

Authors' responses to Referees' comments

Journal: Geoscientific Model Development

Manuscript Number: egusphere-2025-3960

Title: OIRF-LEnKF v1.0: A Self-evolving Data Assimilation System by Integrating Incremental Machine Learning with a Localized EnKF for Enhanced PM_{2.5} Chemical Component Forecasting and Analysis

Authors: Hongyi Li, Ting Yang, et al.

Note:

Comment (12-point black italicized font).

Reply (indented, 12-point blue normal font).

“Revised text as it appears in the text (in quotes, 12-point blue italicized font)”.

Anonymous Referee #2

1 General Comments:

This manuscript presents “OIRF-LEnKF v1.0,” a hybrid data assimilation (DA) system that couples an optimized incremental Random Forest (OIRF) model with a Localized Ensemble Kalman Filter (LEnKF). The primary goal is to address the computational inefficiency and limited forecasting improvement associated with traditional Chemical Transport Model (CTM)-based DA systems. By replacing the CTM ensemble with a machine learning (ML) ensemble that updates itself via incremental learning, the authors claim to achieve significant efficiency gains and improved accuracy in estimating PM_{2.5} chemical components. The topic is highly relevant to the scope of Geoscientific Model Development, as it addresses the critical bottleneck of computational cost in atmospheric chemistry DA. The integration of incremental learning (updating decision trees based on analysis increments) is a novel and interesting approach to handling non-stationary error distributions. The validation against independent sites and other reanalysis datasets suggests the system performs well.

However, there are several major concerns regarding the terminology used (specifically “forecasting”), the dependence on reanalysis inputs, and the circularity of the self-evolving mechanism that must be addressed before publication. The critical distinction

between a “reanalysis generator” and a “forecast system” seems blurred in the current experimental design. The experimental period is very insufficient to support the claims.

Authors’ response:

We sincerely thank the reviewer for the thorough and insightful review, as well as for the positive assessment of the novelty and relevance of our work. We appreciate the constructive criticisms, which have helped us identify crucial areas for clarification and improvement. We will address the major concerns regarding the terminology used (specifically “forecasting”), the dependence on reanalysis inputs, the circularity of the self-evolving mechanism, and the insufficient experimental period point-by-point below.

2 Major Comments:

1) Clarification of “Forecasting” vs. “Hindcasting/Reanalysis”

The title and abstract repeatedly emphasize the system’s “forecasting” capability. However, Section 2.2.1 states that the input features for the OIRF model include meteorological parameters from ERA5 and atmospheric pollutants from CAQRA (a reanalysis dataset). In that setting, the model is effectively learning an instantaneous relationship (features at time $t \rightarrow$ components at time t). In a true operational forecast setting, ERA5 and CAQRA data are not available in real-time; they are retrospective datasets. If the OIRF model relies on con-current reanalysis data as inputs to predict chemical components, this is technically a “diagnostic” application, not a “prognostic forecast.” The authors must clarify this distinction. If the system is intended for operational forecasting, they should discuss how it would perform using forecast meteorology (e.g., IFS or GFS) and forecast pollutants (e.g., CTM forecasts) as inputs, rather than high-quality reanalysis data. This is not a minor wording issue: if inputs are reanalysis fields at the verification time, then improvements in RMSE/CORR do not necessarily translate to operational forecast skill. If the primary purpose is generating reanalysis datasets (as implied by the comparison in Section 3.4), the manuscript should be reframed to reflect that this is a “reanalysis system” or “hindcast system,”

as calling it a “forecast” is misleading given the input data latency.

Authors’ response:

We sincerely thank the reviewer for this crucial and insightful comment, which correctly identifies the most important interpretive limitation of our current model input design. We fully agree that the distinction between a diagnostic application and a real-time prognostic forecast is fundamental.

a. Terminology correction

The reviewer is correct. The OIRF model learns an instantaneous mapping relationship based on retrospective reanalysis datasets and technically performs a “reanalysis-based simulation” rather than a “prognostic forecast”. Therefore, referring to simulation or instantaneous mapping as an operational “forecast” is misleading. We apologize for this imprecision. **In the revised manuscript, we have thoroughly replaced the inappropriate terms such as “forecast”, “forecast field”, and “FOR” with more accurate terms such as “simulation”, “background field”, and “SIM” in the title, abstract, main text, figures (including Fig. 1, Fig. 3, Fig. 4, Fig. 5, Fig. 6, Fig. 7) and tables.** For brevity, only the revised title and abstract are shown below, and all modified figures are displayed at the end of this reply.

Title, Line 1-4: “*OIRF-LEnKF v1.0: A Novel Data Assimilation System by Integrating Incremental Machine Learning with a Localized EnKF for Enhanced PM_{2.5} Chemical Component Simulation and Reanalysis*”

Abstract, Line 12-29: “*Assimilating observational data into numerical simulation is crucial for accurately estimating the spatiotemporal distribution of PM_{2.5} chemical components (NH₄⁺, NO₃⁻, SO₄²⁻, OC, and BC), which is beneficial to quantifying the impact of aerosols on the environment, climate change and human health. However, chemical transport model (CTM)-based data assimilation (DA) is computationally inefficient for large ensemble sizes and offers limited improvements in simulation skill, as it solely provides optimal initial conditions. This paper introduces an incrementally updatable machine learning-based data assimilation system (Optimized Incremental Random Forest coupled with Localized Ensemble Kalman Filter, OIRF-LEnKF v1.0)*

that achieves high efficiency and high quality in generating background and analysis fields for chemical components. Computational efficiency tests indicate that the total time consumed by OIRF-LEnKF v1.0 constitutes only 11.41-16.60 % of that of CTM-based DA, particularly during the simulation process (0.13-0.20 %). Sensitivity tests demonstrate that the self-evolution mechanism in our system enhances the Pearson correlation coefficient (CORR) and reduces the RMSE during the simulation process by 2.28-11.75 % and 32.94-40.98 %, respectively, compared to the stationary training mechanism. A 2-month DA experiment reveals that the RMSE values of chemical components after DA are less than $7.80 \mu\text{g m}^{-3}$ and $2.36 \mu\text{g m}^{-3}$ during the simulation and analysis processes, respectively, indicating reductions of at least 26.38 % and 68.99 % compared to values without DA. Notably, the RMSE values of our system during the simulation process exhibit a significant reduction of 33.16-90.10 % compared to those of the CTM-based DA, highlighting the superior simulation capability of our system. Furthermore, the spatial overestimation and underestimation of chemical components have been significantly mitigated following DA. Compared to multiple reanalysis datasets of inorganic salt aerosols (CORR: 0.56-0.89, RMSE: 2.55-8.52 $\mu\text{g m}^{-3}$), the dataset generated by OIRF-LEnKF v1.0 (CORR: 0.97, RMSE: 1.12 $\mu\text{g m}^{-3}$) demonstrates higher data quality.”

b. Clarification of our study's primary goal

The primary objective of this work is to propose and validate a novel framework that online couples an incrementally updatable AI-based surrogate model and an ensemble data assimilation algorithm, which enables the AI-based surrogate model and the data assimilation component to benefit from the dynamic information provided by the other at each iteration. During the concept proof stage, using optimal reanalysis inputs is deliberate to establish a valid representation of forecast/simulation uncertainty. Specifically, the OIRF-LEnKF utilizes the decision tree members in the OIRF model to estimate the background error covariance without input perturbations, which implicitly assumes that the forecast/simulation uncertainty mainly originates from the OIRF model's inherent incompleteness in learning the mapping relationship between input and target features. Therefore, using reanalysis data as input excludes the additional

uncertainty that would arise from imperfect forecast inputs.

c. Discussion of operational forecasting

The application of the OIRF-LEnKF system in operational forecasting is feasible, but its comprehensive validation would require a broader set of experiments, such as sensitivity tests on forecast ensemble generation. The in-depth investigation on the operational forecasting extends beyond the primary scope of this paper. We sincerely thank the reviewer's constructive suggestions, which provide significant inspiration. Our immediate future work will indeed prioritize these forecasting experiments, such as employing forecast data as input and assessing performance under different ensemble generation strategies (e.g. using perturbed meteorological forecast data alone, jointly perturbing meteorological and pollutant forecasts, or developing hybrid methods that integrate input perturbations with the intrinsic ensemble statistics from the decision tree members).

2) Circularity / information leakage risk in the incremental learning loop

A key design choice is that each decision tree is scored using MAE against the analysis field at the same time step, and trees are replaced by new trees trained on “analysis” targets. I have two concerns on this design. The first one is that the system is self-training on its own analysis. The system progressively trains on DA outputs (which incorporate the observations), not solely on an external reference dataset. This can lead to overly optimistic performance if not carefully controlled. The second one is that the scoring target is non-independent. The analysis field is itself a function of the forecast ensemble (through the forecast covariance used by EnKF). Even with localization, using the analysis as “ground truth” for selecting trees can create a feedback loop where the ensemble is optimized to match its own internally constructed target. At minimum, the paper should include a leakage-aware evaluation, for example, scoring the incremental learning using withheld stations (VE sites) not assimilated, or withheld time blocks, rather than analysis fields produced by assimilating the same network. Providing an ablation where incremental learning is driven by an external target versus DA-derived analysis, to quantify how much of the gain comes from DA self-training can

also be very valuable.

Authors' response:

We thank the Reviewer for this exceptionally insightful comment.

Regarding the first concern, we fully agree that the system progressively trains the OIRF model using DA outputs can lead to overly optimistic performance if not carefully controlled. We would like to clarify several key points as follows.

a) DA outputs serve as a high-quality target set for training, aiming to accurately establish the mapping relationship between the input features and five PM_{2.5} chemical components. Importantly, the re-training utilizes the DA outputs from the previous time step. The updated RF model is then applied to provide background fields for the current and all subsequent time steps until the next update. This design ensures that the model does not gain prior knowledge of future states, thereby preventing artificially optimistic performance.

b) An external and independent target set of high-quality observations typically provides insufficient sample size (only 9 VE sites in our case) for training at a single time step. Meanwhile, as validated in *Section 3.4*, external reanalysis datasets exhibit lower accuracy than DA outputs, making them a suboptimal choice for the “ground truth” target in the re-training process.

c) **Most crucially, our system incorporates specific controls (update frequency and update intensity) to mitigate the risk of overly optimistic performance.** The update frequency parameter, which determines how often the OIRF model integrates DA outputs, is optimized by sensitivity experiments, as detailed in *Section 3.2*. The update intensity parameter is implemented by a controllable threshold (τ_p), which governs the proportion of decision trees (DTs) replaced. A higher threshold ensures that only a small and stable fraction of the DTs is replaced during each update cycle, which prevents model overfitting to the new DA outputs.

We acknowledge that some rationales and controls were not explicitly detailed in the original manuscript, which may have caused confusion. We have revised the text accordingly to provide the necessary clarification as follows.

Section 2.1.1, Line 128-143: “As shown in Fig. 1, the fundamental workflow of OIRF-LEnKF v1.0 is as follows.

Step 1. Initial training of the OIRF model. The training data at the first timestep serve

as the initial conditions for constructing the OIRF model. The input features include meteorological parameters, including temperature, relative humidity, U-component wind, V-component wind, and geopotential, as well as anthropogenic atmospheric pollutants, including $PM_{2.5}$, PM_{10} , SO_2 , NO_2 , CO , and O_3 . The output features are SO_4^{2-} , NO_3^- , NH_4^+ , OC , and BC .

Step 2. Incremental learning of the OIRF model at time steps > 1 . High-quality analysis fields at the last time step, along with the corresponding meteorological and anthropogenic input data, are employed to train a new ensemble of decision trees. The old decision trees, which exhibit poor simulation performance, are subsequently replaced with new decision trees to enhance the simulation accuracy and generalization ability of the OIRF model.

Step 3. Generating a background ensemble of $PM_{2.5}$ chemical component concentrations at the current timestep using the OIRF model, along with the current meteorological and anthropogenic input data.

Step 4. Generating the analysis fields of $PM_{2.5}$ chemical component concentrations at the current timestep by assimilating chemical observations into background fields using the LEnKF algorithm.

Step 5. Scoring the simulation performance of ensemble decision trees in the OIRF model using mean absolute error (MAE) and screening out the decision trees with poor simulation performance based on a predefined threshold. Repeat steps 2-5 until the end of the loop.”

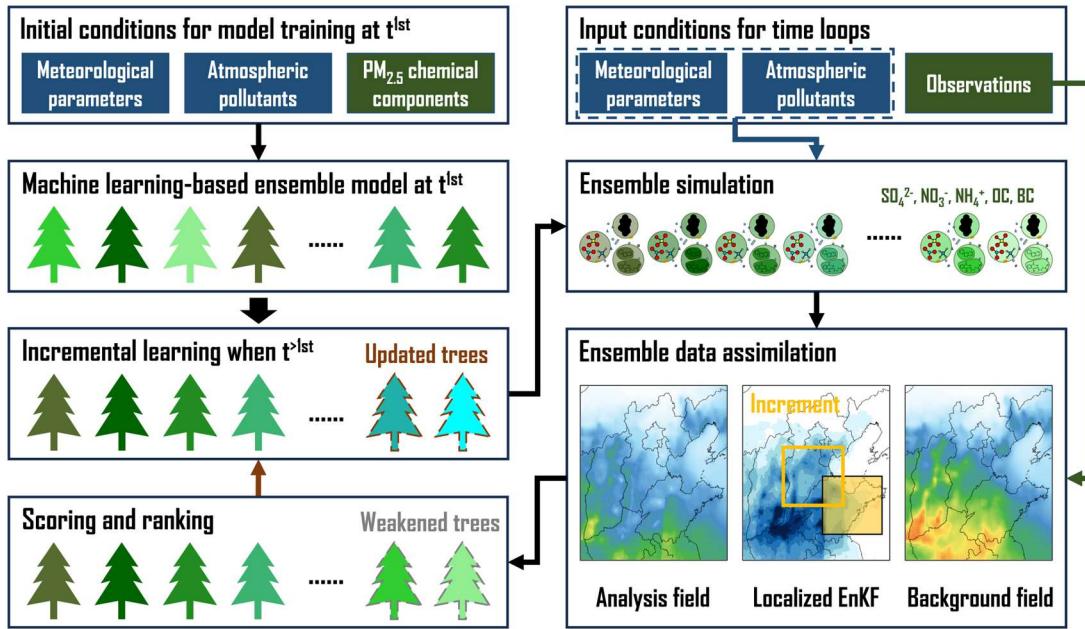


Figure 1. The framework of OIRF-LEnKF v1.0.

Section 2.1.2, Line 175-180: “The incremental learning mechanism introduces a threshold (τ_p) to screen out the DTs with poor simulation performance. The threshold is defined as the p^{th} percentile value of f_n^{score} . The percentile-based threshold ensures a stable and controllable number of DTs are updated, a critical feature for maintaining the smoothness and stability of the estimation of background error covariance within the ensemble data assimilation framework and preventing model overfitting to the new information. As shown in Eq. (3), the old DTs with scores not higher than τ_p are retained, while the old DTs with scores higher than τ_p will be replaced by new DTs obtained from the incremental learning process.”

Section 2.1.2, Line 185-186: “...The p is set at 80 to prevent excessive updating of DTs, which may introduce instability and artificially optimistic performance into ensemble simulation of the OIRF model.”

Regarding the second concern, we fully agree that our work should include a leakage-aware evaluation. Following the Reviewer’s suggestion, we have conducted an experiment in which we score the incremental learning using the observations from VE sites that have not been assimilated.

Section 2.1.2, Line 162-168: “Inspired by the idea of dynamically updating DTs with weak performance (Xie et al., 2016), the OIRF model incorporates a novel incremental learning mechanism into the RF model, enabling it to conduct effective updating from

newly available training data within a simulation-assimilation cycle. In the incremental learning mechanism, the OIRF model scores the simulation performance of each DT based on the mean absolute error (MAE), as shown in Eq. (2). The MAE is quantified by the DT outputs and high-accuracy analysis fields at the same time step. A leakage-aware evaluation indicates that using the analysis field as scoring target did not cause substantial information leakage, while employing the independent high-quality observation as scoring target is also recommended (Sect. S1 in the Supplement)."

Supplement: "Sect. S1: Leakage-aware evaluation of incremental learning

In the incremental learning mechanism, each decision tree (DT) member is scored by comparing its simulation to the analysis field using mean absolute error (MAE). However, using the analysis field as the scoring target for selecting trees could arise a feedback loop risk as the DT ensemble may become optimized toward its own internally constructed target. Therefore, we conducted a leakage-aware evaluation for February 2022 by comparing simulation performance of the OIRF model when the scoring target is set as the analysis field against when it is set as the independent observation at withheld sites (VE sites) not assimilated. Fig. S1 shows that both scoring targets achieved comparable performance across all five $PM_{2.5}$ chemical components, with correlation coefficient (CORR) values of 0.39-0.85 (analysis-field target) versus 0.39-0.86 (independent-observation target), and RMSE values of 1.02-5.85 $\mu g m^{-3}$ (analysis-field target) versus 0.95-5.68 $\mu g m^{-3}$ (independent-observation target). This finding suggests that the theoretical risk of a feedback loop from using the analysis field as the scoring target was limited during the study period. Adopting an independent-observation target is recommended in practice, since it yields slightly superior skill and fully eliminates the theoretical concern of an information leakage risk.

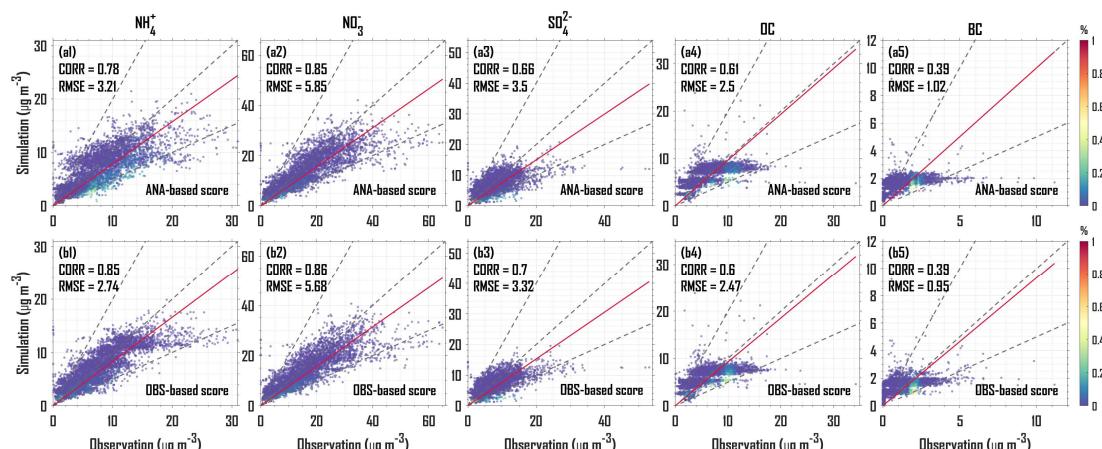


Figure S1: Scatterplots with probability density of simulated versus observed mass

concentrations at independent VE sites correspond to the two scoring targets used in the incremental learning process, including analysis fields (ANA) (a1-a5) and independent observations (OBS) (b1-b5). The gray dotted lines represent the 2:1, 1:1, and 1:2 lines, and the red solid line represents the fitting regression line.”

3) Insufficient experimental period limiting model extrapolation and generalizability

The experimental validation is strictly limited to a two-month period (February–March 2022). This short duration fundamentally undermines the manuscript’s claims regarding the system’s robustness and its “self-evolving” capability, primarily due to the inherent limitations of the chosen machine learning architecture.

From the ML perspective, The OIRF model relies on the Random Forest (RF) algorithm. A well-known limitation of tree-based methods is their inability to extrapolate beyond the range of values encountered in the training data. By restricting the training and validation to a single two-month window, the model is only exposed to a specific subset of atmospheric conditions. If the system encounters pollution episodes more severe or chemically distinct than those in the February–March training set, the RF model will likely “clip” the forecast to the maximum value previously learned, failing to capture new extremes. The current experimental design does not demonstrate that the “incremental learning” mechanism can overcome this fundamental extrapolation barrier when faced with out-of-distribution data.

From the physics perspective, a system trained and validated exclusively on winter/early spring data cannot validly be claimed as a “Self-evolving Data Assimilation System” because it has not been tested against any possible regime shifts of a full annual cycle. There is no evidence presented that the model can “evolve” to handle the volatility of semi-volatile species in warmer months without catastrophic forgetting or significant error.

I strongly encourage the authors to extend their experiment to cover a longer period to genuinely establish the robustness of the incremental learning mechanism. Otherwise the authors need to rescale their claims. For example, the term “self-evolving” should be removed as the system’s evolutionary capability remains unproven beyond Feb–Mar

2022. The authors must also explicitly discuss the theoretical risks of deploying this approach in operational setting outside the training season.

Authors' response:

We sincerely thank the reviewer for the crucial and insightful comment. We fully agree with the reviewer's concerns from both machine learning and physics perspective. Conducting a year-long experiment is crucial for verifying the robustness of the incremental learning mechanism of machine learning models, especially for tree-based models with poor extrapolation capabilities.

a. Re-scaling the claims on “self-evolving”

We fully agree that a two-month experimental period is insufficient to robustly demonstrate “self-evolution” against the full scenarios of atmospheric variability and unprecedented extremes. However, acquiring a year-long hourly observation dataset of five key PM_{2.5} chemical components across a wide spatial range is currently very challenging. To our knowledge, none of the popular reanalysis datasets are generated directly from long-term hourly observations of chemical components. As summarized in **Table R1**, the CAQRA-aerosol dataset was generated indirectly by assimilating ground-level hourly observations of traditional air pollutants. The TAP dataset was generated by fusing daily, monthly, and annual observations of chemical components. The CHAP dataset was generated by fusing daily measurements of four water-soluble inorganic ions. The chemical component fields in both CAMSA and MERRA-2 were generated indirectly by assimilating observations of aerosol optical depth.

Consequently, within the current constraints, we deployed our maximum feasible effort to conduct a two-month hourly measurement campaign at 33 sites. This campaign was designed for a representative period (February-March 2022) and region (Beijing-Tianjin-Hebei region) known for frequent pollution episodes, which directly supports the primary goal of proposing and validating a novel framework online coupling incremental machine learning and ensemble data assimilation. **In response, we have re-scaled the claims on “self-evolving” and replace the term “self-evolving” with more precise descriptions such as “incrementally updatable” in the revised**

manuscript. In the future work, we will extend our measurement campaigns covering a longer period to establish the robustness of the incremental learning mechanism.

b. Discussion on extrapolation risks

In response, we have added a section **3.4 Limitations** to the revised manuscript to discuss the theoretical risks of deploying this approach in operational settings outside the training season. The original section **3.4 Comparison with multiple reanalysis datasets** has been changed to a section **3.3.3. Comparison with multiple reanalysis datasets**.

3.4 Limitations, Line 621-637: “*Although the OIRF model serves as an efficient surrogate for the CTM in generating simulation or forecast ensembles for data assimilation, it inherits a constrained extrapolation capability of tree-based models. Specifically, the OIRF model may exhibit a tendency to saturate at learned extremes when extrapolating beyond its training data distribution, which directly limits its generalizability in diverse and complex atmospheric scenarios, such as the pollution extremes in seasons outside the training period. The poor performance of tree-based models on testing sets has been reported in our previous study (Li et al., 2025). Our incremental learning mechanism is designed to mitigate the extrapolation limitation by dynamically updating the RF model with new knowledge. However, the effectiveness of incremental learning is contingent upon the availability of high-quality analysis fields. A lack of observations, which prevents the generation of analysis fields, exposes the OIRF model to its inherent extrapolation limitations, leading to compromised simulation accuracy.*

Replacing the RF model with an ensemble of deep neural networks (DNNs) holds promise for superior nonlinear mapping and extrapolation. However, the considerably higher computational cost required for both training and inference of DNNs (Debjyoti and Utpal, 2025; Xi, 2022) results in an operational bottleneck that the process of updating and running an ensemble of DNNs can be slower than traditional CTM-based ensemble simulations, which could offset its accuracy advantages. Therefore, balancing the inherent predictive performance of a machine learning model against its

computational cost remains a central challenge for the practical online coupling of machine learning with data assimilation.”

Table R1. The brief description of the observations used in the reanalysis datasets.

Dataset	Reanalysis species	Observed species	Temporal resolution of observation	Citation
CAQRA-aerosol	SO_4^{2-} , NH_4^+ , NO_3^- , OC, BC	$\text{PM}_{2.5}$, PM_{10} , NO_2 , SO_2 , CO , O_3	Hourly	Kong et al., 2025
TAP	SO_4^{2-} , NH_4^+ , NO_3^- , OM, BC	SO_4^{2-} , NH_4^+ , NO_3^- , OC, BC	Daily, monthly, and annual	Liu et al., 2022
CHAP	SO_4^{2-} , NH_4^+ , NO_3^- , Cl^-	SO_4^{2-} , NH_4^+ , NO_3^- , Cl^-	Daily	Wei et al., 2023
CAMSRA	NO_3^- , NH_4^+	Satellite-based AOD	12-hourly	Inness et al., 2019
MERRA-2	SO_4^{2-} , OM, BC	Satellite & ground-based AOD	Hourly	Randles et al., 2017

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The revised figures

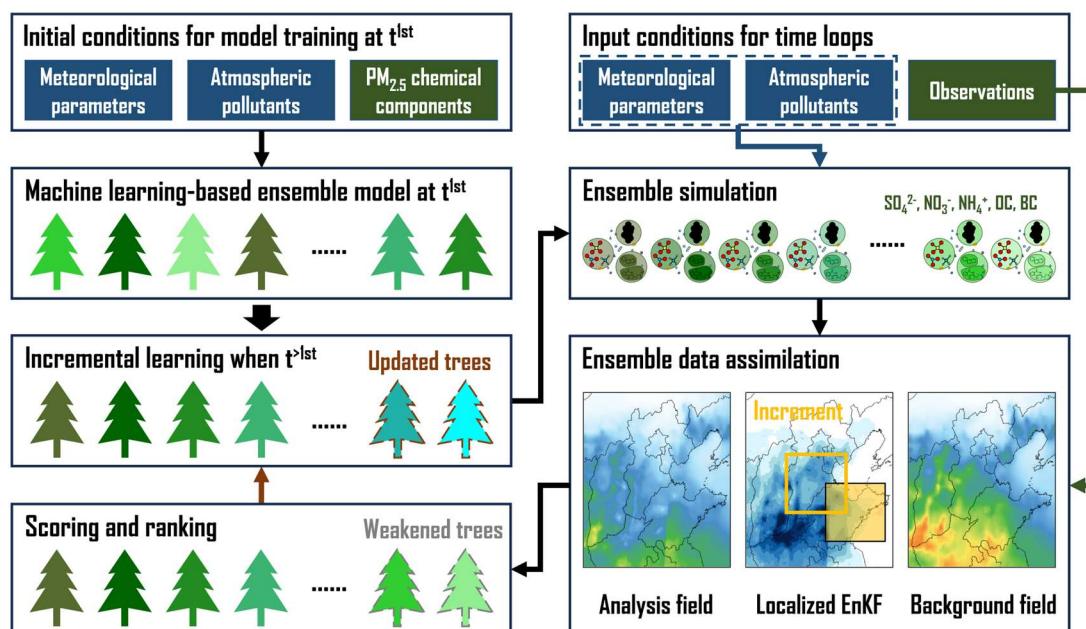


Figure 1. The framework of OIRF-LEnKF v1.0.

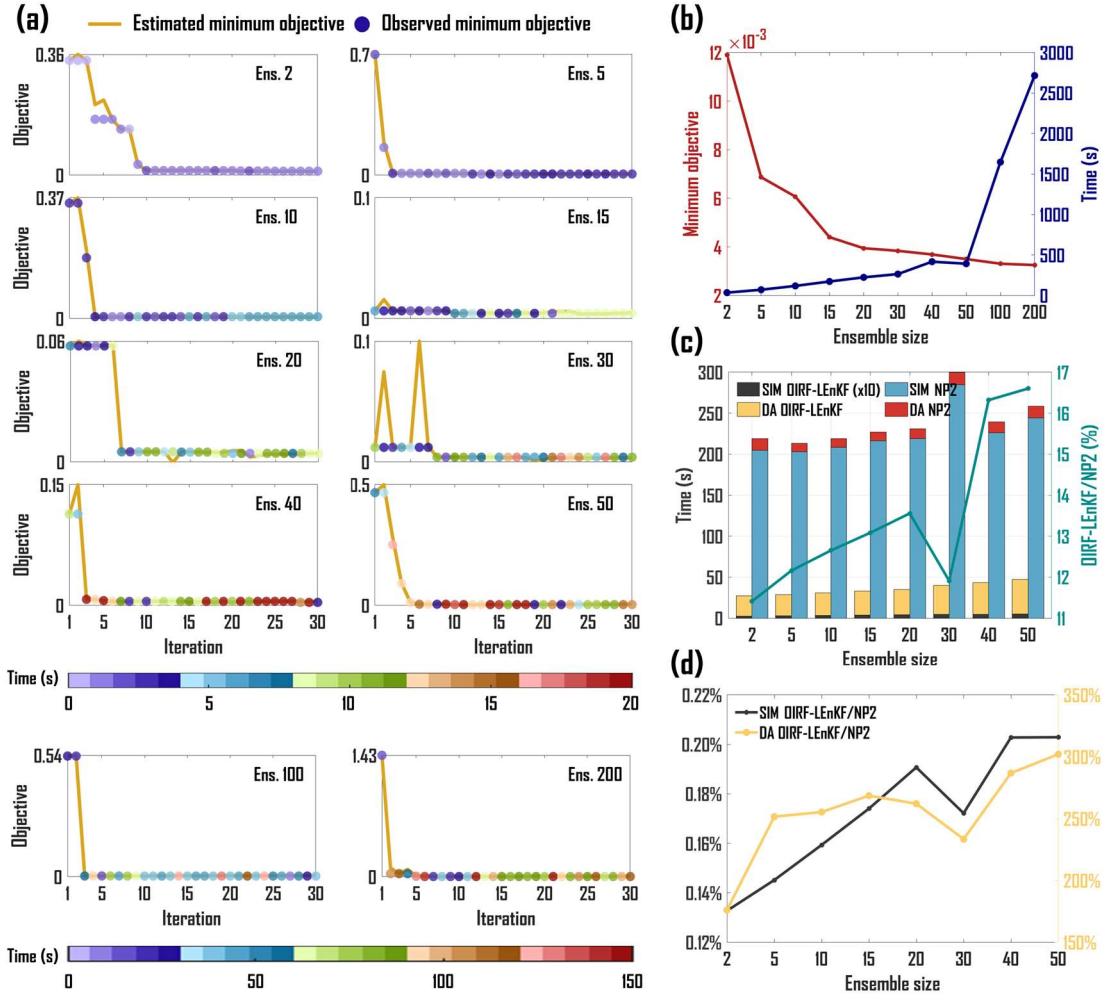


Figure 3. Computational efficiency of OIRF-LEnKF v1.0. (a) Variation in the minimum objective value throughout the Bayesian optimization process and time consumed by each iteration, determined by Eq. (5). (b) minimum value of total observed minimum objectives and total time consumed during Bayesian optimization process for different ensemble sizes, (c) time consumed by model simulation and data assimilation at each timestep for OIRF-LEnKF and NAQPMS-PDAF v2.0 (NP2), and the ratio of total time consumed between OIRF-LEnKF and NP2, (d) the ratio of time consumed by model simulation and data assimilation between OIRF-LEnKF and NP2. SIM represents the simulation phase, and DA represents the data assimilation phase. The elapsed time of the OIRF-LEnKF simulation process in Figure 3c has been magnified by a factor of 10 for better clarity.

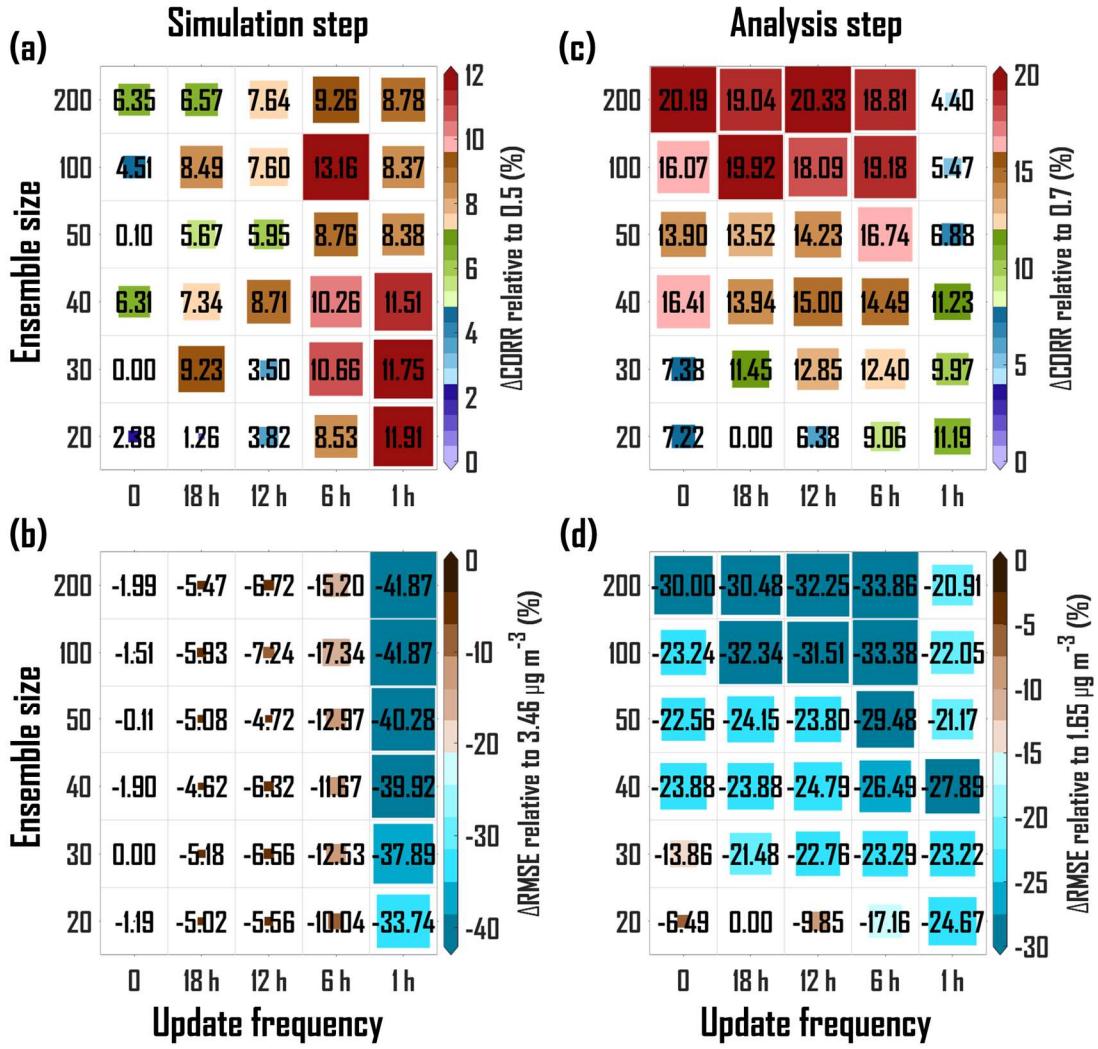


Figure 4. (a) Percentage change of Pearson correlation coefficient (CORR) relative to the minimum CORR (0.5) (ΔCORR , %) for sensitivity test with six ensemble sizes (20, 30, 40, 50, 100, 200) and five update frequencies (no update, 18-hour interval, 12-hour interval, 6-hour interval and 1-hour interval) at the simulation step. (b) Same as (a) but for percentage change of root mean square error (RMSE) relative to the maximum RMSE ($3.46 \mu\text{g m}^{-3}$) (ΔRMSE , %) at the simulation step. (c) Same as (a) but for percentage change of CORR relative to the minimum CORR (0.7) at the analysis step. (d) Same as (a) but for percentage change of RMSE relative to the maximum RMSE ($1.65 \mu\text{g m}^{-3}$) at the analysis step.

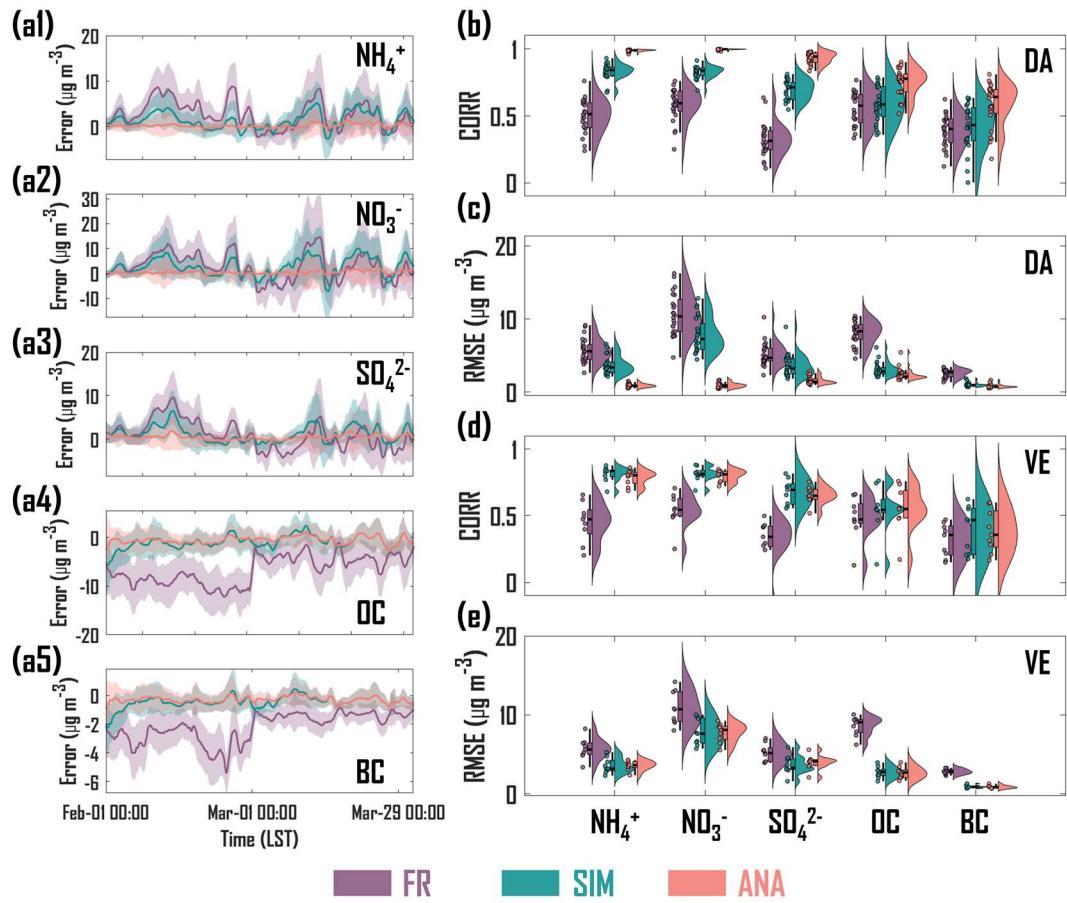


Figure 5. Smoothed variation in the error between observation and model output (including the free-run field (FR), the ML-simulated background field (SIM) and the analysis field (ANA)) for (a1) NH_4^+ , (a2) NO_3^- , (a3) SO_4^{2-} , (a4) OC and (a5) BC at total sites during February and March of 2022. The lines and shading areas represent the mean and standard deviation of the errors, respectively. (b) Correlation coefficient (CORR) between observation and model output for five $\text{PM}_{2.5}$ chemical components at DA sites. (c) Same as (b) but for root mean square errors (RMSE). (d) Same as (b) but for VE sites. (e) Same as (b) but for RMSE at VE sites.

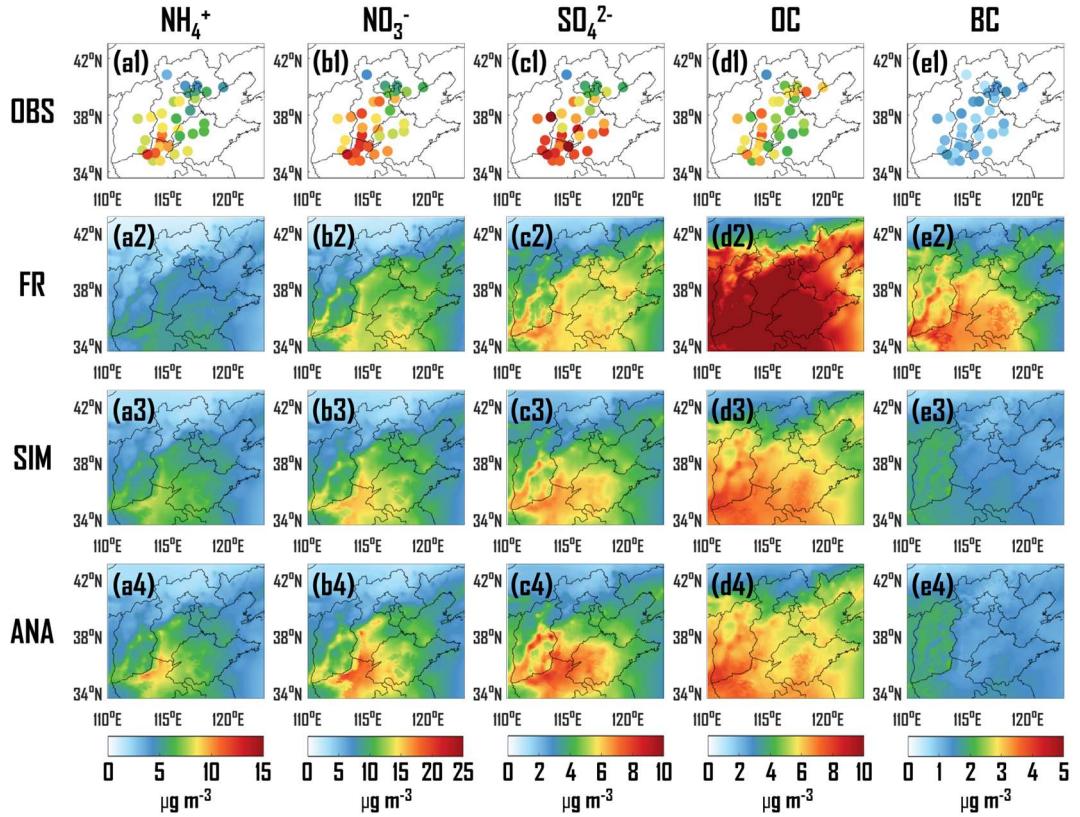


Figure 6. Spatial distribution of observation (OBS), free-run field (FR), ML-simulated background field (SIM) and analysis field (ANA) for NH_4^+ (a1-a4), NO_3^- (b1-b4), SO_4^{2-} (c1-c4), OC (d1-d4) and BC (e1-e4).

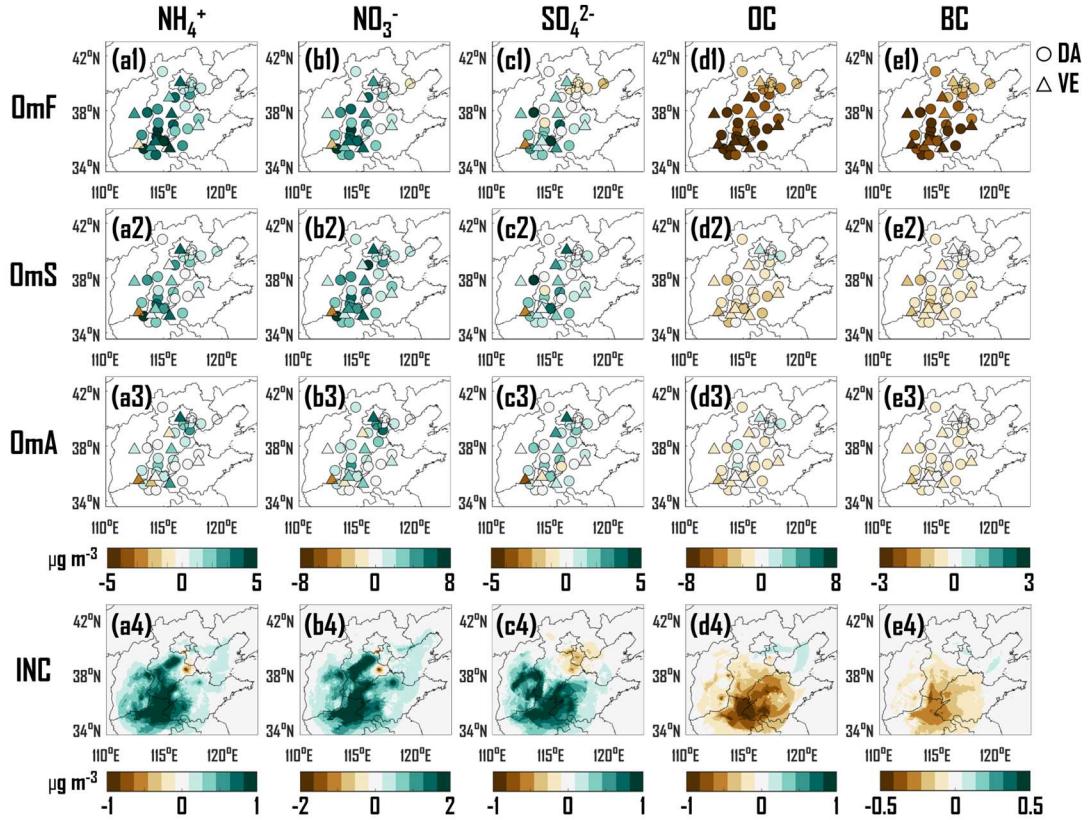


Figure 7. Spatial distribution of observation minus free-run field (OmF), observation minus ML-simulated background field (OmS), observation minus analysis field (OmA) and analysis field minus background field (INC) for NH_4^+ (a1-a4), NO_3^- (b1-b4), SO_4^{2-} (c1-c4), OC (d1-d4) and BC (e1-e4). The circle indicates the DA sites with data assimilation, and the upward-pointing triangle indicates the VE sites without data assimilation.