

Response to referees' comments:

Referee #1

The editor may forward the following comments to the authors, but it is not necessary to be published. Thanks. I still have a little concern about the language. A proof reading and language editing would be helpful to make the paper more readable. I list some examples below.

Response: First of all, thanks for the editor and reviewers for taking time to review our manuscript and offering helpful and constructive suggestions! We have carefully read the kind comments by all reviewers and revised the manuscript accordingly. Please see our detailed point-by-point reply below (in blue).

On the other hand, if the authors could comment or give some directions on how any interested reader can use their suggested method, it would be great.

Reply: Per your kind suggestion, we have revised our manuscript in an attempt to give direction on how to use our suggested method. Please refer to the revised manuscript for more details.

1. Lines 25-28, long sentence difficult to follow, consider to revise it as “On the other hand, these data gaps could introduce significant bias to daily averages of PM2.5 concentration, especially during clean episodes when larger biases would be introduced to PM2.5 daily averages compared to the polluted days (even with the presence of same number of missingness).”

Reply: Revised as suggested.

2. Line 28-31, consider to revise it as “The cross-validation results indicate that our suggested DCCEOF method, with the consideration of local diurnal variation pattern of PM2.5, has a good prediction accuracy particularly in predicting daily peaks and/or minima that cannot be restored by the conventional spline interpolation approach.”

Reply: Revised as suggested.

3. Line 45-46, grammar error.

Reply: Corrected.

Filling the gaps of in-situ hourly PM_{2.5} concentration data with the aid of empirical orthogonal function constrained by diurnal cycles

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Abstract. Data gaps in surface air quality measurements significantly impairs the data quality and the exploration of these valuable data sources. In this study, a novel yet practical method called diurnal cycle constrained empirical orthogonal function (DCCEOF) was developed to fill in data gaps present in data records with evident temporal variability. The hourly PM_{2.5} concentration data retrieved from the national ambient air quality monitoring network in China was used as a demonstration. The DCCEOF method aims at reconstructing the diurnal cycle of PM_{2.5} concentration from its discrete neighborhood field in space and time firstly and then predicting the missing values by calibrating the reconstructed diurnal cycle to the level of valid PM_{2.5} concentrations observed at adjacent times. The statistical results indicate a high frequency of data gaps in our retrieved hourly PM_{2.5} concentration record, with PM_{2.5} concentration measured on about 40% of days suffering from data gaps. Further sensitivity analysis results reveal that data gaps in hourly PM_{2.5} concentration record may introduce significant bias to their daily averages, especially during clean episodes at which PM_{2.5} daily averages are observed to subject to larger uncertainties compared to the polluted days (even in the presence of same number of missingness). The cross-validation results indicate that our suggested DCCEOF method has a good prediction accuracy, particularly in predicting daily peaks and/or minima that cannot be restored by conventional interpolation approaches, thus confirming the effectiveness of the consideration of local diurnal variation pattern in gap filling. By applying the DCCEOF method to the hourly PM_{2.5} concentration record measured in China during 2014 to 2019, the data completeness ratio was substantially improved while the frequency of days with gapped PM_{2.5} records reduced from 42.6% to 5.7%. In general, our suggested DCCEOF method provides a practical yet effective approach to handle data gaps in time series of geophysical parameters with significant diurnal variability, and this method is also transferable to other data sets with similar barriers because of its self-consistent capability.

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1 Introduction

A large variety of ground-based monitoring networks have been established worldwide to provide accurate measurements on various aspects of the atmospheric environment (Lolli and Di Girolamo, 2015).

Many of these in-situ measurements, however, suffer from data losses due to various unexpected reasons

(e.g., instrumental malfunction, interruption of power supply, and internet outage), thus resulting in salient data gaps in the archived data records. Undoubtedly, these gaps significantly impair the data qualities and the exploitation of these valuable data sources. Therefore, filling data gaps present in such datasets is essential to the further exploitation of these in-situ measurements.

Due to the high frequency and severity of haze pollution events, China started to establish the national ambient air quality monitoring network since 2012 by extending the range of the previous sparsely distributed monitoring network to cover most major Chinese cities. To date, more than 1,600 state-controlled monitoring stations are routinely operated to measure concentrations of six primary air pollutants (i.e., PM₁₀, PM_{2.5}, O₃, NO₂, SO₂, CO) on an hourly basis (Guo et al., 2017; Li et al., 2017a).

These in-situ measurements are publicly released online via the China National Environment Monitoring Centre (CNEMC) in near real-time as of 2013 but without providing any direct data download interface.

Consequently, users oftentimes apply an automated software program (also known as a “web crawler”) to retrieve these valuable data sources from the CNEMC website. Such an endeavour empowers us to acquire hourly air quality data more efficiently.

As a critical air quality indicator, PM_{2.5} mass concentration data have been widely used in many haze related studies (Gao et al., 2018; Miao et al., 2018; Bai et al., 2019a, 2019b; Zhang et al., 2019).

Nevertheless, how data gaps were treated in the data exploration process (e.g., data integration and data transformation), especially for those using daily or monthly averaged PM_{2.5} data set (e.g., Guo et al., 2009; Miao et al., 2018; Ye et al., 2018; Zhang et al., 2018; Yang et al., 2019a), is oftentimes unclear. Since ignoring missing values would undoubtedly introduce biases into the final results (Bondon, 2005; Larose et al., 2019), some studies attempted to perform data analysis on a relatively long time scale by integrating hourly records into monthly resolution so as to mitigate the impacts of data gaps (e.g., Bai et al., 2019b; Zhang et al., 2019). On the other hand, many previous studies preferred to exclude records on days subject to a certain degree of missing values (e.g., no more than 6 missing values within 24-h) from their analysis

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(e.g., van Donkelaar et al., 2016; Li et al., 2017; Huang et al., 2018; Manning et al., 2018; Shen et al., 2018; Bai et al., 2019a; Zhang et al., 2019). Such a treatment on data gaps (e.g., ignoring missing values or excluding records on days with missingness) would either introduce new bias to the aggregated data record or make the original PM_{2.5} time series temporally discontinuous, however.

Since a non-gap PM_{2.5} record is essential to PM_{2.5} related haze control and environmental health risk assessment, filling data gaps in hourly PM_{2.5} concentration record is thus of great value. Although there exists versatile gap filling methods in literature (e.g., Beckers and Rixen, 2003; Taylor et al., 2013; Chang et al., 2015; Dray and Josse, 2015; Gerber et al., 2018), most of them fail to properly restore missingness present in data record with high temporal resolution (e.g., hourly) and evident diurnal variability (e.g., PM_{2.5} concentration), in particular the daily extrema. For instance, PM_{2.5} concentrations vary significantly in space and time due to heterogeneous local emissions and atmospheric conditions (Guo et al., 2017; Lennartson et al., 2018; Shi et al., 2018). A similar barrier also applies for many other datasets which are sampled at high temporal resolution. Therefore, a priori knowledge of the diurnal variation pattern of the analysed data is thus essential to the restoration of values for missingness.

In this study, a novel yet practical gap filling method called DCCEOF (that is, the diurnal cycle constrained empirical orthogonal function) was developed to better handle data gaps present in time series with marked variability in space and time, by taking the diurnal variation pattern as a critical constraint in missing value prediction. To our knowledge, none of the existing gap filling methods have accounted for the diurnal variation pattern of the given data in their missing value restoration schemes, and hence the predicted values from such methods would subject to large bias. As an illustration, the hourly PM_{2.5} concentration record retrieved from CNEMC during the time period of 2014 to 2019 was applied to demonstrate the efficacy and accuracy of the suggested DCCEOF method. Science questions to be answered by this study include: (1) how about the data completeness of the Chinese in situ PM_{2.5} record? (2) how much uncertainties can be introduced to PM_{2.5} daily averages by missing values? (3) is it feasible to reconstruct the local diurnal variation pattern of PM_{2.5} from discrete observations in the neighborhood? and (4) are missing values predicted by DCCEOF reliable?

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2 Overview of existing gap filling methods

285 Plenty of methods have been developed or adopted for gap filling with respect to various theoretical bases, ranging from simple replacement with surrogates (e.g., mean value) to spatiotemporal interpolation as well as complicated machine learning techniques. Generally, these methods can be classified into different groups according to different criteria. For instance, two major groups can be classified based on the number of variables (univariate versus multivariate) (Ottosen and Kumar, 2019) and theoretical basis
290 (likelihood-based versus imputation-based) (Junger and Ponce de Leon, 2015). Table 1 summarizes a selection of popular gap filling methods to deal with missingness in geophysical data sets according to the domain specific data dependence (Gerber et al., 2018). Comparisons of the performance of these methods can also be found in other literatures, e.g., Kandasamy et al. (2013), Demirhan and Renwick (2018), Yadav and Roychoudhury (2018), and Julien and Sobrino (2019), to name a few.

295 Since each method is initially proposed to deal with missingness in one specific data set, adopting one method to another data set is often a challenge due to distinct features of missingness (e.g., missing at random versus missing not at random), in particular for data sets with significant spatiotemporal heterogeneity such as air pollutants time series (Junger and Ponce de Leon, 2015). PM_{2.5} concentration often exhibits evident diurnal variation patterns, which are primarily governed by local air pollutants
300 emissions and regional meteorological conditions such as boundary layer height (Guo et al., 2017; Li et al., 2017; Huang et al., 2018; Liu et al., 2018; Miao et al., 2018; Yang et al., 2018, 2019b). Consequently, conventional approaches like those listed in Table 1 may partially fail in accurately predicting missing values in hourly PM_{2.5} time series.

In general, most available gap filling methods in Table 1 suffer from at least one of the following
305 drawbacks: 1) partially fail for data sets with prominent gaps; 2) not self-consistent due to the requirement of supplementary data sets; 3) computationally intensive (e.g., neural networks), and, most critically; 4) unable to fairly predict daily peaks and/or minima due to the lack of essential prior knowledge of diurnal variation cycle of monitoring targets. Given the significant heterogeneity of PM_{2.5} concentration in space and time (Guo et al., 2017; Manning et al., 2018), ignoring the diurnal phases of PM_{2.5} would result in
310 large bias to the gap filled PM_{2.5} data set.

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Table 1. Overview of several popular gap filling methods to impute missingness in geophysical data sets.

	Method	Principle or core technique	Reference
Temporal	Weibull	Weibull frequency distribution mapping	Nosal et al. (2000)
	EM	Expectation-Maximization	Junger and Ponce de Leon (2015)
	Interpolation	Linear regression, Spline, NAR, ARIMA, ARCH	Stauch and Jarvis (2006); Neteler (2010); Demirhan and Renwick (2018)
	Machine learning	Gradient Boosting, neural networks	Körner et al. (2018); Şahin et al. (2011)
	SSA	Imputation using singular spectrum analysis	Mahmoudvand and Rodrigues (2016)
	DS	Conditional resampling of a temporal subset	Dembélé et al. (2019); Oriani et al. (2016)
	TIMESAT	Savitzky–Golay filter, harmonic and asymmetric Gaussian functions	Jönsson and Eklundh (2004)
	Hybrid method	Fuzzy c-means with support vector regression and genetic algorithm	Aydilek and Arslan (2013)
Spatial	IDW	Interpolate using inverse distance weighting	Shareef et al. (2016)
	Kriging	Interpolate neighborhoods using Kriging	Rossi et al. (1994); Zhu et al. (2015); Singh et al. (2017)
	NSPI / GNSPI	Replace or interpolate with adjacent similar pixels	Zhu et al. (2012); Chen et al. (2011)
Spatio-temporal	EOF / DINEOF	Iteratively decompose and reconstruct spatial and temporal subsets using empirical orthogonal function	Beckers and Rixen (2003); Taylor et al. (2013); Liu and Wang (2019)
	Mosaicing	Merge numerical outputs with satellite observations	Konik et al. (2019)
	gapfill	Quantile regression fitted to spatiotemporal subsets	Gerber et al. (2018)
	STWR	Spatially and temporally weighted regression	Chen et al. (2017)
	SMIR	Learning machine created from historical spatial and temporal subsets	Chang et al. (2015)
	RFRE	Learning from other information using random forest	Bi et al. (2018); Chen et al. (2019)

* SSA: Singular Spectrum Analysis; DS: Direct Sampling; IDW: Inverse Distance Weighting; NSPI: Neighborhood Similar Pixel Interpolator; GNSPI: Geo-statistical Neighborhood Similar Pixel Interpolator; EOF: Empirical Orthogonal Function; DINEOF: Data Interpolating Empirical Orthogonal Function; STWR: Spatially and Temporally Weighted Regression; SMIR: SMart Information Reconstruction; RFRE: Random Forest Regression

3 The DCCEOF gap filling method

Given the significant heterogeneity of PM_{2.5} diurnal variation pattern associated with local emissions of air pollutants and atmospheric conditions, we propose to **incorporate** the local diurnal variation pattern of PM_{2.5} to constrain the **prediction** of missing values in **each** hourly PM_{2.5} concentration **record**. The goal

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is to better predict missing $PM_{2.5}$ values, especially for the daily peaks and/or minima, which are poorly predicted by conventional methods due to the absence of prior knowledge of local diurnal phases of $PM_{2.5}$.

Figure 1 presents a schematic illustration of the proposed DCCEOF method, which consists of the following four primary procedures toward the filling of data gaps present in each 24-h $PM_{2.5}$ time series:

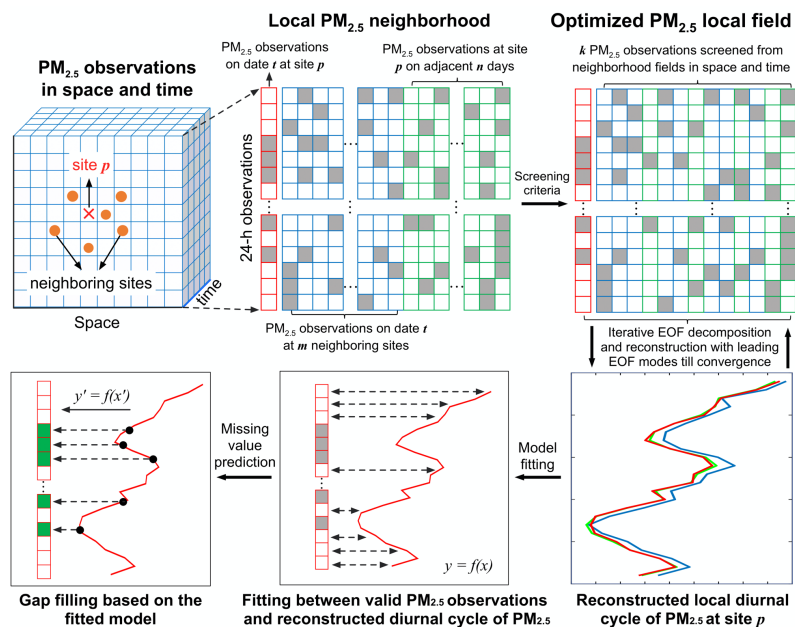
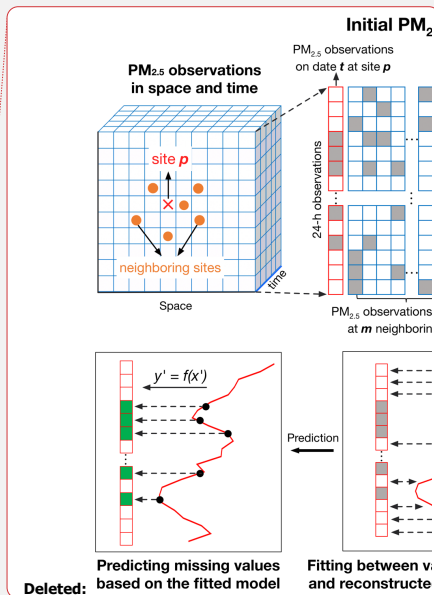


Figure 1. A schematic illustration of the suggested DCCEOF method to fill data gaps in hourly $PM_{2.5}$ concentration records. The grey rectangles denote missing values while the green ones indicate restored data values.

1) Initialize a local $PM_{2.5}$ neighborhood field: For a $PM_{2.5}$ missingness at site p on date t , an initial $PM_{2.5}$ neighborhood field in space and time (denoted as $X_{p,t}^{m,n}$) was first constructed using 24-h $PM_{2.5}$

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observations from nearby m stations on date t and adjacent $2n$ days (n days before and after t respectively) at site p . Mathematically, the neighborhood field $X_{p,t}^{m,n}$ can be expressed as:

$$X_{p,t}^{m,n} = \{x_t^1, x_t^2, \dots, x_t^m, x_p^{t-n}, \dots, x_p^{t-2}, x_p^{t-1}, x_p^{t+1}, x_p^{t+2}, \dots, x_p^{t+n}\} \quad (1)$$

350 It is clear that m and n are two critical factors ~~modulating~~ the dimension of $X_{p,t}^{m,n}$. Considering a too compact neighborhood field may be inadequate to reconstruct the local diurnal cycle of $PM_{2.5}$ fairly due to limited ~~valid~~ samples, (because missingness may also ~~emerge~~ in each candidate 24-h $PM_{2.5}$ concentration ~~record~~), here m ~~was~~ defined as the number of stations within 100 km (spatial window size) to the target station while n ~~was~~ set to 7 (temporal window size) ~~in our case~~. The spatial and temporal
355 window sizes used here ~~are~~ based on our recent results in which an optimal window size of 50 km and 3-day was found to attain a good autocorrelation of $PM_{2.5}$ concentration in space and time, respectively (Bai et al., 2019c). To have adequate samples for the construction of $X_{p,t}^{m,n}$, here we enlarged the both window sizes by simply doubling the ~~values~~ found in our previous study. ~~These~~ two window sizes would have little effect on the performance of the subsequent gap filling once they are large enough (at least greater
360 than the identified optimal window sizes) to cover most similar observations nearby. ~~This is because a~~ sorting scheme ~~is~~ further applied to ~~the neighborhood field to pinpoint~~ observations ~~having~~ similar diurnal variation pattern ~~with~~ the target station. In other words, the two window sizes used here is simply to include adequate samples while avoiding incorporating all available data for the subsequent data reconstruction, ~~in particular~~ those ~~even~~ distant away.

365 2) Construct a compact $PM_{2.5}$ neighborhood field: Since the initial $PM_{2.5}$ neighborhood field $X_{p,t}^{m,n}$ might include many irrelevant observations with distinct diurnal variation patterns ~~due to~~ large spatial and temporal window sizes, a compact neighborhood field needs to be constructed by only retaining observations that are highly related to the target $PM_{2.5}$ time series x_p^t with respect to the diurnal variation pattern. Therefore, the covariance rather than correlation between the target time series x_p^t and every
370 candidate $PM_{2.5}$ time series in $X_{p,t}^{m,n}$ ~~was~~ first calculated (weighted by the number of valid data pairs within 24-h). Subsequently, the candidate $PM_{2.5}$ time series ~~were~~ sorted in terms of the magnitudes of covariances in a descending order. Finally, the first k time series ~~were~~ retained to construct the optimized $PM_{2.5}$ neighborhood field X^k by complying with the criterion that there ~~were~~ at least five valid

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observations at each specific time from 00:00 to 23:00. The aim ~~was~~ to avoid large bias in the subsequent diurnal cycle reconstruction using empirical orthogonal function (EOF), since large outliers ~~might~~ emerge at times without any valid observation. Mathematically, the process to construct \mathbf{X}^k can be formulated as follows:

$$C_{x'} = COV(x_p^t, x' | X_{p,t}^{m,n}) \quad (2)$$

$$\mathbf{X}^k = \{x'_1, x'_2, \dots, x'_k | C_{x'_k} < C_{x'_{k-1}} < \dots < C_{x'_1}\} \quad (3)$$

where x' denotes the 24-h time series of candidate PM_{2.5} in $X_{p,t}^{m,n}$ and COV is the covariance function.

3) Reconstruct the local diurnal cycle of PM_{2.5}: The diurnal cycle of PM_{2.5} at site p on date t (denoted as β_p^t) was then reconstructed from the optimized PM_{2.5} neighborhood field \mathbf{X}^k using EOF in an iterative process similar to the DINEOF method (Beckers and Rixen, 2003). In our ~~suggested~~ DCCEOF method, the target PM_{2.5} time series at site p on date t (denoted as x_p^t) were also included to constrain the reconstruction of β_p^t , and the whole field ~~can be~~ denoted as \mathbf{X} :

$$\mathbf{X} = \{x_p^t, \mathbf{X}^k\} \quad (4)$$

In general, the EOF-based gap filling process can be outlined as follows: a) 20% of valid PM_{2.5} observations in \mathbf{X} were first held out for cross validation and then these data values were treated as gaps by replacing with nulls (i.e., missing value); b) given that a small amount of missing values would not significantly influence the leading EOF mode for the original data set, we ~~assigned~~ a first guess (here we used the mean value of valid data on each specific date) to the data points where missing values ~~were~~ identified to initialize the EOF analysis; c) EOF analysis was performed on the previously generated background field (that is, \mathbf{X} with gaps are filled with daily mean and denoted as $\langle \mathbf{X} \rangle$) in a form of singular value decomposition (SVD) and then data values at value-missing points were replaced by the reconstructed values using the first EOF mode at the corresponding locations. These processes can be expressed as:

$$[U, S, V] = \text{SVD}(\langle \mathbf{X} \rangle) \quad (5)$$

$$\mathbf{X}' = u_1 * s_1 * v_1 \quad (6)$$

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where $\langle X \rangle$ denotes the initial matrix in which the missing values were filled with daily means. U , S , and V are three matrices derived from SVD while u_1 , s_1 , and v_1 denote the SVD components in the first EOF mode. X' is the reconstructed matrix using the first EOF mode; e) iteratively decompose and reconstruct the matrix while updating data values at the value-missing points using the first EOF mode till the convergence **was** confirmed by the mean square error at each iteration; f) repeat the above iterative processes **by including** the following EOF modes till the reach of the final convergence (i.e., **mean square error started** to increase **when a** new EOF mode **was** included). The **diurnal cycle β_p^t** was finally derived by standardizing the identified leading EOF modes **in the last round iteration**.

4) **Prediction of missing values**: A linear relationship was **then** established between valid PM_{2.5} observations in **the original time series x_p^t** and the corresponding values in **β_p^t** . Missing values in **x_p^t** were then predicted by mapping data values in the reconstructed diurnal cycle **β_p^t** at missing times to the level of valid PM_{2.5} observations based on the established linear relationship.

In short, **our suggested** DCCEOF method is a univariate and self-consistent gap filling method since no additional data record is required for **the restoration of** missing values. Rather, the method works relying primarily on the local diurnal cycle of PM_{2.5} that can be reconstructed from discrete PM_{2.5} neighborhood fields in space and time. In contrast to conventional gap filling methods that work on a purely statistical basis (e.g., spline interpolation), the unique feature and novelty of the proposed DCCEOF method lies in the accounting for the **local** diurnal variation pattern **of the input data** in their missing value **predictions**, **thus** making the predicted values **physically meaningful and** with high accuracy.

450 4 Demonstrative case study in China

4.1 China in-situ PM_{2.5} concentration records

The near surface mass concentration of PM_{2.5} across China are measured primarily using the tapered element oscillating microbalance analyzer and/or the beta-attenuation monitor at each monitoring station. The instruments' calibration, operation, maintenance, and quality control are all properly conducted by complying with the China Environmental Protection Standards of GB3095-2012 and HJ 618–2011. PM_{2.5} concentration data are measured by these instruments with an accuracy of $\pm 5 \mu\text{g}/\text{m}^3$ for

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ten-minute averages and $\pm 1.5 \mu\text{g}/\text{m}^3$ for hourly averages (Guo et al., 2017; Miao et al., 2018). Although the hourly $\text{PM}_{2.5}$ observations in China have been publicly available since 2013, the $\text{PM}_{2.5}$ records used in the present study were retrieved since May 2014 via a web crawler program.

Figure 2 depicts the spatial distribution of monitors in the national ambient air quality monitoring network in China as well as the year starting to release of $\text{PM}_{2.5}$ measurements to the public at each individual station. Given the fact that our data were retrieved following May 2014, stations deployed before that are hardly to be separated from those being built in 2014 and hence, they were all designated the same way in Figure 2. At present, this network consists of more than 1,600 stations, in which about 940 stations were established before 2015. The total number was increased to 1,494 in June 2015, and then only four stations were newly deployed in the following one and half years till December 2016. In other words, the vast majority (92.4%) of stations in the current network was deployed before the middle of 2015.

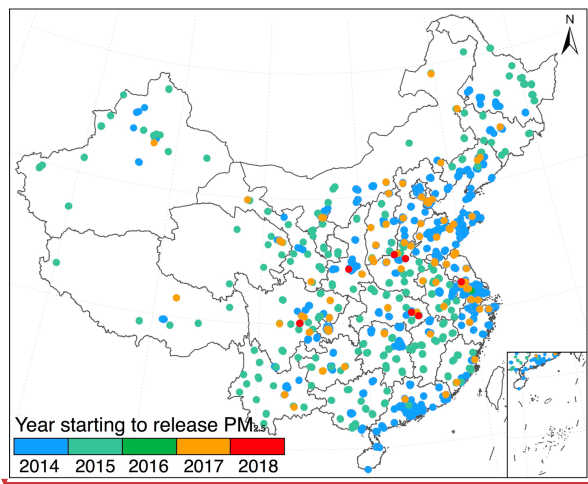


Figure 2. Spatial distribution of national ambient air quality monitoring network stations in China during May 2014 to April 2019. Circles with distinct color indicate the year in which the first $\text{PM}_{2.5}$ observation was publicly released in our retrieved data record.

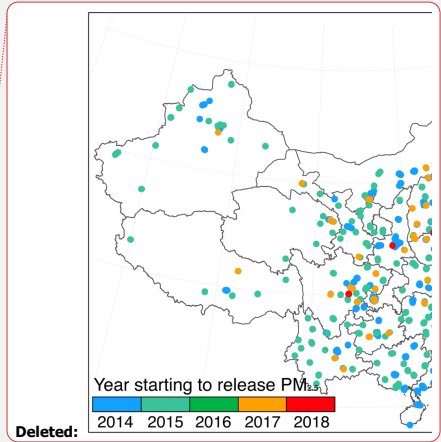
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4.2 Results

4.2.1 Data incompleteness of in-situ PM_{2.5} records in China

Figures 3a–c present the daily averaged missing value ratio, the occurrence frequency of missingness (defined as the ratio of days with missing values in each 24-hour PM_{2.5} observations divided by the total number of days), and the diurnal phases of the most frequently occurring missing values at each monitoring station since the first release of PM_{2.5} observations to the public, while Figures 3d–f show the corresponding histograms, respectively. Although most of stations have a daily-averaged missing value ratio less than 10% (Figures 3a and 3d), significant data gaps are still observed at several monitoring stations (red dots in Figure 3a) with more than 70% of hourly PM_{2.5} observations lost in daily 24-h measurements. After checking the retrieved PM_{2.5} data records over these stations, we find that most of these stations stopped releasing PM_{2.5} observations after the middle of 2015.

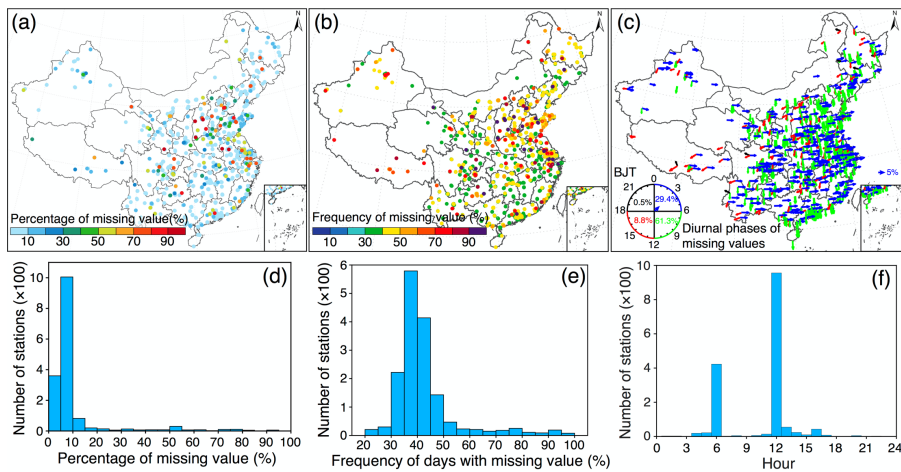
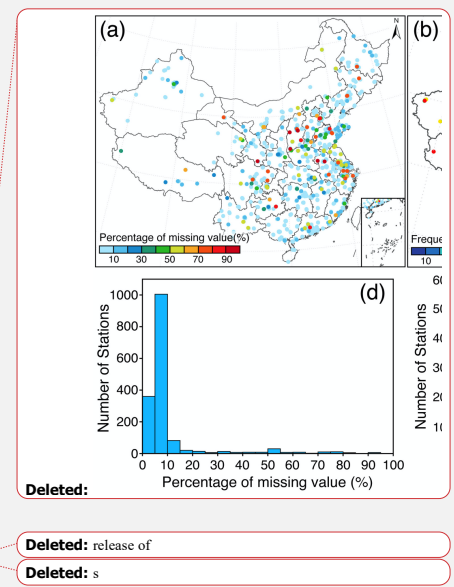


Figure 3. Statistics associated with missing values present in site specific hourly PM_{2.5} concentration record since the time for the release of first PM_{2.5} observation onward. (a) Percentage of missingness in each PM_{2.5} record, (b) frequency of days with missing values, (c) diurnal phases of the maximum frequency of missing values occurred within 24-h, (d–f) histograms for (a–c), respectively. The arrow



515 direction in (c) indicates the local time (Beijing time, BJT) at which missing values occurred most frequently and the arrow length shows the magnitude of frequency. The varying diurnal phases of missing values were represented by different color: blue (00~06 BJT), green (06~12 BJT), red (12~18 BJT), and black (18~24 BJT).

520 Despite the small magnitudes (~10%) of daily-averaged missing value ratios (Figure 3d), data gaps in our retrieved hourly PM_{2.5} record are still significant, which is evidenced by the occurrence frequency of missing values in daily PM_{2.5} observations (Figure 3b). In contrast to the daily averaged missing value ratios (Figure 3a), the frequency of days with missing values has a relatively larger magnitude of about 40% (PM_{2.5} data measured on four out of ten days suffered from missingness), indicating an extraordinary
525 high chance to suffer from data gaps in the retrieved PM_{2.5} record (Figure 3e). These results suggest an urgent need to fill data gaps in our retrieved PM_{2.5} concentration record so as to facilitate the further exploration of this valuable data set.

Figure 3c presents the diurnal variation pattern of the occurrence of missingness in the retrieved PM_{2.5} record in terms of the detailed time (represented by the arrow direction) and frequency (represented by the relative length of each arrow) of the most commonly occurring missing values, while Figure 3f shows the histogram of the local time at which missing values occurred most frequently at each monitoring station. It is note-worthy that missing values occurred more frequently in the morning over most stations (90.7% of total population of stations), particularly at 0600 and 1200 of the Beijing time. Nevertheless,
530 detailed reason for this diurnal variation pattern remains unclear.

535 **4.2.2 Impacts of data gaps on PM_{2.5} daily averages**

Given the common application of daily-averaged PM_{2.5} concentration data in many studies, the possible impacts of data gaps on PM_{2.5} daily averages were thus assessed here to examine how well the estimated PM_{2.5} daily averages can be trusted in the presence of data gaps, especially during different pollution episodes. Toward such a goal, gap-free observations of hourly PM_{2.5} concentration within 24-h were first
540 extracted. To make the computational workload manageable, we randomly sampled 1,000 days observations rather than using observations from all gap-free days. Moreover, days with PM_{2.5} daily

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averages lower than that of the 10th percentile of all gap-free days were considered as clean scenario, while those greater than the 90th percentile were treated as polluted scenario. Subsequently, a varying number (ranging from 1 to 23) of data values were held out and then treated as gaps in every 24-h PM_{2.5} observations randomly. Mean relative differences (MRDs) between PM_{2.5} daily averages derived from hourly records with and without data gaps were finally calculated to examine the potential impacts of missingness on PM_{2.5} daily averages.

Figure 4a shows the estimated MRDs at the 10th, 50th, and 90th percentiles associated with different numbers of missing values in each 24-h PM_{2.5} observations. There is no doubt that larger biases could be introduced to PM_{2.5} daily averages with the increase in the total number of missingness. Given the symmetrical behavior of MRDs around zero (50th percentile) for each given number of missingness, we may infer that random biases could be introduced to PM_{2.5} daily averages if missing values are ignored for the calculation of daily averages of PM_{2.5}. These random biases, in turn, could result in large uncertainties to the subsequent results such as trend estimations. To further evaluate the impacts of missingness on PM_{2.5} daily averages, in particular at different pollution scenarios, MRDs were also calculated on 1,000 clean and polluted days, respectively (Figures 4b–d). On average, MRDs vary with larger deviations on clean days than polluted days (Figure 4b). Regarding MRDs at 10th and 90th percentiles, we may deduce that missing values would result in larger bias to PM_{2.5} daily averages on clean days than in polluted conditions given larger MRDs during clean scenarios (Figures 4c–d). This effect is in line with expectations since PM_{2.5} concentration often exhibits relatively larger diurnal variations on cleaner days than during polluted episodes due to the possible boundary layer height effect (Li et al., 2017; Miao et al., 2018). Moreover, six missing values in 24-h observations would result in as large as approximately 5% of deviations (10% for 12 missing values) to PM_{2.5} daily averages during clean days (Figures 4c–d).

In addition to the total number of missing values, possible impacts of diurnal phases of missing values on PM_{2.5} daily averages were also examined. It shows that different diurnal phases were observed for MRDs associated with missingness at different pollution levels (Figure 5). Specifically, missing values in the afternoon and evening would be more likely to overestimate PM_{2.5} daily averages, whereas an opposite effect (underestimations) was observed for missingness in the morning and night. Moreover, the

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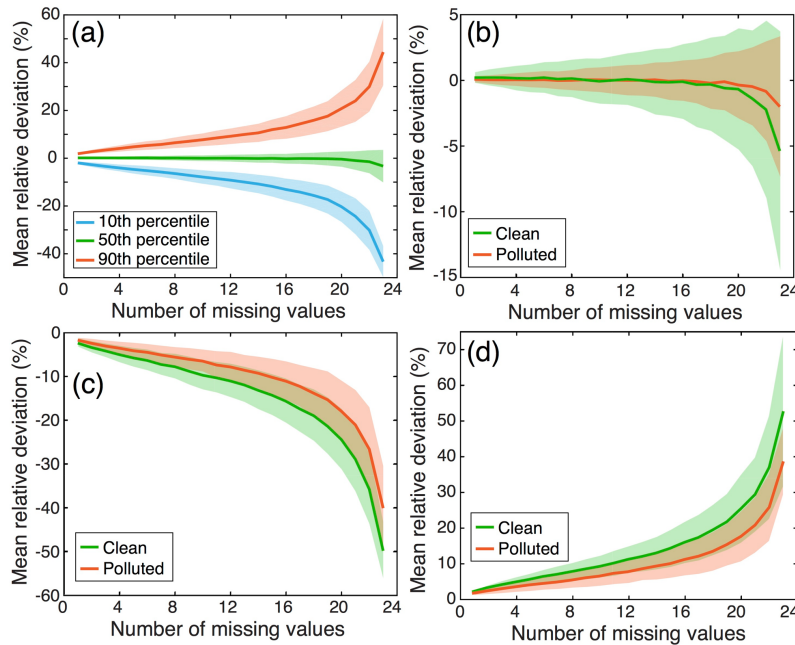
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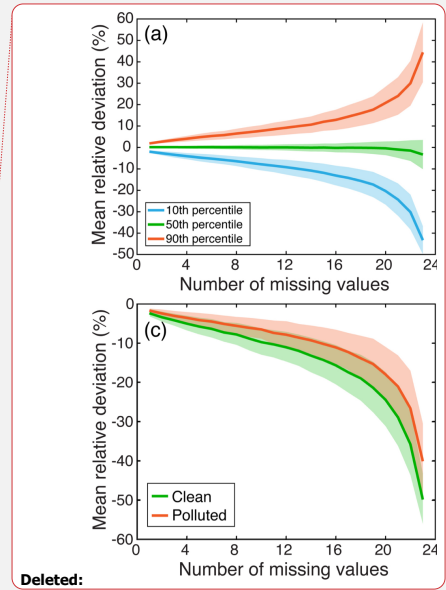
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missingness in the afternoon during clean days has a larger potential to overestimate $PM_{2.5}$ daily averages than during other times. This effect could be largely associated with the diurnal phases of $PM_{2.5}$ as daily peaks are oftentimes observed in the early morning (Wang and Christopher, 2003), though such a diurnal variation pattern may differ by regions (Lennartson et al., 2018). Also, the diurnal phases of $PM_{2.5}$ are largely dominated by the diurnal variation of regional emissions and boundary layer processes (Guo et al., 2016; Lennartson et al., 2018; Miao et al., 2018; Yang et al., 2019b). In contrast, the diurnal phases of MRDs are not evident during polluted days. Above findings collectively suggest the need to fill data gaps in hourly $PM_{2.5}$ observations, especially for those measured during clean days, since missing values would result in larger biases to $PM_{2.5}$ daily averages than those during polluted episodes.



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Figure 4. Impacts of the number of missing values on daily averages of $PM_{2.5}$. Mean relative deviations were calculated between $PM_{2.5}$ daily averages estimated from 1,000 hourly $PM_{2.5}$ records with a given

number of missing values and the original one without missing values. (a) Deviations at different percentiles at all-sky conditions; (b) deviations at the 50th percentile under different pollution scenarios; (c) same as (b) but for the 10th percentile; (d) same as (b) but for the 90th percentile. Thick lines represent mean deviations while shaded regions are uncertainties of one standard deviation from the mean.

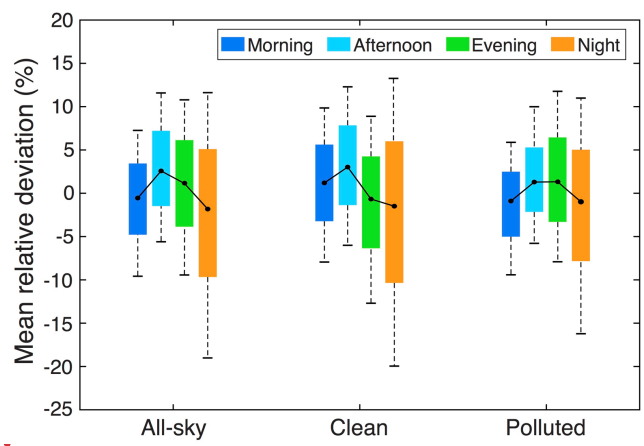
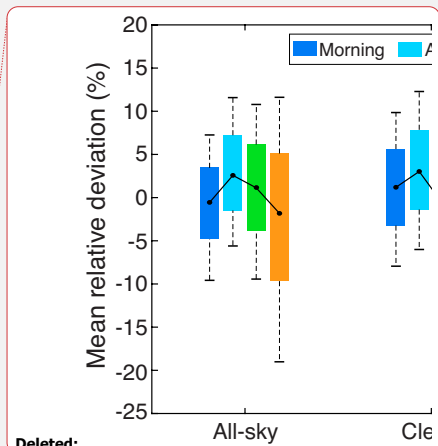


Figure 5. Impacts of diurnal phases of missing values on $PM_{2.5}$ daily averages. Hourly $PM_{2.5}$ values in the morning (07~11 BJT), afternoon (12~16 BJT), evening (17~21 BJT), and night (22~06 BJT) were removed from the original hourly $PM_{2.5}$ time series throughout the day to resemble missing values respectively. On each box, the black dots represent medians of mean relative deviations while the bottom and top edges of the box indicate the 25th and 75th percentiles and the whiskers extend to the 10th and 90th percentiles, respectively.

4.2.3 Performance of the DCCEOF method

Cross-validation experiments were first conducted at two monitoring stations to evaluate the efficacy of the suggested DCCEOF method in reconstructing the diurnal cycle of $PM_{2.5}$ from the discrete neighborhood field. Specifically, gap-free $PM_{2.5}$ records on three distinct days with different pollution levels were first extract randomly at each station and then six valid observations in each 24-h record were

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held out. Subsequently, the DCCEOF method was applied to reconstruct the diurnal cycle of $PM_{2.5}$ for each specific case. Figure 6 compares the reconstructed diurnal cycles of $PM_{2.5}$ with their actual $PM_{2.5}$ concentrations. The results indicate that the reconstructed diurnal cycles of $PM_{2.5}$ have a good fit with their actual observations, thus confirming the **effectiveness** of the DCCEOF method in reconstructing the diurnal variation pattern of $PM_{2.5}$ from the discrete neighborhood field. In particular, the DCCEOF method also succeeded to restore the missing $PM_{2.5}$ information even at the inflection times, e.g., the peak value in Figure 6c and the minimum value in Figure 6e, which are **hardly** to be recovered by statistical interpolation approaches. Nonetheless, compared with actual $PM_{2.5}$ observations, the reconstructed diurnal cycle of $PM_{2.5}$ is still unable to totally restore all types of local variations (e.g., $PM_{2.5}$ observations between 0700 and 1100 shown in Figure 6f). This is consistent with our initial understanding that $PM_{2.5}$ concentrations vary **substantially** in space and time whereas the reconstructed diurnal cycle of $PM_{2.5}$ is derived from a limited number of leading EOF modes and hence it only captures the dominant variation pattern of $PM_{2.5}$ in the neighborhood field while some local variations could be **thus** ignored. In spite of this potential defect, **the suggested** DCCEOF method still exhibits promising accuracy in restoring the local diurnal cycle of $PM_{2.5}$ even from a discrete neighborhood field.

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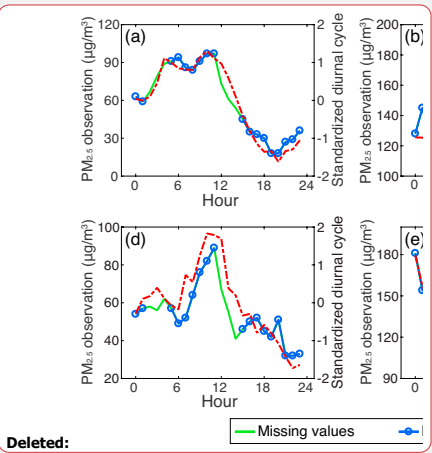
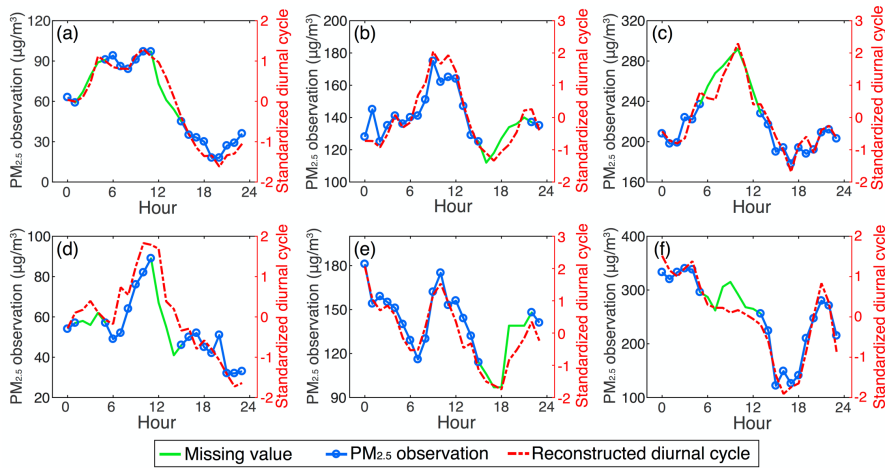
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675 **Figure 6.** Comparisons of reconstructed diurnal cycles of PM_{2.5} with their actual concentrations at distinct pollution levels. For each trial, six valid PM_{2.5} observations were held out to simulate gapped PM_{2.5} time series prior to the diurnal cycle reconstruction for a given day. Note that the number of neighboring stations differs between these two cases (58 for the top panel and 16 for the bottom).

To better assess the performance of the DCCEOF method, we retrieved the hourly PM_{2.5} observations at one monitoring station in Beijing during the time period of August 1 to 7, 2014 and then some valid observations were held out and then treated as missing values for the subsequent gap filling practices. Both the DCCEOF method and a spline interpolation approach were used to practically restore the retained PM_{2.5} observations. The comparison results shown in Figure 7 indicate higher accuracy of the DCCEOF method than the spline interpolation approach in restoring those retained PM_{2.5} observations, especially for those at the inflection times at which spline interpolation failed to predict with good accuracy (e.g., peak values on August 3). However, both methods failed in predicting the minimum values on August 2. After checking the original data record, we found that the local variation of PM_{2.5} at this station differed largely from all neighboring stations at that time. For such situation, the proposed DCCEOF method also fails to properly predict the missing values given large difference in diurnal variation patterns in space and time.

685 Figure 8 presents a more general evaluation of the prediction accuracy of the suggested DCCEOF method, which compares the predicted values with the retained observations at distinct pollution levels. As indicated, there is a good fit between the predicted values and the actual observations, with a correlation coefficient of 0.82 on clean days (Figure 8a) and 0.95 during polluted episodes (Figure 8b), respectively. This is in line with our expectation as higher prediction accuracy would be reached by the DCCEOF method in filling data gaps on polluted days given smaller variability of PM_{2.5} concentrations. This effect can also be evidenced by spread scatters shown in Figure 8a, which in turn reveals the large spatiotemporal
690 heterogeneity of PM_{2.5} concentrations during clean scenarios.

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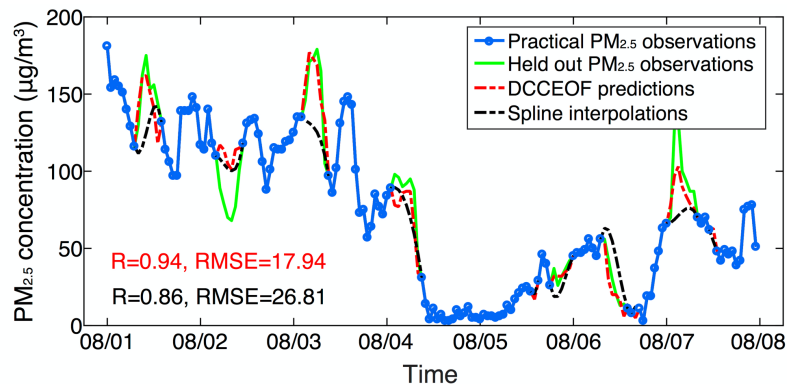


Figure 7. Comparison of predicted hourly $PM_{2.5}$ concentration values between the suggested DCCEOF method and spline interpolation at the Wanshou Temple station in Beijing during the period of August 1 to 7, 2014. The green line shows the practical $PM_{2.5}$ observations that were held out to simulate data gaps while their original values were retained for cross validation.

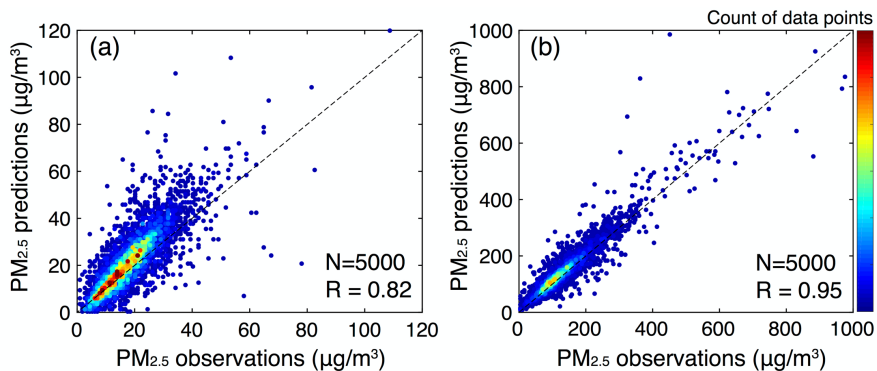
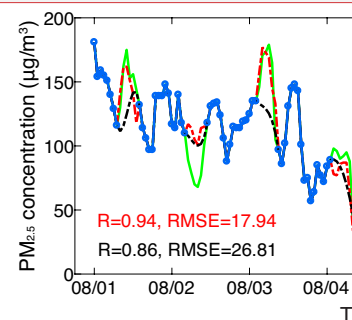


Figure 8. Comparisons of $PM_{2.5}$ observations with the reconstructed data values during clean (a) and polluted (b) phases. For each scenario, the results were derived from 1,000 days of gap-free $PM_{2.5}$



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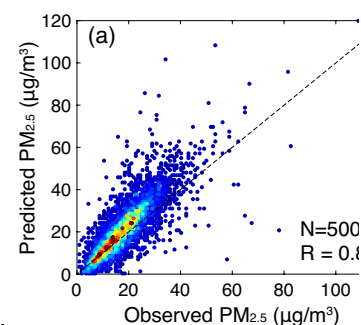
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760 observations with 5 valid values being randomly retained from 24-h observations on each sampled date
for cross validation.

Given the ~~inherent~~ principle of utilizing discrete neighborhood field in space and time to reconstruct the
local diurnal cycle of PM_{2.5} for the subsequent missing value ~~prediction~~, the performance of the DCCEOF
765 method could be ~~impacted by~~ the number of missing values and the total number of neighboring stations.
To ~~examine~~ the possible dependence of prediction accuracy on these two factors, sensitivity experiments
were also conducted. Figure 9a shows the response of prediction accuracy (in terms of correlation
coefficient) of the DCCEOF method to the varying number of missing values in each sampled 24-h PM_{2.5}
time series. It clearly shows that the prediction accuracy generally decreases with the increase in the
770 number of missing values. This effect can be ascribed to the fact that the target PM_{2.5} time series is also
applied as a critical constraint for the screening of similar PM_{2.5} observations in space and time to
construct the neighborhood field for the reconstruction of local diurnal cycle of PM_{2.5}. ~~As a consequence~~,
more missingness would make the constructed neighborhood field ~~be prone to~~ larger uncertainties due to
less information for the selection of ~~relevant~~ time series of PM_{2.5}, which in turn undermines the overall
775 accuracy of the final predictions.

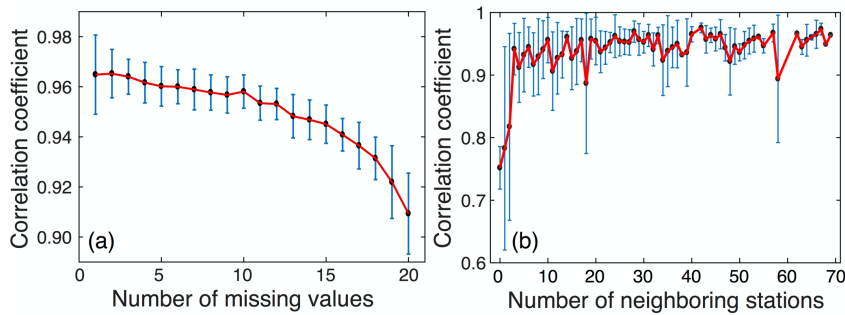


Figure 9. Impacts of the number of missing values present in hourly PM_{2.5} records for every 24-h (a) and the total number of neighboring stations within 100 km (b) on the performance of the proposed gap filling method. The error bars denote one standard deviation of each value from the mean on each side.

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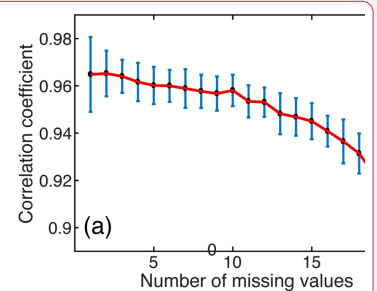
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790 Figure 9b shows the influence of the total number of neighboring stations on the prediction accuracy at the target station. The total number of neighboring stations within a radius of 100 km to the target station was first calculated and then sensitivity experiments were performed for each specific number. Specifically, ten stations were randomly selected for each given number, and then 20 days gap-free PM_{2.5} observations were sampled at each individual station. For each gap-free PM_{2.5} observation within 24-h, 795 six values were retained and then treated as gaps for cross validation while the DCCEOF method was finally applied to restore these values. It is indicative that the DCCEOF method would have high prediction accuracy with an adequate number of neighboring stations, as three neighboring stations suffice to yield promising prediction accuracy (Figure 9b). On the other hand, large biases could be introduced to the final predictions with a limited number of neighboring stations (<3) due to the lack of sufficient 800 prior information from neighbors to reconstruct the diurnal cycle of PM_{2.5}. Nevertheless, good accuracy still can be guaranteed even in the absence of prior spatial information (that is, no neighboring station within 100 km), which in turn corroborates the beneficial effect of the inclusion of temporal neighborhood in gap filling. Although the prediction accuracy improves with the increase in the number of neighboring stations, the gains of accuracy is not significant at stations with more than three neighboring stations. This 805 is because we only use the similar observations rather than all available observations within 100 km to reconstruct the diurnal cycle of PM_{2.5}; otherwise, irrelevant observations would distort the reconstructed diurnal variation pattern and in turn the final predictions. Nevertheless, the increase in the number of neighboring stations would reduce the uncertainties in the final predictions, which is evidenced by smaller standard deviations of correlation coefficients for those with more neighboring stations (Figure 9b). 810 Moreover, the diurnal cycle reconstructed from the neighborhood field in space is more accurate than using PM_{2.5} observations from near-term days, which is evidenced by smaller correlation values with limited neighboring stations. Such effect is also in line with our recent results when comparing the beneficial effects of spatial and temporal neighboring terms in advancing the gridded PM_{2.5} concentration mapping (Bai et al., 2019c). 815 Figure 10 shows the benefits of the DCCEOF method on our retrieved in-situ hourly PM_{2.5} concentration record at each individual monitoring station in terms of the improvement of data completeness ratio as

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well as the reduction of gap frequency. After applying the DCCEOF method, the data completeness ratio of hourly PM_{2.5} concentration records in China has been improved by approximately 5% on average nationwide, with the overall data completeness ratio increasing from 89.2% to 94.3% (Figure 10a).
 830 Despite the small magnitude of data completeness improvement ratio, the occurrence frequency of missingness has been significantly reduced, with the averaged frequency of days with missingness declined from 42.6% to 5.7% nationwide (Figure 10b). In general, the gap-filled PM_{2.5} record is temporally more complete given fewer data gaps and this data set can thus be used as a promising data source for PM_{2.5}-related studies in the future.

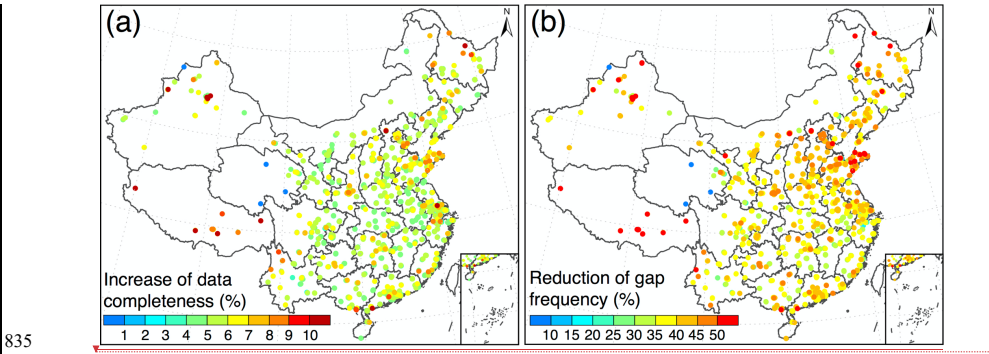
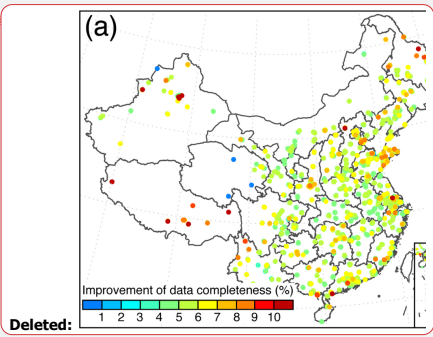


Figure 10. Benefits of the DCCEOF method on China in-situ hourly PM_{2.5} records at each individual monitoring station. (a) Improvement of data completeness ratio, and (b) reduction of the percentage of days with missingness.

840 4.3 Discussion

Compared with conventional interpolation approaches, the suggested DCCEOF method has better accuracy in predicting missing values for the emerged data gaps in hourly PM_{2.5} time series given the principle of accounting for the local diurnal variation pattern of PM_{2.5} concentration. Specifically, site specific diurnal cycle of PM_{2.5} was reconstructed from the discrete spatial and temporal neighborhood
 845 using EOF and was then used as a reference to predict missing values. Such a scheme enabled DCCEOF to capture the local variation pattern of PM_{2.5} with good accuracy in regions with dense neighboring



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stations (e.g., eastern China) and less temporal dynamics of PM_{2.5}. In contrast, relatively poor accuracy could be attained in the western part of the country where stations are sparsely distributed given the lack of adequate neighboring information. In such context, the performance of DCCEOF could be further improved using a general diurnal variation pattern of PM_{2.5} that is firstly determined though a typical classification. However, this endeavor needs us to have a clear prior information of diurnal variability of PM_{2.5} in space and time. On the other hand, the diurnal variation pattern of other relevant factors that are highly related to PM_{2.5} variations, e.g., meteorological factors such as mixing layer height, might be also applied to advance the reconstruction of the local diurnal cycle of PM_{2.5}. Although the DCCEOF method has a promising accuracy in filling data gaps present in hourly PM_{2.5} concentration time series, the current method only works for days with at least several valid observations. In other words, the DCCEOF method is incapable of restoring values for days with all 24-h data missed. This is because the remnant valid observations within 24-h are used as a critical constraint not only to convolve with other neighboring observations in space and time to identify similar observations but also to determine the data magnitude of predicted values for missingness. Moreover, the severity of data gaps in the initial neighborhood field is also associated with the final prediction accuracy because significant data gaps in the neighborhood field could introduce large bias to the reconstructed local diurnal variation pattern. In such context, aforementioned proxy information such as the diurnal variation pattern of meteorological conditions could be applied as a good complementary.

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5 Conclusions

A novel yet practical gap filling method termed DCCEOF was introduced in this study to cope with annoying data gaps in geophysical time series, particularly for those with significant diurnal variability. Compared with the conventional interpolation methods, the suggested DCCEOF method is self-consistent, physically meaningful, and more accurate, given the accounting for the local diurnal variation pattern of the monitoring factor in missing value restorations. Such an endeavor enables the DCCEOF method to predict missing values even at inflection times, like daily peaks or minima that conventional methods always fail to predict properly, with promising accuracy.

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895 A practical application of the DCCEOF method to the China in-situ hourly PM_{2.5} concentration record reveals a good prediction accuracy of the DCCEOF method in restoring PM_{2.5} missingness. The method performs even better in predicting missing values during polluted phases than on clean days given smaller variations of PM_{2.5} concentration in space and time. Further sensitivity experiments suggest that the overall accuracy of the DCCEOF method would slightly decrease (from 0.96 to 0.9) with the increase in
900 the amount of missingness in daily 24-h PM_{2.5} observations. This effect is associated with larger uncertainties in the reconstruction of local PM_{2.5} neighborhood fields since valid observations are required to convolve with other observations ~~to pinpoint~~ observations with similar variation pattern. Also, an adequate number of neighboring stations in space is essential to the final prediction accuracy of missing value restoration. The experimental results suggest that three neighboring stations within 100 km to the
905 target station would yield a promising prediction accuracy, and the more the neighboring stations, the less the uncertainties in the final predicted values.

In addition, we also assessed the severity of data gaps in our retrieved China in-situ hourly PM_{2.5} records. In general, the missingness ratio was less than 10% over most stations across China while data gaps occurred more frequently at 06:00 and 12:00 BJT than during other times. After gap filling, the data
910 completeness ratio of China in-situ hourly PM_{2.5} ~~concentration~~ record was improved to 94.3% while the frequency of days with missingness was markedly reduced from 42.6% to 5.7%. The gap-filled hourly PM_{2.5} concentration record can thus be used as a promising data source for better PM_{2.5} concentration mapping and exposure assessment.

Overall, the ~~suggested~~ DCCEOF method provides a realistic and promising way to deal with missingness
915 emerged in hourly PM_{2.5} concentration record which oftentimes exhibits significant diurnal variation patterns. Given the self-consistent nature, the ~~suggested~~ DCCEOF method can be ~~easily~~ applied to PM_{2.5} datasets measured in other regions and ~~other geophysical records~~ with similar barriers. A more general comparison of this method with many other conventional gap filling methods will be conducted in the future to further examine the performance and accuracy of the DCCEOF method in handling various types
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