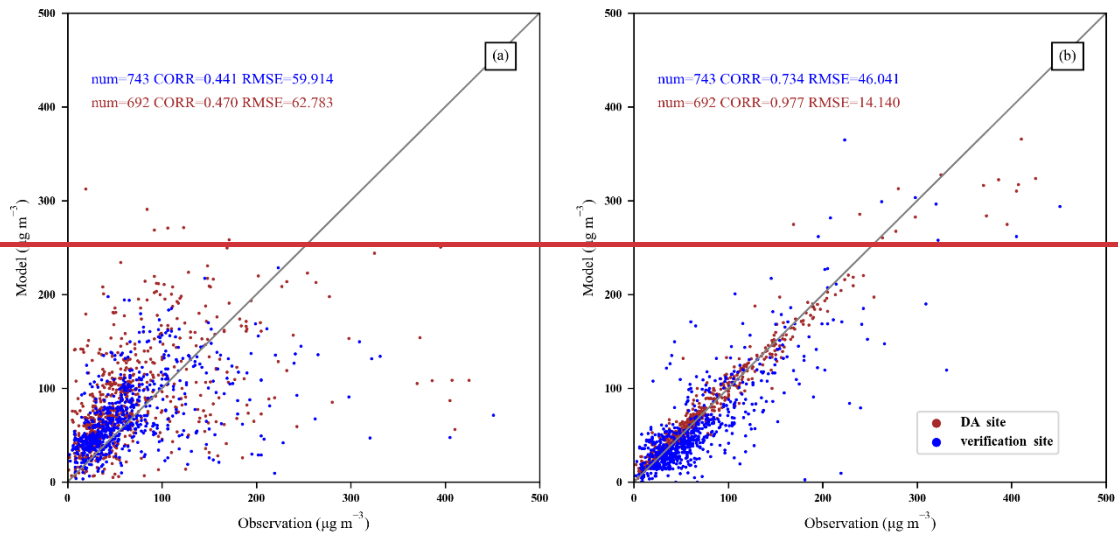
Figure ~~5~~4. Spatial distribution of correlation coefficients of background error for site A (38.0° N, 114.5° E) (rows 1, 2) and site B (36.6° N, 116.9° E) (rows 3, 4) with ~~ensemble error for assimilation experiments using length-scale of 80km and~~ different ensemble size N of 24, 48, 72, 96, 120, 144 and wind vectors at 0000 UTC on 15 December 2016.

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Figure 6. Scatter plot of PM$_{2.5}$ verification sites compared to assimilation (DA) sites for the control experiment (a) and assimilation experiment (b) using length-scale L=40km and ensemble size N=96 at 0000UTC on 15 December 2016.

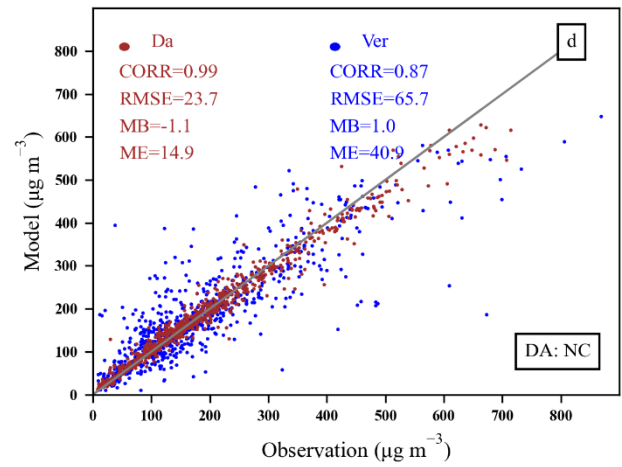
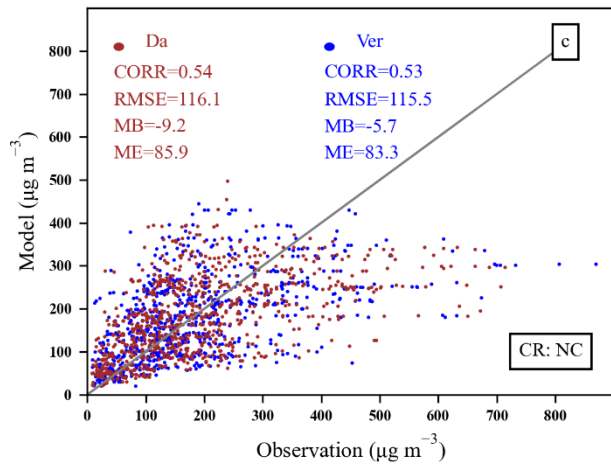
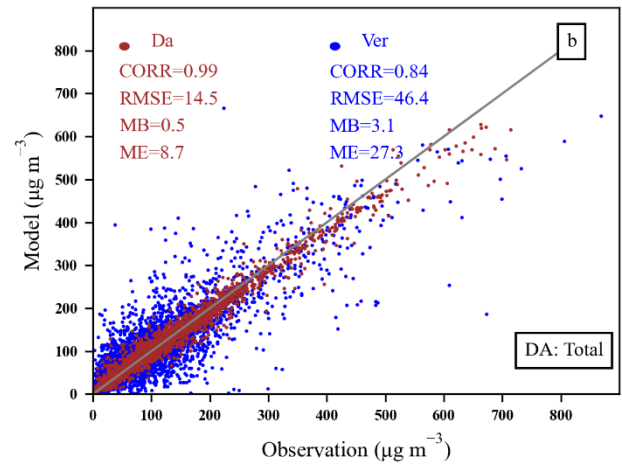
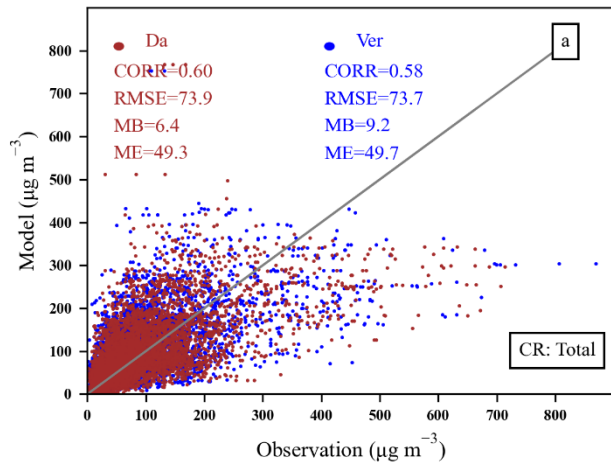



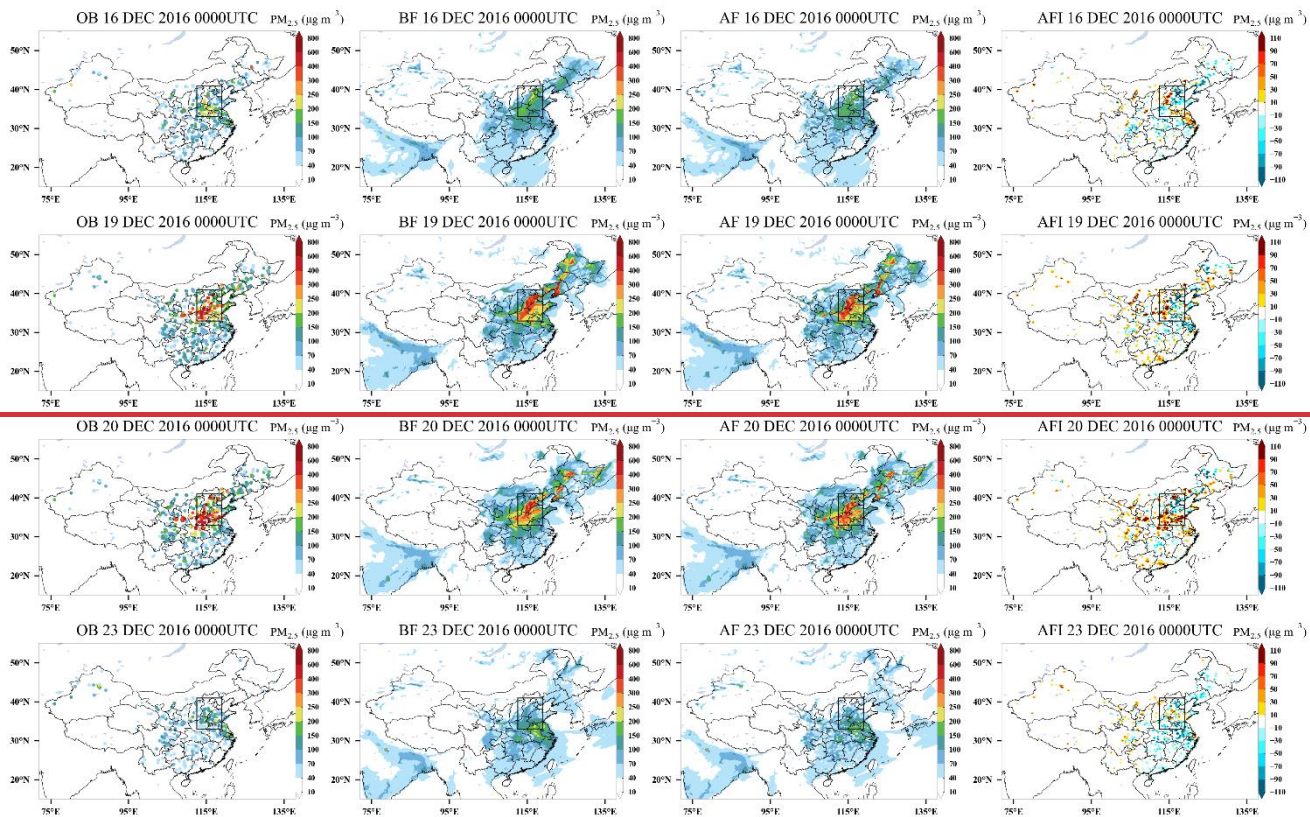
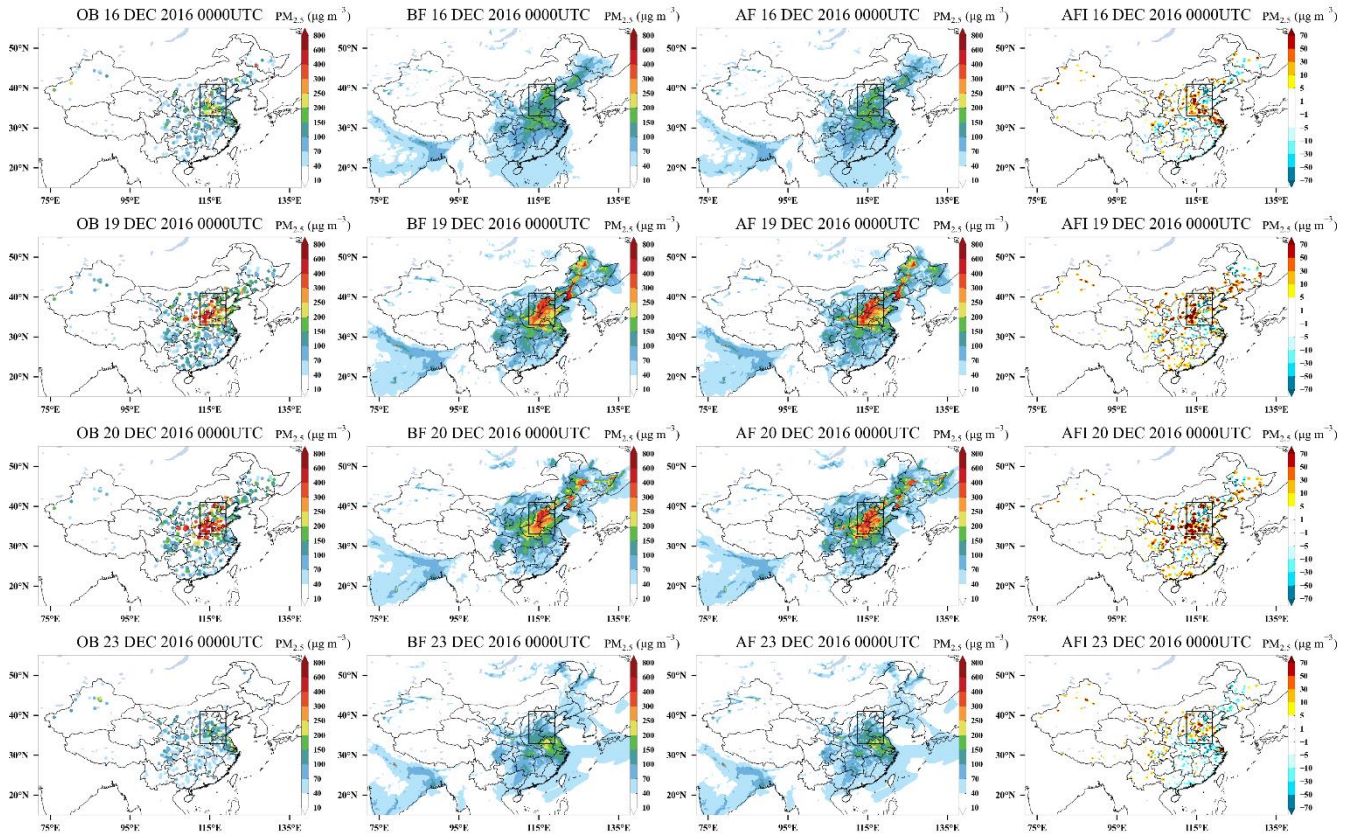
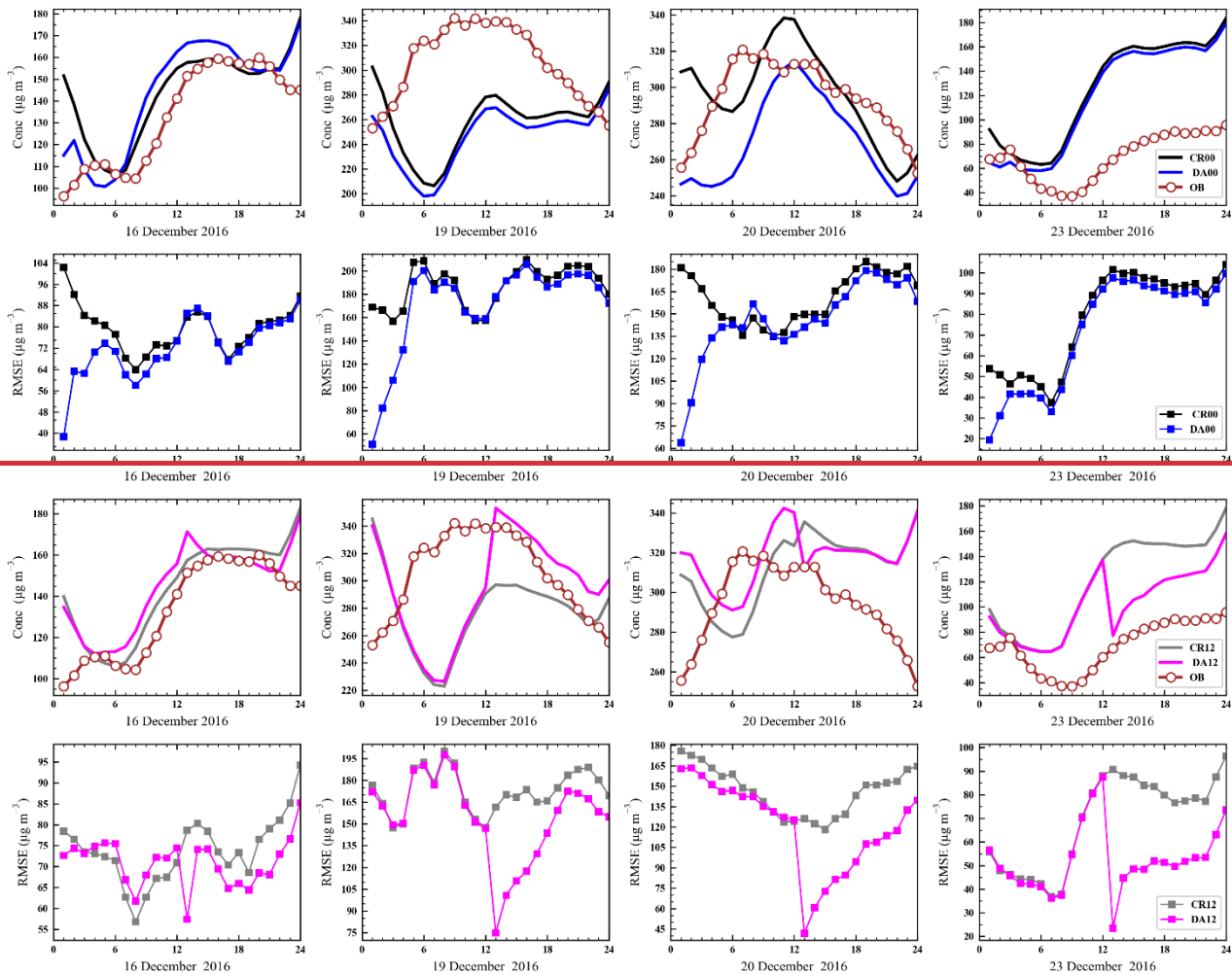
Figure 5. Scatter plot of PM₂.₅ concentrations from the control experiment (a,c) and the assimilation experiment (b, d). The ensemble size in the assimilation experiment is 96 and the length-scale L is 40 km. Brown (Da) and blue (Ver) dots are assimilation sites and validation sites respectively. a, b is for mainland China (Total), c, d is for North China (NC).

895 **Figure 7~~6~~. Snapshots of the horizontal distributions of PM~~2.5~~ observation (OB), before (BF) and after (AF) the application of EnOI technique, analysis field increment (AFI) at 0000 UTC on 16, 19, 20, and 23, December 2016. The black box area, representing ~~northern~~North China (NC)~~, has the most serious PM2.5 pollution~~.**

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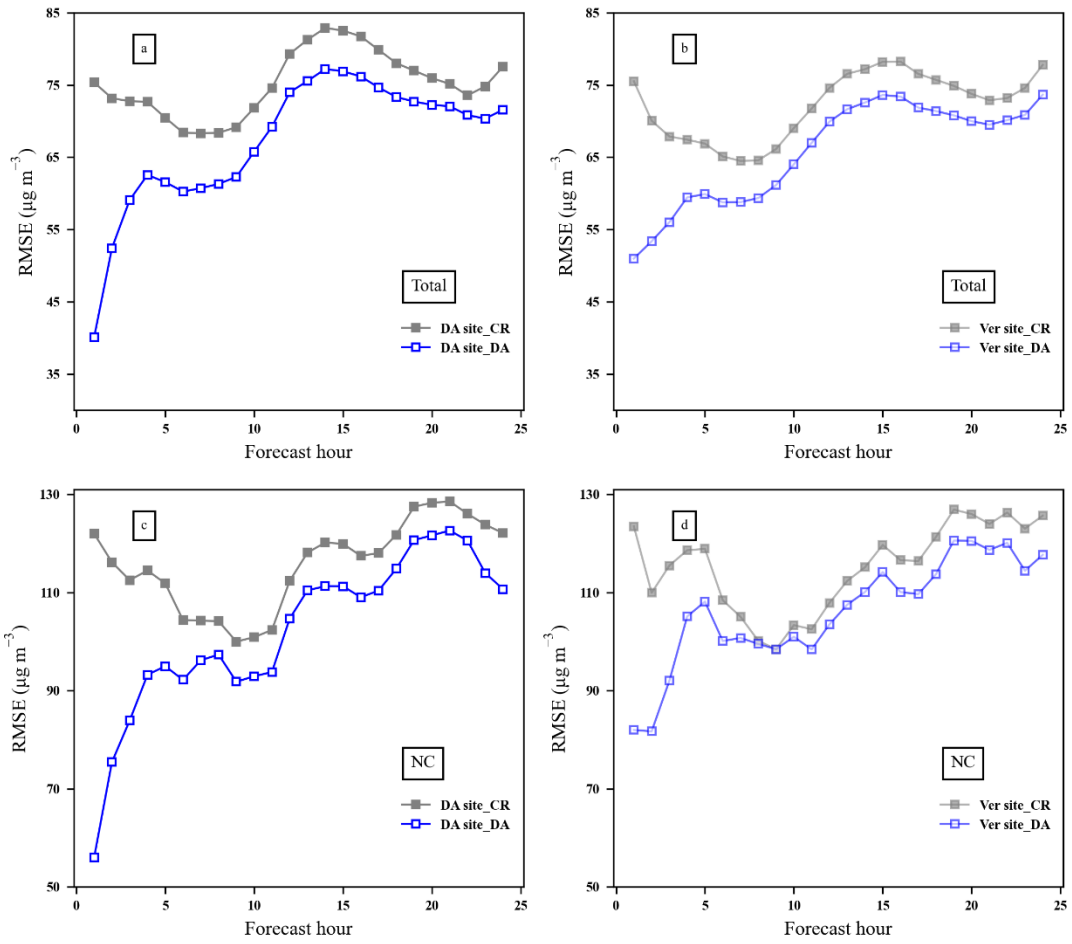
Figure 9. Relative RMSE time series for 15 to 23 December 2016 for Mainland China (a), North China (b). The relative RMSE is calculated by the daily average RMSE.

910 **Figure 7. The average RMSE value of surface PM$_{2.5}$ forecasts as a function of forecast time over (a) China Mainland for DA sites, (b) China Mainland for verification sites, (c) North China for DA sites, (d) North China for verification site.**

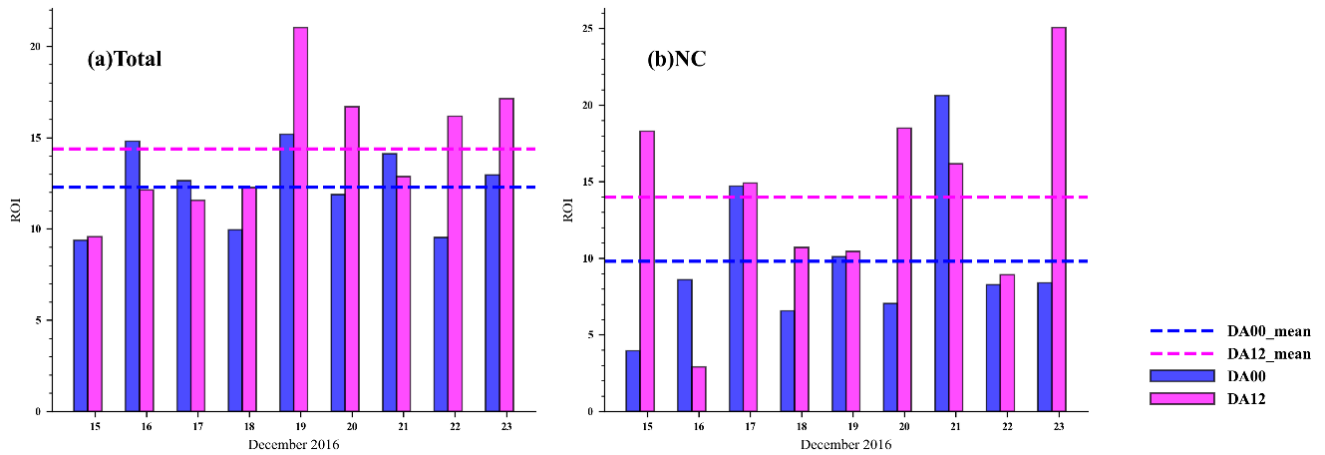



Figure 8. Rate of improvement (ROI, unit: %) by data assimilation in 1d (24h) predictions for 15 to 23 December 2016 over Mainland China (a), North China (b). The ROI is the ratio of the reduced RMSE statistical metrics to those for the CR simulation. DA00 and DA12 represent the initial field assimilation using EnOI at 0000 UTC and 1200 UTC each day, respectively. DA00_mean and DA12_mean represent the mean ROI over 15 to 23 December 2016 of DA00 and DA12, respectively.

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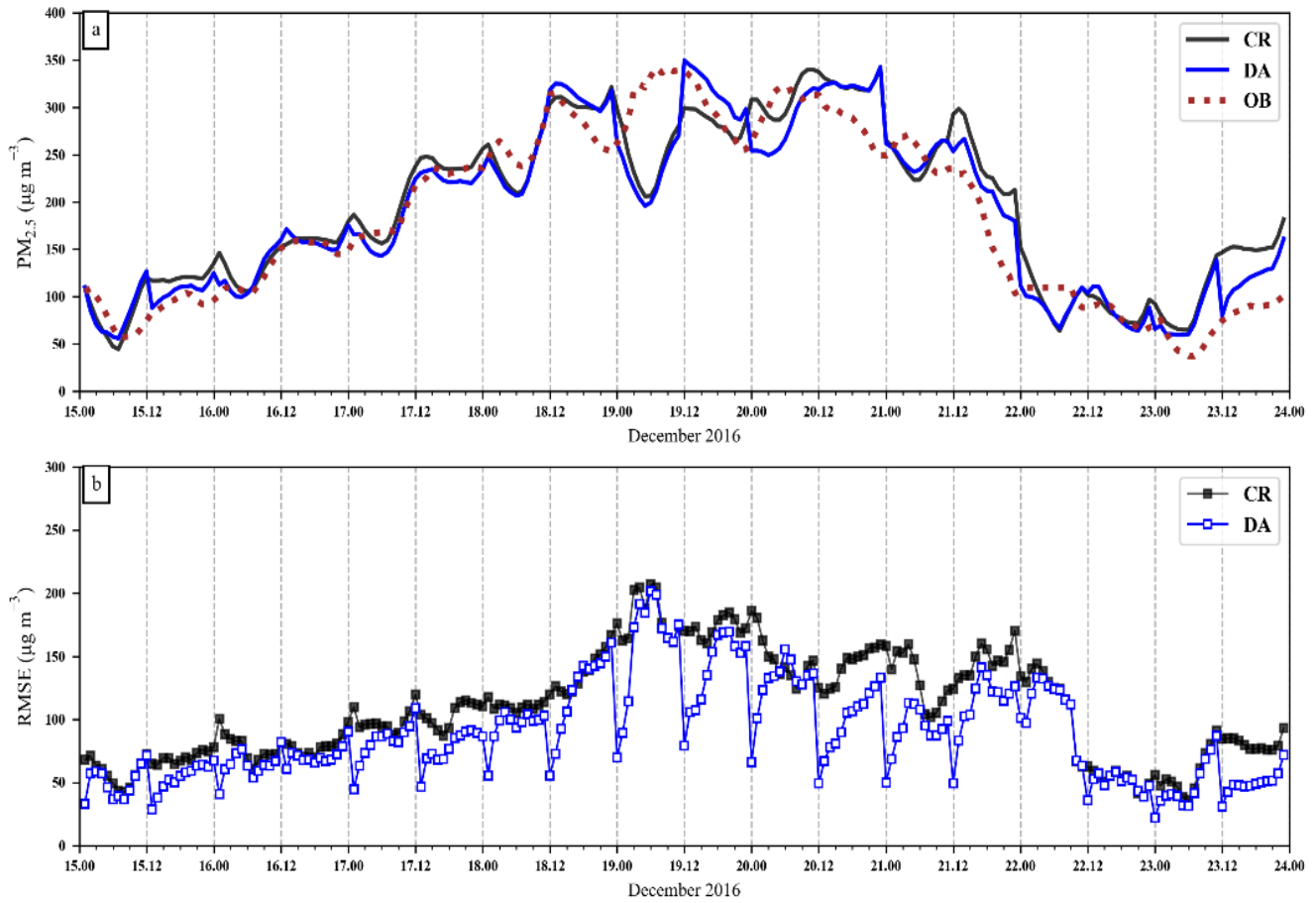
Figure 9. Time series of hourly PM₂.₅ concentration (a) and RMSE between forecasts and observations (b) from 15 December to 23 December 2016 in North China. Red dots: observations, black line: forecasts from control experiment (CR), blue line: forecasts from experiment with initial field assimilation at 0000UTC and 1200UTC (DA), black line with dots: RMSE between CR forecasts and observations, blue line with dots: RMSE between CR forecasts and observations. The values are averages calculated against all the observation sites in North China.
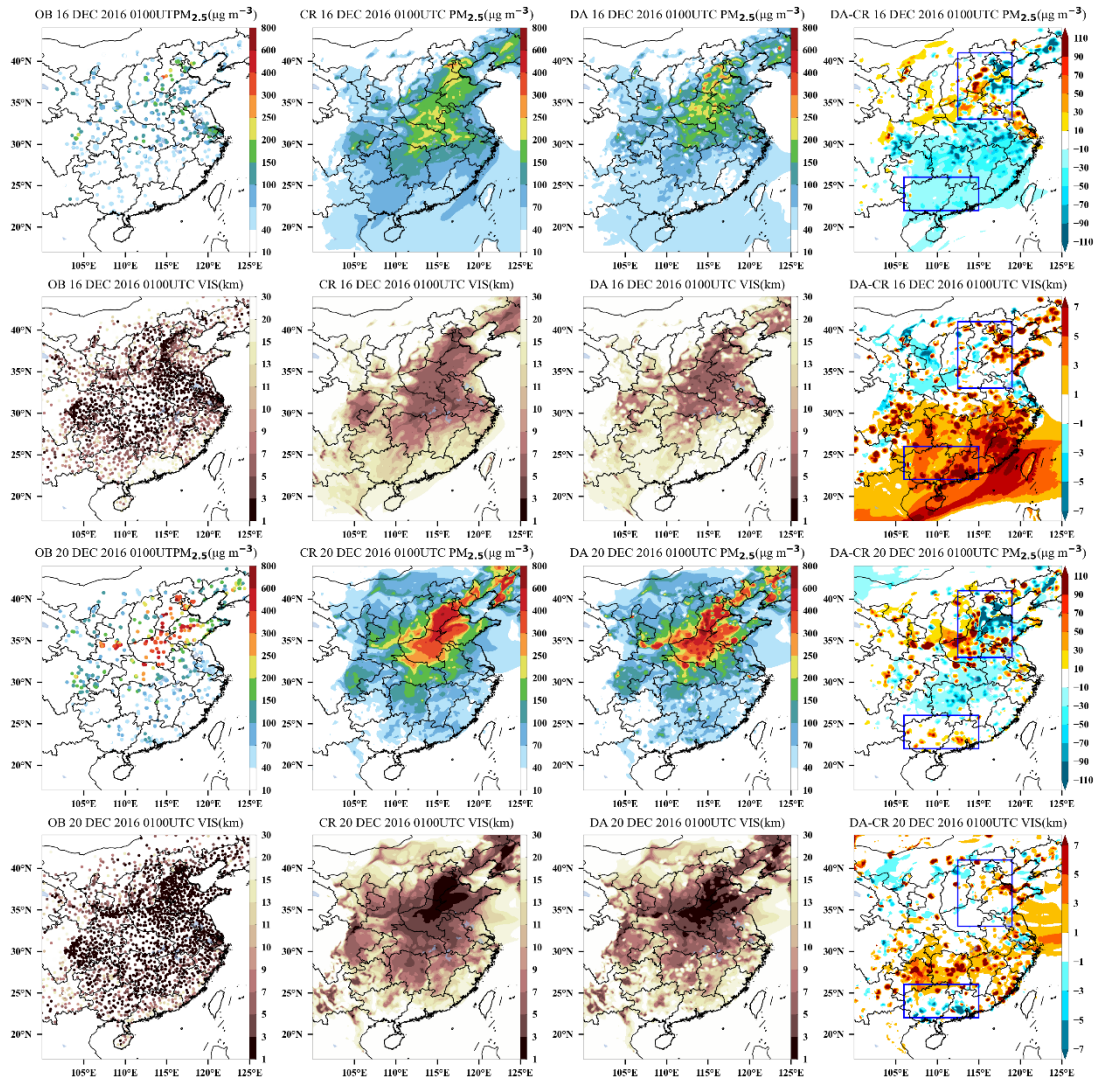
Figure 10. Snapshots of PM₂.₅ and visibility horizontal distribution for control (CR), assimilation (DA), observation (OB), and increment (DA-CR) at 0100 UTC after assimilation of the initial field at 0000 UTC on 16 and 20 December 2016.The upper box represents ~~northern~~ North China and the lower box represents Guangxi and Hainan in China.
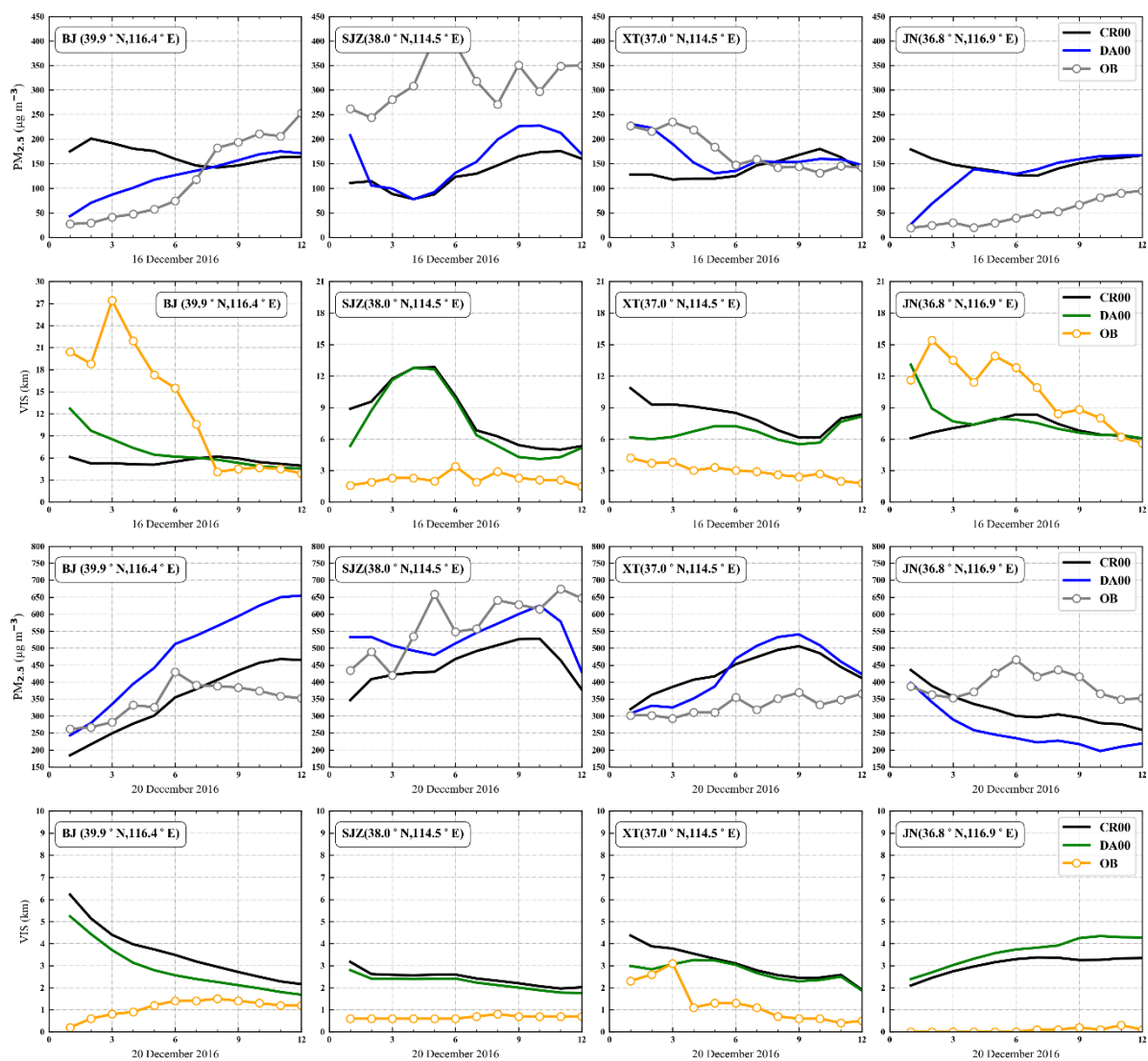
Figure 11. Comparison ~~between~~of PM$_{2.5}$ and visibility observations and model forecast at four cities without (CR00) and with assimilation, ~~assimilation experiment simulations (DA00), and control experiment simulations (CR00) after assimilation~~ (DA00) of the initial field at 0000 UTC on 16 and 20 December 2016~~each day~~. Four cities are exemplified, from left to right, Beijing (BJ), Shijiazhuang (SJZ), Xingtai (XT), Jinan (JN). The labels on the x-axis refer to the first 12 forecast hours of ~~16 and 20 December 2016~~the day, PM$_{2.5}$ observations: grey line with circles, visibility observations: orange line with circles, PM$_{2.5}$ and visibility model forecast without assimilation: black line, PM$_{2.5}$ model forecast with assimilation: blue line, visibility model forecast with assimilation: green line.