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An optimized semi-empirical physical approach for satellite-based $PM_{2.5}$ retrieval: embedding machine learning to simulate complex physical parameters

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ABSTRACT

Satellite remote sensing of $PM_{2.5}$ mass concentration has become one of the most popular atmospheric research aspects, resulting in the development of different models. Among them, the semi-empirical physical approach constructs the transformation relationship between the aerosol optical depth (AOD) and $PM_{2.5}$ based on the optical properties of particles, which has strong physical significance. Also, it performs the $PM_{2.5}$ retrieval independently of the ground stations. However, due to the complex physical relationship, the physical parameters in the semi-empirical approach are difficult to calculate accurately, resulting in relatively limited accuracy. To achieve the optimization effect, this study proposes a method of embedding machine learning into a semi-physical empirical model (RF-PMRS). Specifically, based on the theory of the physical $PM_{2.5}$ remote sensing approach (PMRS), the complex parameter (VE_f, a columnar volume-to-extinction ratio of fine particles) is simulated by the random forest model (RF). Also, a fine mode fraction product with higher quality is applied to make up for the insufficient coverage of satellite products. Experiments in North China show that the surface $PM_{2.5}$ concentration derived by RF-PMRS has an average annual value of 57.92 μg/m³ versus the ground value of 60.23 μg/m³. Compared with the original

method, RMSE decreases by 39.95 μg/m³, and the relative deviation reduces by 44.87%. Moreover, validation at two AERONET sites presents a trend closer to the true values, with an R of about 0.80. This study is also a preliminary attempt to combine model-driven and data-driven models, laying a foundation for further atmospheric research on optimization methods.

Keywords: $PM_{2.5}$; Physical approach; Machine learning; Volume-to-extinction ratio; Fine mode fraction

1. Introduction

Epidemiological studies have indicated that $PM_{2.5}$ (fine particulate matter with an aerodynamic equivalent diameter no greater than 2.5 μm) can adversely affect human health, such as increasing the risk of diabetes and respiratory diseases (Bowe et al., 2018; Pope III et al., 2002; Xu et al., 2013), and accurate surface $PM_{2.5}$ concentration is the basis of air pollution-health related research. Satellite remote sensing has the advantages of high resolution and global coverage (Ma et al., 2014; Wu et al., 2020), including variables strongly associated with $PM_{2.5}$ such as aerosol optical depth (AOD). Therefore, it has become a mainstream method for fine particles estimation (Zhang et al., 2021).

There are mainly three satellite-based ways of retrieving $PM_{2.5}$. 1) Chemical transport models-based method. It calculates a scaling factor η between AOD and $PM_{2.5}$ simulated by atmospheric chemical transport models (CTM) (Lyu et al., 2022) and then transfers the proportional relationship to satellite AOD data when calculating surface $PM_{2.5}$ concentration (Geng et al., 2015; Van Donkelaar et al., 2006). However, the assumption of a constant factor between simulated and observed values has large spatiotemporal limitations. 2) Univariate/Multivariate regression. This kind of method establishes a statistical model between AOD, auxiliary variables, and ground $PM_{2.5}$ observations. Machine learning is a common tool for such data-driven methods due to its powerful nonlinear fitting ability between multiple variables (Irrgang et al., 2021). But the regression is affected by the distribution and density of ground stations (Gupta and Christopher, 2009; Li et al., 2017). 3) Semi-empirical physical approach. Taking

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the physical theory as the basis, surface $PM_{2.5}$ is derived through an empirical formula constructed from AOD and some PM-related key parameters, including an important empirical parameter related to the optical properties (S). The process steps are explicit and independent of ground station observations. Meanwhile, this approach has stronger physical interpretability than the previous two methods with a large space for optimization.

Due to the complexity of the physical parameters, many studies have optimized the semi-empirical physical approach. Raut and Chazette (2009) introduced a specific extinction cross-section to simplify the expression of S and $PM_{2.5}$ concentration was estimated based on 355nm-band radar observations. Kokhanovsky et al. (2009) constructed a particle effective radius model, which can obtain the particle concentrations throughout the atmospheric column. Furthermore, Zhang and Li (2015) proposed the physical $PM_{2.5}$ remote sensing method (PMRS). It replaced S by defining a volume-to-extinction ratio of fine particles (VE_f) and used a quadratic polynomial of fine mode fraction (FMF) to simulate VE_f, showing certain advantages (Li et al., 2016; Zhang et al., 2020).

However, the above semi-physical empirical models have some shortcomings. Firstly, the satellite data used in the models are blocked by clouds and fog in some areas, thus high-coverage and high-precision products need to be excavated and applied; secondly, there are still large uncertainties in estimating physical parameters (such as a simple polynomial fit to S in the PMRS method) and their expressions need to be improved. To date, machine learning (ML) has developed rapidly. It can detect complex nonlinear relationships of multiple data and model their interaction (Yuan et al., 2020; Lee et al.,2022), which provides an idea for improving the accuracy of physical parameter acquisition, thereby estimating high-precision $PM_{2.5}$ through semi-physical empirical models.

According to this idea, our study proposes an optimized semi-empirical physical model (RF-PMRS) based on the PMRS theory, which attempts to explore the possibility of combining physical models and ML. To be specific, we creatively embed ML (the random forest model) into the PMRS method to simulate the physical parameter (i.e.,

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VE_f) derived from FMF and related variables, thus optimizing the previous polynomial expression. Besides, to further improve the $PM_{2.5}$ retrieval accuracy, the physical-deep learning FMF (Phy-DL FMF) dataset generated by a hybrid retrieval algorithm of ML and physical mechanisms is introduced. Ultimately, we comprehensively validate the performance of the $PM_{2.5}$ obtained by our optimized approach.

The remained part of our article is as follows. Section 2 illustrates the specific derivation process of the proposed method. Section 3 describes the experimental datasets and analyzes the evaluation results. Some supporting experiments are discussed in section 4. And the final part provides the conclusion.

2. Methods

Based on the basic physical properties of atmospheric aerosols, the semi-physical empirical approach starts from the integration of PM mass concentration and AOD. Then it combines several key factors related to $PM_{2.5}$, to derive the in situ $PM_{2.5}$ concentration through multiple remote sensing variables (Koelemeijer et al., 2006). The overall empirical relationship can be represented as:

$$PM_{2.5} = AOD\frac{\rho}{H \cdot f(RH)}S \tag{1}$$

where ρ denotes the particle density and H denotes the atmospheric boundary layer height. $f(RH)$ represents the hygroscopic growth factor related to relative humidity (RH). S is an optical characteristic parameter that should be simulated.

2.1. PMRS method

2.1.1. The expression of VE_f

To illustrate S more precisely, PMRS defines the columnar volume-to-extinction ratio of fine particles (i.e., VE_f), which can be regarded as the basis of our optimization method. So equation (1) is transformed into:

$$PM_{2.5} = AOD\frac{\rho}{H \cdot f(RH)}VE_f \tag{2}$$

Related to particle size, aerosol extinction, and other properties, VE_f can be

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expressed as:

$$VE_f = \frac{V_{f,column}}{AOD_f} \tag{3}$$

$$AOD_f = AOD \cdot FMF \tag{4}$$

Here, AOD_f is the fine particle AOD and FMF is the fine mode fraction. $V_{f,column}$ can be expressed by the vertical integral of particle volume size distributions (PVSD) within a certain aerodynamic diameter range:

$$V_{f,column} = \int_0^{D_{p,c}} V(D_p) dD_p \tag{5}$$

$D_{p,c}$ represents the cutting diameter, and the empirical value of 2.0 μm is chosen based on previous literature (Hand and Kreidenweis, 2002; Hänel and Thudium, 1977). And $V(D_p)$ represents the PVSD corresponding to the geometric equivalent diameter (D_p).

2.1.2. Specific process and limitations

The PMRS method is developed from equation (2). Based on satellite AOD, the near-surface $PM_{2.5}$ can be obtained through multi-step transformation. Fig. 1(a) shows its specific process. Each arrow refers to a step, respectively: size cutting (output: AOD_f), volume visualization (output: $V_{f,column}$), bottom isolation (output: V_f, fine particle volume near the ground), particle drying (output: $V_{f,dry}$, dry V_f) and $PM_{2.5}$ weighting. The overall expression is as follows:

$$PM_{2.5} = AOD \frac{FMF \cdot VE_f \cdot \rho_{f,dry}}{PBLH \cdot f_0(RH)} \tag{6}$$

$$f_0(RH) = \left(1 - \frac{RH}{100}\right)^{-1} \tag{7}$$

where FMF denotes the fine mode fraction, $\rho_{f,dry}$ denotes the dry mass density of $PM_{2.5}$, and $PBLH$ represents the planet boundary layer height. $f_0(RH)$ represents the approximation of $f(RH)$ in equation (2), as expressed as equation (7). Considering the aerosol types in different regions, PMRS fits VE_f to a quadratic polynomial relation of FMF:

$$VE_f = 0.2887 FMF^2 - 0.4663 FMF + 0.356 \quad (0.1 \le FMF \le 1.0) \tag{8}$$

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PMRS has strong physical significance, the calculation steps are well-defined and site-independent. Zhang and Li (2015) tested the performance of PMRS on 15 stations, and the validation results had an uncertainty of 34%. Compared with the ground value of Jinhua city in China, a 31.3% relative error was generated in Li et al. (2016). Besides, Zhang et al. (2020) applied it to the $PM_{2.5}$ change analysis and prediction experiments in China over 20 years. However, there may be a more complex nonlinear relationship between VE_f with FMF, not just a simple quadratic formula. Since VE_f is related to the aerosol type, adding other spatiotemporal variables may optimize the fitting process. Additionally, high-quality FMF data is the basic guarantee for the estimated $PM_{2.5}$ quality. In a word, to further improve the physical method, a better nonlinear model between VE_f and related variables from reliable datasets needs to be explored.

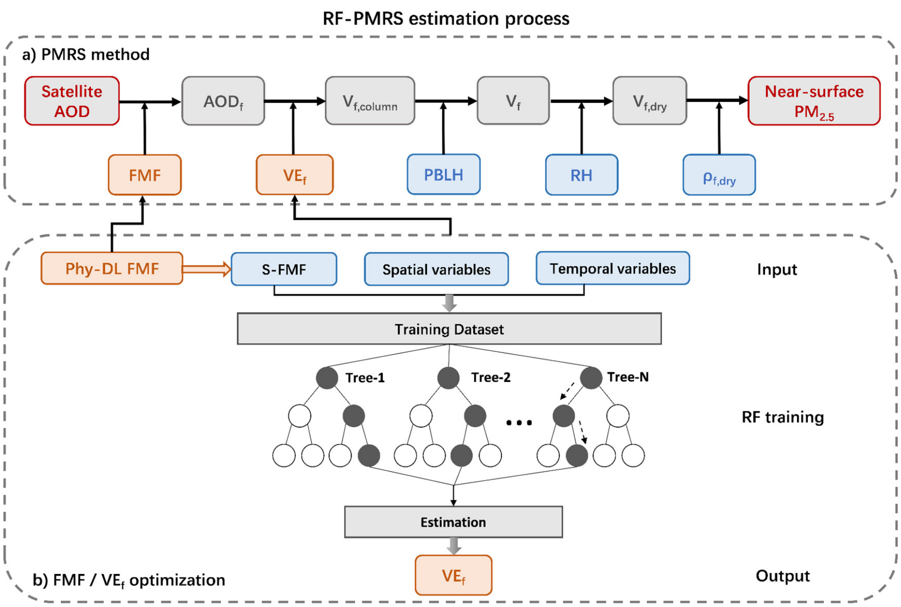

Fig. 1. Surface $PM_{2.5}$ estimation flow of RF-PMRS. a) The five steps of the PMRS method. Gray boxes are the intermediate outputs, blue boxes are the input data, and orange ones denote the variables to be optimized. b) The specific optimization of RF-PMRS: FMF dataset replacement and VE_f simulation by RF model.

2.2. Optimization method: RF-PMRS

Therefore, to overcome the above disadvantages, an optimized method called RF-

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PMRS is proposed. Fig. 1(b) shows the process of our method, while optimizations for FMF and VE_f are described separately below.

1) FMF dataset selection

We introduce the Phy-DL FMF dataset into the PMRS method to improve the accuracy of size-cutting results. In the comparison experiment against Aerosol Robotic Network (AERONET) FMF, Phy-DL FMF shows a higher accuracy (R = 0.78, RMSE = 0.100) than Moderate-resolution Imaging Spectroradiometer (MODIS) FMF (R = 0.37, RMSE = 0.282) (Yan et al., 2022). Also, it performs better spatiotemporal continuity.

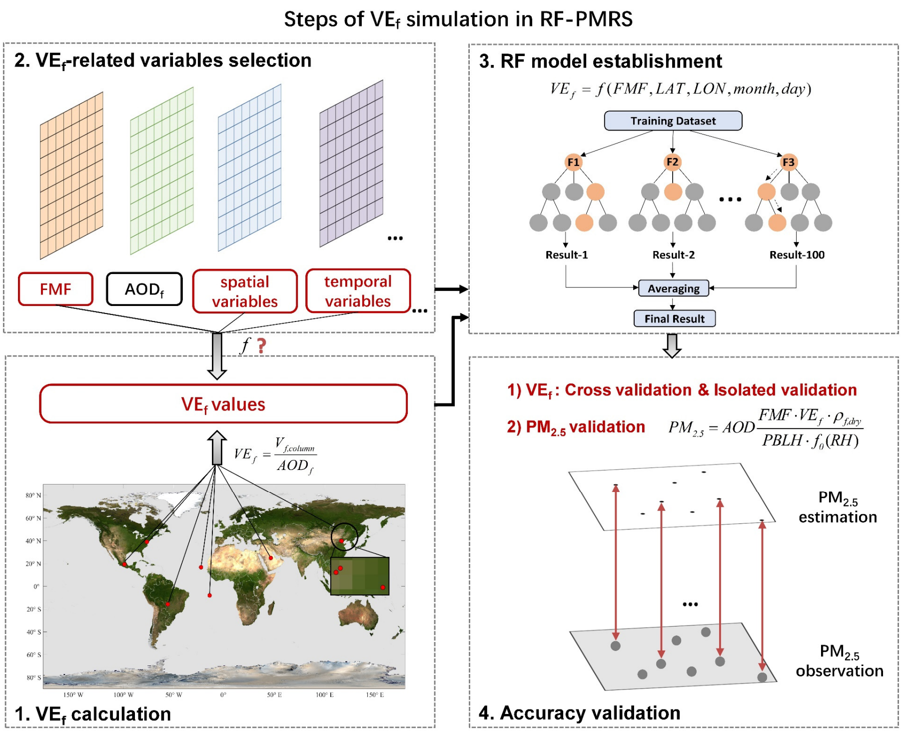

Fig. 2. Specific steps for simulating VE_f based on ML in our RF-PMRS method. The map used in the step 1 is from NASA Visible Earth (https://visibleearth.nasa.gov/images/57752/blue-marble-land-surface-shallow-water-and-shaded-topography). The red points in step 1 represent the distribution of the 9 AERONET sites.

2) VE_f simulation based on ML

The main idea is to establish an ML model between the VE_f truth obtained from multiple AERONET sites and related variables, thus improving the subsequent VE_f-simulation accuracy (Fig. 2).

Step 1 VE_f calculation

The VE_f true values are calculated concerning equations (3)-(5). A total of 9 AERONET sites corresponding to four typical aerosol types participate in the training. Table 1 shows the specific information.

Table 1. Data information of 9 AERONET sites classified by aerosol types. Location indicates the latitude and longitude, where '-' means the south latitude and west longitude. Two sites in bold fonts participate in the $PM_{2.5}$ validation experiment.

Aerosol Type	Site	Location (LAT, LON)	Training period	Isolated-validation period
Urban–industrial	**Beijing**	**39.98°, 116.38°**	2001-2017	2018-2019
	Beijing-CAMS	**39.93°, 116.32°**	2012-2017	2018-2019
	XiangHe	39.75°, 116.96°	2004-2017	/
	Ascension Island	-7.98°, -14.41°	2010-2017	2018-2019
	Capo Verde	16.73°, -22.94°	2010-2017	2018
Biomass burning	CUIABA MIRANDA	-15.73°, -56.07°	2010-2017	2018-2019
Desert dust	GSFC	38.99°, -76.84°	2010-2017	2018-2019
	Mexico City	19.33°, -99.18°	2010-2017	/
Oceanic	Solar Village	24.91°, 46.40°	2010-2013	/

Step 2 VE_f-related variables selection

According to the theory, FMF is selected as the most important modeling variable. Previous studies have also shown that the FMF-VE_f relationship has a good single-value correspondence, which is not affected by AOD. Compared with AOD_f and $V_{f,column}$, FMF is a better indicator for estimation (Zhang and Li 2015). In addition, considering the spatiotemporal heterogeneity of VE_f, the latitude, longitude (LAT, LON), and data time (month, day) of each site are added to the training.

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Step 3 RF model establishment

From step 2, VE_f can be expressed as:

$$VE_f = f(FMF, LAT, LON, month, day) \tag{9}$$

We optimize VE_f expression based on random forest (RF). RF is made up of multiple decision trees that can build high-accuracy models based on fewer variables (Yang et al., 2020). This ensemble supervised learning method randomly samples the original dataset into multiple sets and considers random subsets of features in node splitting, which reduces correlation and the sensitivity to noise (Belgiu and Drăguţ, 2016). Note that the station FMF values ($S\text{-}FMF$) are used when training.

Step 4 Accuracy validation

The VE_f estimation is also based on equation (9), where *f* is the optimal relationship after RF parameter adjustment, and Phy-DL FMF is applied to realize the extension of model results from point to surface. 10-fold cross-validation (Rodriguez et al., 2009) and isolated-validation are used to evaluate model performance (see Appendix A1).

3) $PM_{2.5}$ value estimation and evaluation

Then, calculate $PM_{2.5}$ according to the corresponding process (equation (6)). The statistical indicators used in the evaluation include correlation coefficient (R), mean bias (MB), relative mean bias (RMB), root mean square error (RMSE), and mean absolute error (MAE). In addition, relative predictive error (RPE) is added to validate the accuracy of the RF-based VE_f model. See Appendix A2 for the specific information on these indicators.

3. Experiment data and results

3.1. Data

3.1.1. MODIS AOD

MCD19A2, the MODIS C6 Level-2 gridded (L2G) land AOD product, is selected in this study. It is derived by the Multi-Angle Implementation of Atmospheric Correction

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(MAIAC) algorithm, which can improve the accuracy in cloud detection and aerosol retrieval (Lyapustin et al., 2011). Besides, this new advanced algorithm jointly combines MODIS Terra and Aqua into a single sensor (Lyapustin et al., 2014). The product is produced daily with a 1km resolution, including aerosol parameters such as 470nm/550nm AOD, quality assurance (QA), and uncertainty factors.

The processing of MCD19A2 data (HDF format) is mainly divided into five steps: AOD/QA band extraction, best quality AOD selection, Terra/Aqua data synthesis, missing information reconstruction, and mosaic. Finally, the daily AOD distribution in GeoTiff format is obtained.

3.1.2. Phy-DL FMF dataset

To enhance the reliability of the global land FMF product, Yan et al. (2022) have released a satellite-based dataset (daily scale) called Phy-DL FMF, which integrates physical and deep learning methods. The product has a spatial resolution of 1° and covers from 2001 to 2020. In terms of performance, it exhibits higher accuracy and wider space-time coverage than satellite products (Yan, 2021).

3.1.3. Meteorological data

The values of PBLH and RH are obtained from the ERA5 dataset. As the fifth-generation reanalysis product released by the European Center for Medium-Range Weather Forecasts (ECMWF), ERA5 provides atmospheric data at 0.25° every hour based on the data assimilation principle (Hersbach et al., 2018). It should be noted that RH is not archived directly in ERA5, thus should be calculated by 2m temperature T and dew point temperature T_d (referred to ERA-Interim: documentation).

$$RH = 100 \times \frac{e_s(T_d)}{e_s(T)} \tag{10}$$

Here, $e_s(t)$ represents the saturation vapor pressure related to a Celsius temperature t (Simmons et al., 1999).

$$e_s(t) = 6.112 \times \exp\left(\frac{17.67 \times t}{t + 243.5}\right) \tag{11}$$

3.1.4. AERONET data

The Aerosol Robotic Network (AERONET) is a federation of ground-based sun-sky radiometer networks, providing worldwide remote sensing aerosol data for more than 25 years (Holben et al., 1998). Until now, the Version 3 dataset has been released (Giles et al., 2017). Due to its high quality, the data from AERONET have been regarded as theoretical true values to evaluate satellite-based products in related studies (Chen et al., 2020; Gao et al., 2016; Wang et al., 2019). AOD, FMF, and Volume Size Distribution products with Level 2.0 (quality-assured) are applied to implement our purpose.

3.1.5. Ground $PM_{2.5}$ measurements

The near-surface hourly $PM_{2.5}$ values are obtained from the China National Environmental Monitoring Center (CNEMC). Nowadays, over 1600 ground-based monitors are working continuously and a total of 232 stations (2017) in the North China Region (NC) participate in this work. Fig. 3 displays the site distributions of our validation area.

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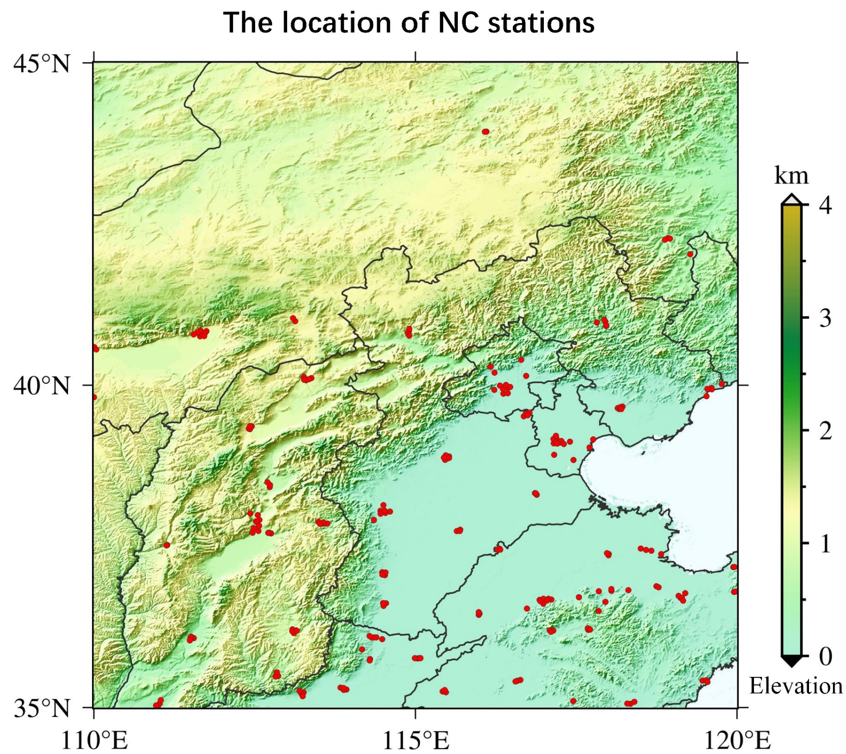

Fig. 3. The location of ground stations in the NC region (35°-45°N, 110°-120°E). The red points represent NC stations.

The above variables are spatially matched to ground sites at their respective resolutions. And based on UTC, the experiment is conducted on a daily scale in 2017. Note that we select the measured empirical value of $\rho_{f,dry}$ (i.e., 1.5 g/cm^3) for the NC region from Gao et al. (2007).

3.2. Experimental results

3.2.1. RF model performance for training VE_f

The simulation model of VE_f is trained based on the data in Table 1 and see Appendix A3 for the adjustment of the model parameters. Table 2 shows that RF can capture the complex relationship between VE_f and related variables well. R is as high as 0.974 (0.975), RMSE and MAE are both small, and RPE is around 30%, which suggests the desired estimation accuracy. Overall, the CV results represent the great performance of

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the RF model for extracting information, that is, the relationship of multi-source data to VE_f. In the meantime, the statistical results in CV and IV experiments are similar, indicating that the RF model has no obvious overfitting phenomenon.

Table 2. Performance statistics of the RF model for training VE_f. N represents the number of data, and VE_f has no unit.

	R	RMSE	RPE	MAE	N
Cross-validation（CV）	0.974	0.076	32.9%	0.034	6463
Isolated-validation（IV）	0.975	0.067	29.8%	0.037	814

3.2.2. Accuracy evaluation of PMRS/RF-PMRS at AERONET stations

After applying the Phy-DL FMF data to the calculation process, the experiment compares $PM_{2.5}$ results of PMRS and RF-PMRS at Beijing (BJ) and Beijing-CAMS (BC) AERONET sites in 2017. Here, RF-PMRS simulates VE_f based on RF, replacing the polynomial of the PMRS method. Note that the results of the two sites are compared with their respective nearest ground $PM_{2.5}$ stations (distances of 3.64 km and 3.91 km, respectively, in line with the representative range of ground stations in previous studies (Shi et al., 2018)).

Fig. 4 displays the $PM_{2.5}$ value trends of different models at two sites. The blue line fits the red line better than the gray one, confirming that the $PM_{2.5}$ results of RF-PMRS are closer to the true values. Within the range of the black circles at positions 1 and 2, the variation trend of RF-PMRS results has better consistency with the ground truth, while the PMRS results show dislocation and excessive growth. The overall performance of the RF-PMRS estimations can signify the effectiveness of our proposed method framework. As observed in the red boxes at positions 3 and 4, both models have a certain degree of deviation, which is found to be consistent with the time regularity of the AOD high values. It is worth noting that our method has well mitigated the apparent overestimation of the original model (PMRS) in the case of above-normal aerosol loadings. Furthermore, the average $PM_{2.5}$ values from ground stations, PMRS, and RF-PMRS are compared. As for the two sites, the RF-PMRS results are satisfactory. As depicted in Fig. 5, the RF-PMRS and station mean values are close, with a difference

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of 4.82 μg/m³ (BJ) and 2.73 μg/m³ (BC), suggesting a good estimation. Nevertheless, the PMRS results have deviations greater than 40 μg/m³, and overestimation basically exists at both sites. It can be inferred that, in our proposed method, the optimization of VE_f can greatly improve the $PM_{2.5}$ estimation accuracy.

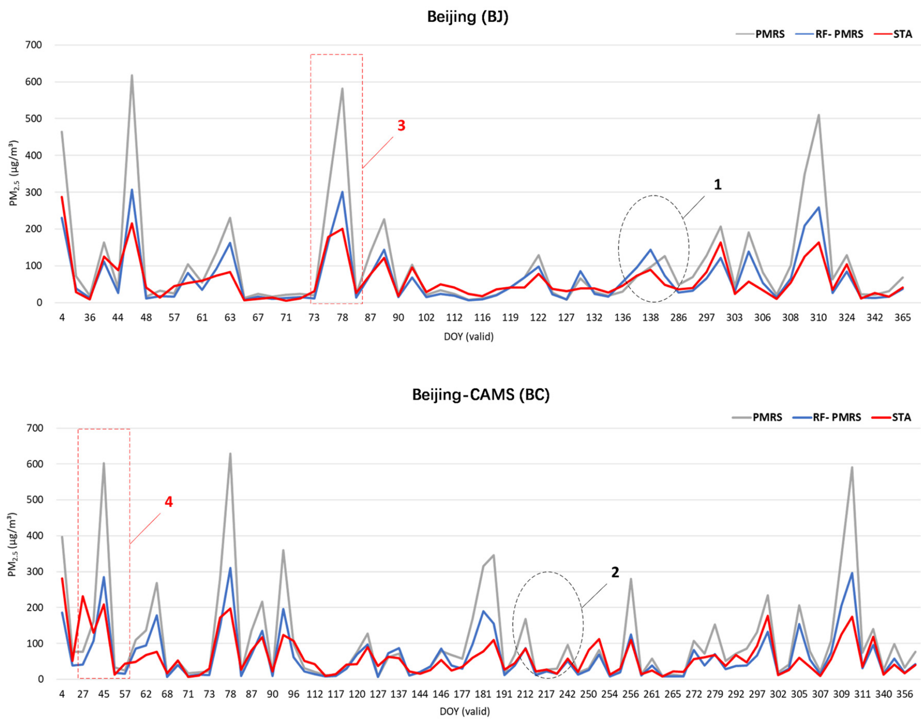

Fig. 4. Three $PM_{2.5}$ trends at the Beijing (BJ) and Beijing-CAMS (BC) sites under their respective valid DOYs in 2017. Grey, blue, and red lines represent $PM_{2.5}$ values of PMRS, RF-PMRS, and stations (STA), respectively. The red boxes and black circles select a specific period for analysis.

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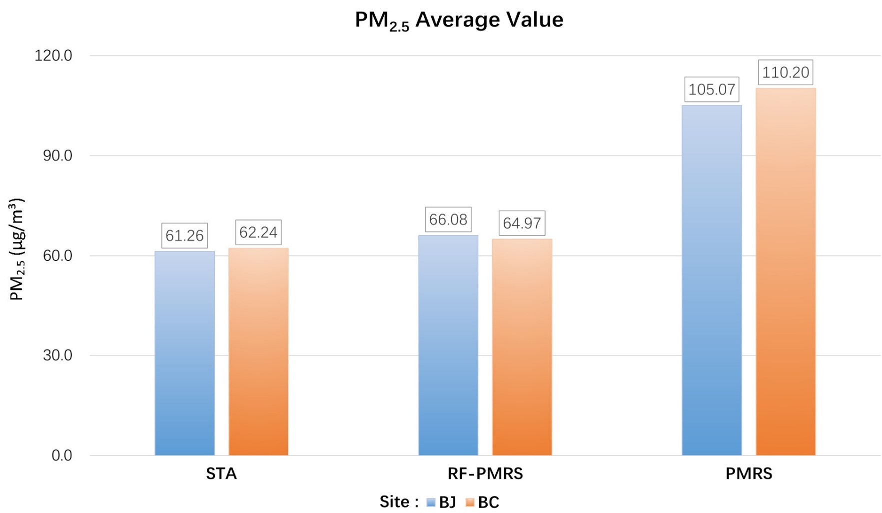

Fig. 5. Annual average $PM_{2.5}$ values from stations (left), RF-PMRS (middle), and PMRS model (right) at the BJ and BC sites.

Aiming at visually comparing the optimization effect, Fig. 6 plots the $PM_{2.5}$ bias distribution patterns for two methods. From the boxplot, the average $PM_{2.5}$ bias of RF-PMRS is close to zero (less than 5 μg/m³), which is greatly lower than that of PMRS. Besides, PMRS $PM_{2.5}$ has a larger deviation range, which manifests in two aspects. One is the maximum bias, specifically, it has exceeded 100 μg/m³ at the BC site. The other is the overall distribution of the data bias, the BJ site ones are mostly distributed below zero, indicating an obvious overestimation. As for RF-PMRS, the above circumstances are not obviously reflected in it. In addition, as can be seen from the indicators, RMSE and MAE of RF-PMRS $PM_{2.5}$ decrease by about half in comparison with PMRS. And the experiment has confirmed that the RF-PMRS $PM_{2.5}$ values have a strong linear relationship with the ground truth at both sites, with R around 0.8 (0.82 at BJ and 0.78 at BC). Such a large optimization effect is attributed to the VE_f expression replacement to the fitted RF model.

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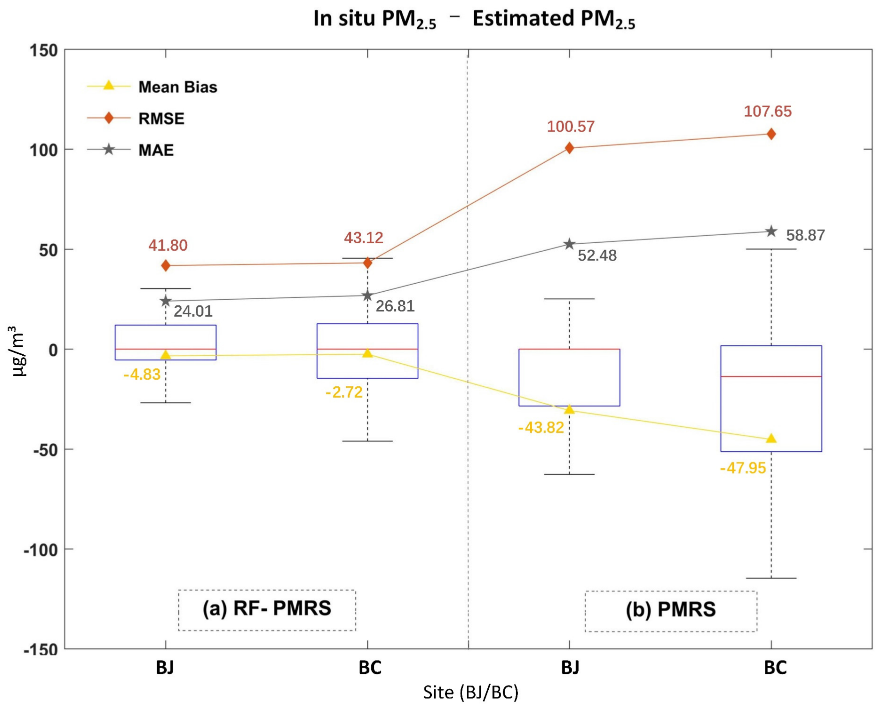

Fig. 6. Boxplots of RF-PMRS (a) and PMRS (b) $PM_{2.5}$ bias at the BJ and BC sites. The upper (lower) black line of each box represents the largest (smallest) value, the blue upper (lower) border represents the upper (lower) quartile, and the red line denotes the median. Besides, the yellow, orange and gray symbols are the MB, RMSE, and MAE of the corresponding $PM_{2.5}$ concentration.

3.2.3. Generalization performance of RF-PMRS

Then, we estimate $PM_{2.5}$ based on PMRS and RF-PMRS within North China (Fig. 3 exhibits the distribution pattern of the validation stations). Table 3 shows the accuracy statistics. It can be seen that RF-PMRS greatly reduces the bias (about 44.87%), with MB of about 2.31 μg/m³. Similar to the results at the sites, the RF-PMRS method can derive $PM_{2.5}$ concentration with practically no overestimation (underestimation). Although there is not much difference in R values of the two models (R of RF-PMRS is only improved by 0.01), RMSE and MAE of which decrease by about 39.96 μg/m³ and 18.86 μg/m³, respectively. As a result, the optimized method deserves to be considered excellent.

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Table 3. Validation results of PMRS and RF-PMRS $PM_{2.5}$ in North China.

Method	R	MB (μg/m³)	RMB (%)	RMSE (μg/m³)	MAE (μg/m³)
PMRS	0.69	-29.34	48.71%	79.98	44.72
RF-PMRS	0.70	2.31	3.84%	40.02	25.86

Meanwhile, $PM_{2.5}$ scatterplots are presented below. As depicted in Fig. 7, there are sufficient estimated samples (28305) in the NC region, which guarantees the credibility of our validation results. In general, the RF-PMRS $PM_{2.5}$ values are distributed around the true values evenly, with a slightly higher R of 0.70 compared to that of the original method. And the slope of the linear fitting relationship reaches 0.82, which indicates that the proposed method greatly reduces the overestimation of PMRS with a linear slope of 1.46. Although the overall performance of the RF-PMRS estimations maintains an excellent level, defects do remain. To be specific, in areas with high $PM_{2.5}$ concentration (especially greater than 150 μg/m³), RF-PMRS results exist a slight underestimation. It may be caused by the relatively small number of high-value points (only 1319 out of 28305), which is difficult to adequately reflect the fitting effect of the method.

As for RF-PMRS, the deviation is reduced to a large extent, so the probability density function maps based on the bias of PMRS and RF-PMRS are further drawn. Fig. 8 visualizes the probability densities within different bias ranges. In terms of distribution characteristics, the overall bias of RF-PMRS from the zero value (black solid line) is small. With regard to the curve shape, it is high and narrow, manifesting that the bias has a lower standard deviation (STD) and is more prone to appear around the mean. However, PRMS shows a more discrete distribution pattern, and there are many outliers outside the range of greater than 600 μg/m³. Simultaneously, as can be concluded from the three boxes, within the bias range of ±20 μg/m³ and ±40 μg/m³, the data numbers of RF-PMRS results increase by 8.32% and 12.81%, respectively. Outside the range of ±100 μg/m³, the number decreases by 9.10%. Therefore, as far as the accuracy is concerned, RF-PMRS results have lower bias and better stability.

In a word, the above analysis demonstrates that compared with the simple quadratic

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polynomial relationship (equation (8)), the established RF model in RF-PMRS can more accurately capture the relationship between VE_f and multiple variables, thereby improving the $PM_{2.5}$ estimation accuracy.

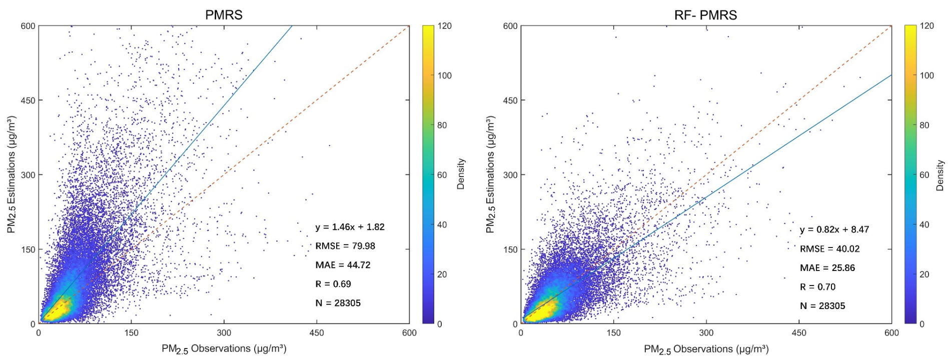

Fig. 7. Validation scatterplots of $PM_{2.5}$ results from PMRS (left) and RF-PMRS (right). Red dashed lines are 1:1 reference lines, and blue solid lines stand for the linear fits. The right legends show the point densities (frequency) represented by different colors.

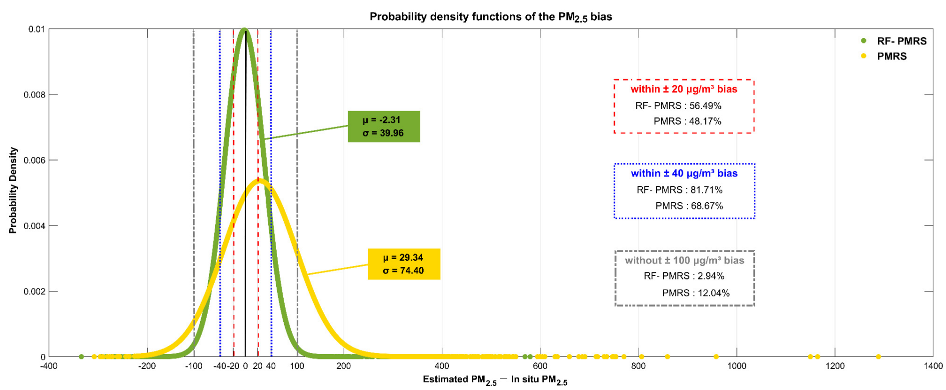

Fig. 8. Probability density functions of PMRS (yellow) and RF-PMRS (green) $PM_{2.5}$ bias. The red, blue and grey dotted lines indicate the bias boundaries of ±20 μg/m³, ±40 μg/m³, and ±100 μg/m³, respectively. μ and σ represent the mean value and standard deviation of each data.

4. Discussion

4.1. Accuracy comparison of PMRS using MODIS/Phy-DL FMF

To confirm the superiority of the Phy-FMF data adopted in our method framework, taking the BJ and BC sites as examples, the experiment compares the $PM_{2.5}$ accuracy

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and the number of effective days calculated by PMRS based on different FMF. Table 4 presents the overall day-level results. As can be seen, after the FMF replacement, the valid DOY turns out to become more (an increase of 113 days), which illustrates that the number of effective $PM_{2.5}$ concentration has gone up by about 5 times. Moreover, the accuracy has been significantly enhanced, with R increased by about 0.30, RMSE and MAE decreased by 26.14% and 16.47% accordingly. On the whole, Phy-DL FMF contributes to the improvement of PMRS results, signifying the first step optimization of the proposed RF-PMRS method is effective.

Table 4. Validation results of the PMRS method using different FMF data. The valid DOY refers to the number of days that the AOD, FMF, and other data are not missing when calculating $PM_{2.5}$. Note that since the valid days of the two schemes are different, the MB and RMB are not compared.

	Valid DOY	R	RMSE (μg/m³)	MAE (μg/m³)
PMRS with MODIS FMF	30	0.38	63.01	35.64
PMRS with Phy-DL FMF	143	0.68	46.54	29.77

4.2. Performance compared with other ML models

Different machine learning models are suitable for diverse research data, and decision tree (DT) models can better fit experiments with fewer variables, such as this study. For comparison, except for RF, the Extremely Randomized Tree (ERT) (Geurts et al., 2006) and Gradient Boosting Decision Tree (GBDT) (Friedman, 2001) models have also been established. The results of training VE_f based on the above three DT models are presented in Table 5 and Table 6. By contrast, RF performs best in CV and IV experiments, as indicated by the multiple accuracy indicators. Although ERT and GBDT models are comparable to RF in some indicators, there exists a certain degree of overfitting in the above two models, which is manifested in that their IV results are clearly worse than their respective CV ones. Thus, the RF model is applied to our study.

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Table 5. Cross-validation results in comparison of the decision tree models for training VE_f. N represents the number of data, and VE_f has no unit.

CV results					
	R	**RMSE**	**RPE**	**MAE**	**N**
RF	0.974	0.076	0.330	0.034	6463
ERT	0.972	0.079	0.343	0.035	
GBDT	0.973	0.078	0.339	0.036	

Table 6. Isolated-validation results in comparison of the decision tree models for training VE_f. The indicators are the same as those in Table 5.

IV results					
	R	**RMSE**	**RPE**	**MAE**	**N**
RF	0.975	0.067	0.299	0.037	814
ERT	0.967	0.076	0.340	0.042	
GBDT	0.969	0.074	0.331	0.040	

5. Conclusion

Among various satellite remote sensing methods for $PM_{2.5}$ retrieval, the semi-empirical physical approach has strong physical significance and clear calculation steps, and derives the $PM_{2.5}$ mass concentration independently of in situ observations. However, the parameters with the meaning of optical properties are difficult to express, which need to be optimized. Hence, the study proposes a method (RF-PMRS) that embeds machine learning in a physical model to obtain surface $PM_{2.5}$: 1) Based on the PMRS method and select the Phy-DL FMF product with a combined mechanism; 2) Use the RF model to fit the parameter VE_f, rather than a simple quadratic polynomial. In the point-to-surface validation, RF-PMRS shows great optimized performance. Experiments at two AERONET sites show that R reaches up to 0.8. And in North China, RMSE decreases by 39.95 μg/m³ with a 44.87% reduction in relative deviation. In the future, we will further explore the combination of atmospheric mechanism and machine learning, then research the $PM_{2.5}$ retrieval methods with physical meaning and higher accuracy.

Appendix A

A1. 10-fold cross-validation and isolated-validation

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The sample-based 10-fold cross-validation method is applied to test the fitting and predictive ability of our model. The original dataset is randomly divided into ten parts, nine of which are used as the training set for model fitting, and the remaining one is used for prediction, then the cross-validation process is repeated ten rounds until each data has been used as the test set.

At the same time, when verifying the RF-based VE_f model, the dataset in the time period that did not participate in the training in Table 1 is used for isolated-validation.

A2. Statistical indicators

$$R = \frac{\sum_{i=1}^{m}(y_i - \bar{y})\sum_{i=1}^{m}(f_i - \bar{f})}{\sqrt{\sum_{i=1}^{m}(y_i - \bar{y})^2}\sqrt{\sum_{i=1}^{m}(f_i - \bar{f})^2}}$$

$$MB = \bar{y} - \bar{f}$$

$$RMB = \mathrm{abs}\left(\frac{\bar{y} - \bar{f}}{\bar{y}}\right)$$

$$RMSE = \sqrt{\frac{1}{m}\sum_{i=1}^{m}(y_i - f_i)^2}$$

$$MAE = \frac{1}{m}\sum_{i=1}^{m}\left|y_i - f_i\right|$$

$$RPE = \frac{\sqrt{\frac{1}{m}\sum_{i=1}^{m}(y_i - f_i)^2}}{\bar{y}}$$

where m is the total number of observations, i is the number of measurements, y_i is the i-th observation, f_i is the corresponding estimation result. And $\bar{y}$ and $\bar{f}$ are the averages of all observations and estimates, respectively.

A3. Parameter adjustments of the RF model

The four parameters of RF are adjusted, that is the correlation coefficient r changes

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with (a) the number of trees, (b) maximum depth, (c) maximum number of features when splitting, (d) minimum number of split samples. Experiments shows that the maximum depth varies greatly in a small range. To prevent overfitting, the four parameters of RF are adjusted to 60, 10, 2, and 8. It can ensure high accuracy while improving training efficiency.

Code and data availability

All relevant codes as well as the intermediate data of this work are archived at https://doi.org/10.5281/zenodo.7183822 (Jin, 2022). The MCD19A2 data can be downloaded on https://ladsweb.modaps.eosdis.nasa.gov (last access: 30-09-2022) (Lyapustin and Wang, 2015). Detailed information about Phy-DL FMF dataset can be found at https://doi.org/10.5281/zenodo.5105617 (Yan, 2021). Meteorological data used in this work are obtained on https://cds.climate.copernicus.eu/cdsapp#!/dataset/reanalysis-era5-single-levels (lass access: 30-09-2022) (Hersbach et al., 2018). AERONET data was downloaded from https://aeronet.gsfc.nasa.gov/ (last access: 30-09-2022) (Giles et al., 2019).

Author contributions

Caiyi Jin: Data curation, Methodology, Formal analysis, Writing - original draft. **Qiangqiang Yuan:** Conceptualization, Supervision, Project administration, Writing - review and editing. **Tongwen Li:** Resources, Methodology, Writing - review and editing, Formal analysis. **Yuan Wang:** Methodology, Validation, Writing - review and editing. **Liangpei Zhang:** Supervision, Writing - review and editing.

Competing interests

The contact author has declared that none of the authors has any competing interests.

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