Investigating PM$_{2.5}$ responses to other air pollutants and meteorological factors across multiple temporal scales

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It remains unclear on how PM$_{2.5}$ interacts with other air pollutants and meteorological factors at different temporal scales, while such knowledge is crucial to address the air pollution issue more effectively. In this study, we explored such interaction at various temporal scales, taking the city of Nanjing, China as a case study. The ensemble empirical mode decomposition (EEMD) method was applied to decompose time series data of PM$_{2.5}$, five other air pollutants, and six meteorological factors, as well as their correlations were examined at the daily and monthly scales. The study results show that the original PM$_{2.5}$ concentration significantly exhibited non-linear downward trend, while the decomposed time series of PM$_{2.5}$ concentration by EEMD followed daily and monthly cycles. The temporal pattern of PM$_{10}$, SO$_2$ and NO$_2$ is synchronous with that of PM$_{2.5}$. At both daily and monthly scales, PM$_{2.5}$ was positively correlated with CO and negatively correlated with 24-h cumulative precipitation. At the daily scale, PM$_{2.5}$ was positively correlated with O$_3$, daily maximum and minimum temperature, and negatively correlated with atmospheric pressure, while the correlation pattern was opposite at the monthly scale.

Due to rapid urbanization and industrialization and mounting energy consumption, fine particulates have become a major air pollutant in China’s urban atmosphere$^1$. As one of the most important fine particles, PM$_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 $\mu$m) consists of heavy metals, volatile organic compounds, and carbonaceous substances$^3$, which could produce detrimental effects on human respiratory system$^3$. The World Health Organization (WHO) considers a concentration of PM$_{2.5}$ over 10 $\mu$g/m$^3$ as hazardous to human health$^4$, while this measurement in large cities of China can range from 50 to 125 $\mu$g/m$^3$.$^5$ It is estimated that long-term exposure to high level of particulate pollution caused 1.36 million premature deaths per year in China$^6$.

To better inform air quality control and build sustainable cities, tremendous efforts have been devoted to understanding potential contributing factors of PM$_{2.5}$ concentration. It is widely recognized that PM$_{2.5}$ are mainly influenced by human activities, natural environment, and atmospheric chemical reactions. Among these three factors, human activities, such as vehicle exhaust emission and industrial production$^7,8$, are the dominant factors of PM$_{2.5}$ pollution$^7$. Natural environment, such as meteorological conditions (e.g., precipitation and wind speed)$^9$, facilitate the transportation and diffusion of PM$_{2.5}$, while atmospheric chemical reactions stimulate the secondary formation of PM$_{2.5}$$^{10}$. All of these factors interact with PM$_{2.5}$ at different spatial and temporal scales, and thus can have varying effects on PM$_{2.5}$ distribution. Tremendous efforts have been paid to understanding such varying effects at multiple spatial scales, such as the national, provincial, and urban cluster scales. In existing literature, the effects of these factors on PM$_{2.5}$ have been primarily investigated at the seasonal scale, while those at the hourly, daily and monthly scales have been understudied. Lack of such knowledge makes it imperative to identify temporally dependent strategies for PM$_{2.5}$ control.

Ensemble Empirical Mode Decomposition (EEMD), a new time-series signal processing method, was recently proposed with the support of signal detection technology$^{12}$. EEMD was designed for non-stationary and non-linear signal detection and can gradually separate different oscillation or trend components from the original

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signal. In recent years, EEMD has been adopted by a number of scholars to analyze time series data of PM2.5, but few of them used it to investigate PM2.5 over multiple temporal scales. This article attempts to fill this gap by taking the city of Nanjing, China as the study area. Specifically, the objectives of this study are to: (1) explore the variation of PM2.5 at multiple temporal scales, and (2) investigate how PM2.5 responds to air pollutants and meteorological factors over different temporal scales. Findings from our research can offer better understanding on the temporal variability of PM2.5, and improve regional air quality assessment and air pollution source apportionment.

Data and methods

Study area. Nanjing is the capital city of Jiangsu Province in eastern China (Fig. 1). It is situated in the subtropical monsoon climate zone and has four distinct seasons. On the one hand, Nanjing is one of the four garden cities in China, with park green area of 16.01 m² per capita. On the other hand, it is a heavily air-polluted area by PM2.5, attributed to the combined effects of large amount of population and on-road vehicles, valley basin landform, numerous polluting enterprises in the suburbs (e.g., petrochemical factories), and the prevailing wind direction of east-south-east (Fig. 1).

Data. This study acquired three datasets regarding PM2.5 concentrations, other air pollutants, and meteorological factors as described below:

PM2.5 and air pollution factors. Besides PM2.5, five other major air pollutants monitored in China were included in the analysis, namely, PM10, SO2, NO2, CO and O3. We retrieved the hourly concentrations of PM2.5 and five other air pollutants of Nanjing between January 1, 2014 and December 31, 2018 from the Jiangsu environmental data public service station (https://218.94.78.75:20001/sjzx/).

There are nine national monitoring stations in Nanjing (Fig. 1). The hourly concentrations of PM2.5 and other five pollutants were derived from the nine stations, and were averaged to indicate daily concentrations of the entire city.

Meteorological factors. Based on previous studies, wind speed, precipitation, atmospheric pressure, temperature and humidity have shown a strong correlation with PM2.5, and thus were considered as meteorologi-
cal factors in this research. For the same time period (January 1, 2014–December 31, 2018), the daily average wind speed (WS), 24-h cumulative precipitation (PR), daily average atmospheric pressure (AP), daily maximum temperature (MaxT), daily minimum temperature (MinT) and daily surface air relative humidity (RH) for the Nanjing city were retrieved from the National Meteorological Information Center website (https://data.cma.cn/) which were reported from the Nanjing weather station, shown in Fig. 1.

**EEMD.** EEMD was developed from the Empirical Mode Decomposition (EMD). EMD is a signal analysis method suitable for non-stationary, nonlinear data that can decompose any type of signal in an adaptive manner. The aim of EMD is to decompose raw signals into a set of intrinsic mode functions (IMFs) to better reveal the signal characteristics. The IMFs need to meet the following requirements: (1) the number of extreme and the number of zero crossings must be equal to each other or at most differ by one; (2) at any point, the mean value of the envelope defined by the local maximum and local minimum must be zero.

Although EMD has better adaptability, there is a defect of mode mixing due to the influence of the original signal frequency. Therefore, on the basis of EMD, Wu and Huang proposed a noise-assisted data analysis method—Ensemble Empirical Mode Decomposition (EEMD). EEMD effectively processes the ‘mode mixing’ issue in EMD by adding white noise to filter IMF components. Meanwhile, EEMD can also complete the significance test of the IMF, and the IMF component and the residual of the total signal of the same scale in the original signal (quasi-periodic changes) were also revealed by the IMF components, which reflect the nonlinear relationship between natural driving processes within the atmospheric system and the external human activities, and accurately and succinctly reflect the relationships between PM<sub>2.5</sub> and both other air pollutants and meteorological factors. The inherent oscillations of different temporal scales in the original signal (quasi-periodic changes) were also revealed by the IMF components, which reflect the nonlinear relationship between natural driving processes within the atmospheric system and the external human activities, and accurately and succinctly reflect the relationships between PM<sub>2.5</sub> and both other air pollutants and meteorological data.
Results

Descriptive statistics of time series data. Descriptive statistics of PM$_{2.5}$ data with other air pollutants and meteorological factors data of Nanjing from January 1, 2014 to December 31, 2018 are calculated (Table 1). The average PM$_{2.5}$ concentration is 52 μg/m$^3$, but the standard deviation is large, indicating the sample data is fluctuant greatly, which has certain research significance.

| Air pollution indicator | Unit | Mean | Standard deviation | Meteorological indicator | Unit | Mean | Standard deviation |
|-------------------------|------|------|--------------------|--------------------------|------|------|--------------------|
| PM$_{2.5}$              | μg/m$^3$ | 52   | 36                 | WS                       | m/s  | 3    | 1                  |
| PM$_{10}$               | μg/m$^3$ | 92   | 54                 | PR                       | mm   | 28   | 104                |
| SO$_2$                  | μg/m$^3$ | 17   | 10                 | AP                       | Pa   | 1013 | 9                  |
| NO$_x$                  | μg/m$^3$ | 47   | 19                 | MaxT                     | °C   | 21   | 9                  |
| CO                      | mg/m$^3$ | 0.94 | 0.35               | MinT                     | °C   | 13   | 9                  |
| O$_{3,8h}$              | μg/m$^3$ | 104  | 51                 | RH                       | %    | 73   | 14                 |

Table 1. Descriptive statistics of air quality and meteorological factors data of Nanjing in 2014–2018. WS—daily average wind speed, PR—24-h cumulative precipitation, AP—daily average atmospheric pressure, MaxT—daily maximum temperature, MinT—daily minimum temperature, RH—daily surface air relative humidity.

Temporal variation of PM$_{2.5}$. Undecomposed temporal variation of PM$_{2.5}$. Results from the anomaly processing of the average daily PM$_{2.5}$ data suggest that the original daily average PM$_{2.5}$ concentration in Nanjing presented a significant volatility reduction trend during 2014–2018 (Fig. 2). Before December 2017, the PM$_{2.5}$ concentration was relatively higher and presented a weak downward trend, while after that the PM$_{2.5}$ was lower and exhibited the seasonality of “decrease–increase”, i.e., high concentration in spring and winter and low concentration in summer and autumn.

The PM$_{2.5}$ concentration varied dramatically from day to day. The difference between the highest pollution and the lowest was up to 297 μg/m$^3$. Meanwhile, the 30-day moving averages curve of PM$_{2.5}$ concentration implied non-linear and non-stationary volatility characteristics of PM$_{2.5}$. Therefore, the EEMD method was needed to further reveal the complex process of PM$_{2.5}$ in Nanjing.

Decomposed temporal variation of PM$_{2.5}$. The daily average time series of original PM$_{2.5}$ concentration was decomposed by EEMD. For decomposition, the ensemble number was set to 100 and the amplitude of added noise was set to 20% of the standard deviation of the original data. The EEMD analysis produced nine IMF components (IMF1-9) and one trend component (RES), as shown in Fig. 3. Each IMF component a range of frequencies from high (HF, less than a 30-day period) to low (LF, greater than or equal to a 30-day period) at different temporal scales, and the final trend component represents the trend of the original PM$_{2.5}$ data over time.
The significance of each IMF component was tested (Table 2). Except for IMF4, all other components passed the 5% significance level test, indicating that these IMF components contain information with actual physical meaning and the corresponding oscillation periods are the main oscillation period of the PM$_{2.5}$ time series. The average daily concentration change of PM$_{2.5}$ has a relatively stable quasi-periodicity (Fig. 3 and Table 2), following the daily circle and the monthly circle. At the daily scale, it exhibits periodic changes of quasi-3 days (IMF1), quasi-7 days (IMF2), quasi-14 days (IMF3) and quasi-27 days (IMF4); At the monthly scale, it exhibits periodic changes of quasi-60 days (IMF5), quasi-140 days (IMF6), quasi-406 days (IMF7), quasi-913 days (IMF8) and quasi-1826 days (IMF9).

According to Table 2, the variance contribution rates of IMF1, IMF2, IMF3, IMF4, IMF5, IMF6, IMF7, IMF8, IMF9 and RES of the daily average concentration of PM$_{2.5}$ in Nanjing are 19.98%, 16.33%, 8.59%, 4.72%, 3.96%, 4.86%, 11.55%, 1.96%, 1.53% and 26.55%, respectively, showing the fact that the contribution rate of high-frequency signal to low-frequency signal of PM$_{2.5}$ concentration gradually decreases. Although the IMF4 component obtained by EEMD decomposition does not pass the significance test, it still contributes to the fluctuation of the original data series.

The influence of PM$_{2.5}$ daily oscillation (IMF1 ~ 4) on the overall variation of PM$_{2.5}$ is 49.61%, while the monthly oscillation (IMF5 ~ 9, RES) is 50.39%, and the contribution of monthly oscillation is slightly higher than that of daily oscillation. Therefore, the daily and monthly scales are two important temporal scales in the PM$_{2.5}$ time series in Nanjing, which is consistent with the temporal of PM$_{2.5}$ that found in other studies as daily, monthly, and annual.

The results of PM$_{2.5}$ decomposition in Nanjing were reconstructed at both the daily scale and monthly scale and compared with the original PM$_{2.5}$ concentration and trend (Fig. 4). The inter-day variation was obtained by adding the IMF1, IMF2, IMF3, and IMF4, which can be considered as filtering out large-scale oscillations, while the inter-month variation was obtained by adding the IMF5, IMF6, IMF7, IMF8, IMF9 and RES (including larger time scale fluctuation over the study period).

The reconstructed inter-day variation of PM$_{2.5}$ is basically consistent with the original sequence change trend, which can well show the slight change of PM$_{2.5}$ concentration during the study period, while the reconstructed inter-month variation of PM$_{2.5}$ shows a fluctuation of high concentration in spring and winter and low concentration in summer and autumn throughout the study period, which can effectively reveal the high-low state of PM$_{2.5}$ in Nanjing in different seasons. The inter-day and inter-month fluctuations of PM$_{2.5}$ can reflect the fluctuations of the original PM$_{2.5}$ series from different temporal and have obvious inter-modulation effects as well.

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**Figure 3.** The results of EEMD decomposition of PM$_{2.5}$ daily average concentration of Nanjing in 2014–2018.

| Time sale | On the inter-day scale | On the inter-month scale |
|-----------|------------------------|--------------------------|
| IMF component | IMF1 | IMF2 | IMF3 | IMF4 | IMF5 | IMF6 | IMF7 | IMF8 | IMF9 | RES |
| Relief period (days) | 3 | 7 | 14 | 27 | 60 | 140 | 406 | 913 | 1826 |
| Confidence level | > 95% | > 95% | > 95% | > 50% | > 95% | > 95% | > 95% | > 95% | > 95% |
| Variance contribution rate (%) | 19.98 | 16.33 | 8.59 | 4.72 | 4.86 | 11.55 | 1.96 | 1.53 | 26.55 |

**Table 2.** Period, significance level and variance contribution rate of each IMF component of PM$_{2.5}$ concentration.
Response of PM$_{2.5}$ to air pollutants and meteorological fluctuations. Undecomposed temporal variation of air pollutants and meteorological factors. During the study period, the trends of air pollutants PM$_{10}$, SO$_2$, NO$_2$, CO, O$_3$$_8$h were "decrease–increase", "progressively decrease", "decrease–increase", "increase–decrease" and "increase–decrease", respectively (Fig. 5a). The trends of meteorological fluctuation—Wind Speed, Precipitation, Air Pressure, MaxT, MinT, and Relative Humidity were "progressively decrease", "increase–decrease", "decrease–increase", "increase–decrease" and "increase–decrease" and "progressively decrease", respectively (Fig. 5b). The temporal patterns of PM$_{10}$, SO$_2$ and NO$_2$ are similar to PM$_{2.5}$.

The turning points of CO, O$_3$$_8$h, Precipitation, Air Pressure, MaxT, and MinT were December 2014, September 2015, April 2016 and March 2017, respectively, while that of PM$_{2.5}$ was December 2017, indicating that the change of PM$_{2.5}$ had time lags. As important indicators of air quality, CO and O$_3$ were inextricably linked to the formation of PM$_{2.5}$. Therefore, PM$_{2.5}$ was found to be responsive to CO, O$_3$, PR, AP, MaxT, and MinT.
Decomposed temporal variation of air pollutants and meteorological factors. Absolute anomalies in the air pollutants concentration and meteorological factors data were decomposed by EEMD, and the periods were counted (Table 3). The inter-day variation period of air pollutants in Nanjing was highly consistent with PM$_{2.5}$, while at the inter-month scale, the variation period of air pollutants was different from PM$_{2.5}$. Similarly, the inter-day variation period of meteorological factors in Nanjing was generally consistent with PM$_{2.5}$. However, on the inter-month scale, the meteorological factors show larger inter-year variation, which was consistent with the large time scale characteristics of meteorological fluctuation.

Correlation between PM$_{2.5}$ and atmospheric pollutions and meteorological factors. Correlation analysis was conducted between PM$_{2.5}$ and the six metrics at the both reconstructed scale (at the inter-day and the inter-month scale) and original data. As shown in Fig. 6, PM$_{2.5}$ was differently correlated with CO, O$_{3,8h}$, PR, AP, MaxT, and MinT at different temporal scales.

The correlation between PM$_{2.5}$ and CO was positively on the daily scale, and the correlation coefficient was slightly higher than that on the monthly scale. The correlation coefficient of the original data was between the two scales. The correlation between PM$_{2.5}$ and O$_{3,8h}$ was slightly negative when using the original data, but it was positive at the daily scale. At the monthly scale, the correlation was negative and the correlation coefficient was greater than that at the daily scale.

The correlation between PM$_{2.5}$ and PR was negative at both scales and more intense at a broader time scale (inter-month scale). PM$_{2.5}$ and AP were negatively correlated at the inter-day scale, while at the inter-month scale, it was a stronger positive correlation. The original PM$_{2.5}$ showed a significantly negative correlation with MaxT and MinT, and the correlation was positive on the smaller time scale (inter-day scale) but strongly negative at the larger time scale (inter-month scale).

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Table 3. Period of PM$_{2.5}$ concentration, atmospheric pollutants and meteorological factors in Nanjing from 2014 to 2018. *The factor period is consistent with the PM$_{2.5}$ concentration change period.

| Relief period (days) | PM$_{2.5}$ | Atmospheric pollution factor | Meteorological factor |
|----------------------|------------|-----------------------------|-----------------------|
|                      |            | PM$_{10}$ | SO$_2$ | NO$_2$ | CO   | O$_{3,8h}$ | WS  | PR  | AP   | MaxT | MinT | RH  |
| Day scale            | 3          | 3*        | 3*    | 3*    | 3*   | 3*        | 3*  | 3*  | 3*   | 3*   | 3*   |
|                      | 7          | 7*        | 7*    | 7*    | 6    | 7*        | 7*  | 7*  | 7*   | 7*   | 7*   |
|                      | 14         | 14*       | 14*   | 14*   | 14*  | 13        | 12  | 15  | 14*  | 13   | 15   |
|                      | 27         | 27*       | 29    | 29    | 29   | 26        | 24  | 29  | 27*  | 26   | 31   |
| Month scale          |            | 60        | 56    | 57    | 61   | 55        | 59  | 48  | 43   | 55   | 58   | 63   |
|                      |            | 140       | 140*  | 135   | 146  | 140*      | 146 | 118 | 94   | 365  | 365  | 166  |
|                      |            | 406       | 406*  | 406*  | 406* | 332       | 406*| 281 | 159  | 522  | 522  | 609  | 365  |
|                      |            | 913       | 913*  | 730   | 913* | 730       | 1217| 406*| 365  | 1826 | 1217 | 457  |
|                      |            | 1826      | 1826* | 1217  | 1826*| 1217      | 1826*| 1217| 730  | 3652 | 3652 | 1826*|

Figure 6. Multi-scale correlations between PM$_{2.5}$ and CO, O$_{3,8h}$, PR, AP, MaxT and MinT.
In summary, the responses of PM$_{2.5}$ to O$_3$, 8h, AP, MaxT and MinT were in opposite directions at the daily and monthly scales. The responses were in the same direction for PR and CO at both temporal scales, while the response to PR is more intense at the monthly scale but the response to CO is more obvious at the daily scale. PM$_{2.5}$ has different degrees of significant correlation with other different pollutants and meteorological factors at different temporal, so it has a multi-scale response.

**Discussion**

This study contributes to answering the broader question on the anthropogenic and environmental factors that affect the formation of PM$_{2.5}$ pollution. The sources of pollution caused by human activities mainly include fixed sources with fossil fuel combustion and mobile sources with vehicle exhaust emissions. The sulfur and nitrogen oxides produced by them can be directly converted into PM$_{2.5}$. Natural environment mainly involves meteorological factors such as wind speed, precipitation, air pressure, temperature, and humidity, etc. The favorable meteorological conditions conducive to the diffusion and dilution of PM$_{2.5}$ and reduce the risk of pollution.

Human activities are the main cause of PM$_{2.5}$ pollution, and natural environment plays an important role in the aggregation and diffusion of particulate matter. On a short time scale, human activities and environmental factors (especially meteorological conditions) can change dramatically, and thus the contribution of human activities to PM$_{2.5}$ is fluctuant; On a long time scale, human activities have undergone major changes due to the regulation of macroeconomic policies, while overall changes in environmental conditions are small, so the contribution of human activities to PM$_{2.5}$ is stable.

Our findings advance the understanding of how PM$_{2.5}$ responds to the other air pollutants at different temporal scales. PM$_{10}$ is a particle with the size less than 10 μm, which contains PM$_{2.5}$, and its variability is often synchronous to PM$_{2.5}$. SO$_2$ and NO$_2$ are important precursors in the formation of PM$_{2.5}$, and their trends are similar to PM$_{2.5}$. CO is an intermediate product formed during the combustion of carbon-containing fuels, and it is not directly related to PM$_{2.5}$. However, studies have shown that the spatial distribution of CO and particulate matter concentration tends to be the same in most parts of the day, which supports the conclusion in this study that PM$_{2.5}$ has a strong positive correlation with CO. Under the macro-control of low-carbon emissions, the development of a low-carbon economy in Nanjing controls carbon emissions from adjusting industrial structure, improving energy efficiency and increasing ecological land. Carbon management was carried out from both “source” and “sink”, which also effectively reduced the production of CO. Meanwhile, the vigorous development of new energy vehicles in recent years has alleviated carbon emissions from mobile sources. The production of PM$_{1.0}$, which is based on carbon fuel combustion and vehicle exhaust emissions, is effectively controlled. Therefore, low-carbon policies of energy saving, new energy development is conducive to the regulation of PM$_{2.5}$.

O$_3$ is a pollutant formed by the conversion of nitrogen oxides under light radiation and suitable meteorological conditions, and it is also not directly related to PM$_{2.5}$. However, studies have shown that the correlation between O$_3$ and PM$_{2.5}$ in different seasons is reversed, which is similar to the findings in this study. PM$_{2.5}$ and O$_3$ showed significant inverse correlations on the temporal of inter-day and inter-month and the correlation coefficient on the inter-month scale is larger. The source of PM$_{2.5}$ and O$_3$ is consistent. The high concentration of O$_3$ can promote the formation of secondary particles under strong atmospheric oxidation conditions and increase the concentration of PM$_{2.5}$ and high concentration of PM$_{2.5}$ can weaken solar radiation and inhibit the production of O$_3$. On the small time scale, source consistency is an important reason for the significant positive correlation between PM$_{2.5}$ and O$_3$, while on the large time scale, the rapid advancement of urbanization, industrialization, and motorization, and massive emission of atmospheric active substances are the main reasons for the increase in surface O$_3$ concentration. Since the implementation of the "Air Pollution Prevention Action Plan", the cooperation mechanism for air pollution prevention and control in the three provinces and one city of the Yangtze River Delta has effectively curbed the growth of PM$_{2.5}$, but O$_3$ pollution has become increasingly severe, which indicates that the issue of O$_3$ pollution should be further addressed in the continued implementation of the ten policies of the atmosphere.

How does PM$_{2.5}$ respond to the meteorological factors at different temporal scales? The dilution effect of wind speed on PM$_{2.5}$ rises at first and then tends to be gentle, and high humidity environment has the effect of agglomerating PM$_{2.5}$. Studies have shown that precipitation contributes to the dilution of PM$_{2.5}$, and PM$_{2.5}$ concentration before and after precipitation has an important effect on its dilution effect. This study shows that the dilution effect of PM$_{2.5}$ is more significant with the increase of time scale, which provides a new idea for the impact of precipitation on the macroscopic time scale of PM$_{2.5}$.

Few studies have investigated the effect of air pressure on PM$_{2.5}$, and this study found that the response of PM$_{2.5}$ to air pressure is inversely correlated at different temporal scales and is more strongly correlated on the large time scale. Under the low-pressure circulation situation, there are more rainy days and the wind direction changes more frequently, which helps the diffusion and dilution of particulate matter; while the high-pressure circulation situation brings more sunny days and the weather system is relatively stable, forcing the particulate matter to stagnate in the near-surface layer. Hence, the response of PM$_{2.5}$ to air pressure appears to be positive on a large time scale.

This study found that PM$_{2.5}$ and temperature have a significant correlation at the daily scale, meanwhile, the results show that PM$_{2.5}$ and temperature are significantly negative on the monthly scale, in line with the distribution characteristics of PM$_{2.5}$’ “winter-high, summer-low” in Nanjing.

This study has shown that there exist seasonal variations of the correlation between PM$_{2.5}$ concentration and meteorological factors. Therefore, the multi-temporal scales cannot be ignored when studying the influence of the natural environment (especially meteorological factors) on PM$_{2.5}$. Meanwhile, the multi-temporal scale provides a new perspective for PM$_{2.5}$ research.
Conclusion

By investigating PM$_{2.5}$ responses to other air pollutants and meteorological factors at across multiple temporal scales, this research generated three major findings. First, the original daily average concentration of PM$_{2.5}$ exhibited a significant downward trend and the periodic law of "decrease–increase"; which had time lags compared with other air pollutants (CO, O$_3$) and meteorological fluctuation (PR, AP, MaxT, and MinT). Second, the decomposed temporal variation of PM$_{2.5}$ by EEMD has a relatively stable quasi-periodicity in the inter-day and the inter-month temporal scale, and followed daily circle and monthly circle. Third, the responses of PM$_{2.5}$ to other air pollutants and meteorological factors varied at different temporal scales. The temporal pattern of PM$_{2.5}$, SO$_2$, and NO$_x$ is synchronous with that of PM$_{2.5}$. At the daily and monthly scales, PM$_{2.5}$ was positively correlated with CO and negatively correlated with 24-h cumulative precipitation. At the daily scale, PM$_{2.5}$ was positively correlated with O$_3$, daily maximum and minimum temperature, and negatively correlated with atmospheric pressure, while the correlation pattern was opposite at the monthly scale.

Such findings help improve our understanding on how PM$_{2.5}$ responds to the changes of other air pollutants and meteorological fluctuation at different temporal scales, which contribute to future regional air quality assessment and source apportionment studies. They also suggest that in order to mitigate PM$_{2.5}$ and other air pollution problems, different measures should be devised to target different time scales (especially at inter-day scale and inter-month scale). Especially, the issue of O$_3$ increase should be further addressed when we have effectively curbed the growth of PM$_{2.5}$ at a longer temporal scale.

It is important to point out that China has begun to publicize the PM$_{2.5}$ concentration data since December 2013, thus the PM$_{2.5}$ time series of Nanjing selected in this study started on January 1, 2014. With the continuous accumulation of data, longer temporal scales can be considered in future research, especially the inter-annual and inter-decadal variations of PM$_{2.5}$. In addition, the source analysis of the multi-temporal response of PM$_{2.5}$ from two perspectives of human activities (air pollutants) and natural environment (mainly meteorological factors) is mainly realized by qualitative methods so far, and can be further explored by quantitative methods in the future research.

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References

1. Chan, C. K. & Yao, X. Air pollution in mega cities in China. Atmos. Environ. 42, 1–42. https://doi.org/10.1016/j.atmosenv.2007.09.003 (2008).
2. Bandowe, B. A. et al. PM$_{2.5}$-bound oxygenated PAHs, nitro-PAHs and parent-PAHs from the atmosphere of a Chinese megacity: seasonal variation, sources and cancer risk assessment. Sci. Total Environ. 473–474, 77–87. https://doi.org/10.1016/j.scitotenv.2013.11.108 (2014).
3. Wu, S. et al. Fine particulate matter, temperature, and lung function in healthy adults: findings from the HVNR study. Chemosphere 108, 168–174. https://doi.org/10.1016/j.chemosphere.2014.01.032 (2014).
4. Organization, W. H. WHO air quality guidelines for particulate matter, ozone, nitrogen dioxide and sulfur dioxide: global update 2005: summary of risk assessment (World Health Organization, Geneva, 2006).
5. Liu, Z. et al. Characteristics of PM$_{2.5}$ mass concentrations and chemical species in urban and background areas of China: emerging results from the CARE-China network. Atmos. Chem. Phys. 18, 8849–8871. https://doi.org/10.5194/acp-18-8849-2018 (2018).
6. Lelieveld, J., Evans, J. S., Finais, M., Giannadaki, D. & Pozzer, A. The contribution of outdoor air pollution sources to premature mortality on a global scale. Nature 525, 367–371. https://doi.org/10.1038/nature15371 (2015).
7. Huang, R. I. et al. High secondary aerosol contribution to particulate pollution during haze events in China. Nature 514, 218–222. https://doi.org/10.1038/nature13774 (2014).
8. Zhou, L., Zhou, C., Yang, F., Wang, B. & Sun, D. Spatio-temporal evolution and the influencing factors of PM$_{2.5}$ in China between 2000 and 2011. Acta Geogr. Sin 29, 253–270 (2017).
9. Polissar, A. V., Hopke, P. K. & Poiriot, R. L. Atmospheric aerosol over Vermont: chemical composition and sources. Environ. Sci. Technol. 35, 4604–4621 (2001).
10. Zhang, H., Wang, Y., Hu, J., Ying, Q. & Hu, X. M. Relationships between meteorological parameters and criteria air pollutants in three megacities in China. Environ. Res. 140, 242–254. https://doi.org/10.1016/j.envres.2015.04.004 (2015).
11. Tang, C. H., Coulil, B. A., Schwartz, J., Di, Q. & Koutrakis, P. Trends and spatial patterns of fine-resolution aerosol optical depth-derived PM$_{2.5}$ emissions in the Northeast United States from 2002 to 2013. J. Air Waste Manag. Assoc. 67, 64–74. https://doi.org/10.1080/10962247.2016.1218393 (2017).
12. Wu, Z. & Huang, N. E. Ensemble empirical mode decomposition: a noise-assisted data analysis method. Adv. Adapt. Data Anal. 1, 1–41 (2009).
13. Qin, S., Liu, E., Wang, J. & Sun, B. Analysis and forecasting of the particulate matter (PM) concentration levels over four major cities of China using hybrid models. Atmos. Environ. 98, 665–675. https://doi.org/10.1016/j.atmosenv.2014.09.046 (2014).
14. Zhou, Q., Jiang, H., Wang, J. & Zhou, J. A hybrid model for PM$_{2.5}$ forecasting based on ensemble empirical mode decomposition and a general regression neural network. Sci. Total Environ. 496, 264–274. https://doi.org/10.1016/j.scitotenv.2014.07.051 (2014).
15. Ausati, S. & Amanollahi, J. Assessing the accuracy of ANFIS, EEMD-GRNN, PCR, and MLR models in predicting PM$_{2.5}$. Atmos. Environ. 142, 465–474. https://doi.org/10.1016/j.atmosenv.2016.08.007 (2016).
16. Niu, M., Gan, K., Sun, S. & Li, F. Application of decomposition-ensemble learning paradigm with phase space reconstruction for day-ahead PM$_{2.5}$ concentration forecasting. J. Environ. Manag. 196, 110–118. https://doi.org/10.1016/j.jenvman.2017.02.071 (2017).
17. Bai, Y., Zeng, B., Li, C. & Zhang, J. An ensemble long short-term memory neural network for hourly PM$_{2.5}$ concentration forecasting. Chemosphere 222, 286–294 (2019).
18. Fu, H. & Zhang, Y. Improving the EEMD-GRNN model for PM$_{2.5}$ prediction based on time scale reconstruction. J. Geo-inf. Sci. 21, 1132–1142. https://doi.org/10.1007/s12220-019-00598 (2019).
19. Zheng, S. et al. The spatiotemporal distribution of air pollutants and their relationship with land-use patterns in Hangzhou city, China. Atmosphere 8, 110 (2017).
20. Huang, N. E. et al. The empirical mode decomposition and the Hilbert spectrum for nonlinear and non-stationary time series analysis. Proc. R. Soc. Lond. A Math. Phys. Eng. Sci. 454, 903–995 (1998).
21. Wu, Z. & Huang, N. E. A study of the characteristics of white noise using the empirical mode decomposition method. Proc. R. Soc. Lond. A Math. Phys. Eng. Sci. 460, 1597–1611 (2004).
22. Rahman, M. M., Mahamud, S. & Thurston, G. D. Recent spatial gradients and time trends in Dhaka, Bangladesh, air pollution and their human health implications. *J. Air Waste Manag. Assoc.* 69, 478–501 (2019).

23. Wang, Z., Cai, M., Peng, Z. & Gao, Y. Spatiotemporal distributions of roadside PM$_2.5$ and CO concentrations based on mobile observations. *China Environ. Sci.* 37, 4428–4434 (2017).

24. Chui, X. & Feng, J. High resolution carbon emissions simulation and spatial heterogeneity analysis based on big data in Nanjing City, China. *Sci. Total Environ.* 686, 828–837. [https://doi.org/10.1016/j.scitotenv.2019.05.138](https://doi.org/10.1016/j.scitotenv.2019.05.138) (2019).

25. Yuanxuan, S. et al. Interaction mechanism between PM$_{2.5}$ and O$_3$ in winter and summer in Yunnan–Guizhou plateau: a case study of Guiyang. *Ecol. Environ. Sci.* 27, 2284–2289. [https://doi.org/10.1016/j.ees.2018.12.014](https://doi.org/10.1016/j.ees.2018.12.014) (2018).

26. Chen, J. et al. Temporal and spatial features of the correlation between PM$_2.5$ and O$_3$ concentrations in China. *Int. J. Environ. Res. Public Health* [https://doi.org/10.3390/ijerph16234824](https://doi.org/10.3390/ijerph16234824) (2019).

27. Li, Y. et al. Ozone source apportionment (OSAT) to differentiate local regional and super-regional source contributions in the Pearl River Delta region, China. *J. Geophys. Res. Atmos.* [https://doi.org/10.1029/2011JD017340](https://doi.org/10.1029/2011JD017340) (2012).

28. Jiang, Q., Wang, F., Zhang, H. & Lv, M. Analysis of temporal variation characteristics and meteorological conditions of reactive gas and PM$_2.5$ in Beijing, China. *Environ. Sci.* 37, 829–837 (2017).

29. Deng, L., Qian, J., Liao, R. & Tong, H. Pollution characteristics of atmospheric particulates in Chengdu from August to September in 2009 and their relationship with meteorological conditions, China. *Environ. Sci.* 32, 1433–1438 (2012).

30. Zhou, B., Liu, D., Wei, J. & Peng, H. A preliminary analysis on scavenging effect of precipitation on aerosol particles. *Resour. Environ. Yangtze River Basin* 24, 160–170 (2015).

31. Jia, M. et al. Seasonal variations in major air pollutants in Nanjing and their meteorological correlation analyses. *China Environ. Sci.* 36, 2567–2577 (2016).

32. Zhao, H., Zheng, Y. & Wu, X. Atmospheric compound pollution characteristics and the effects of meteorological factors in Jiangsu Province. *China Environ. Sci.* 38, 2830–2839 (2018).

33. Chen, Z. et al. Detecting the causality influence of individual meteorological factors on local PM$_{2.5}$ concentration in the Jing-Jin-Ji region. *Sci. Rep.* 7, 1–11 (2017).

34. Yang, Q., Yuan, Q., Li, T., Shen, H. & Zhang, L. The relationships between PM$_{2.5}$ and meteorological factors in China: seasonal and regional variations. *Int. J. Environ. Res. Public Health* [https://doi.org/10.3390/ijerph14121510](https://doi.org/10.3390/ijerph14121510) (2017).

35. Chen, Z. et al. Understanding meteorological influences on PM$_{2.5}$ concentrations across China: a temporal and spatial perspective. *Atmos. Chem. Phys.* 18, 5343 (2018).

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Author contributions

H.F. and Y.Z. organized, performed data analysis and wrote the manuscript. C.L. contributed to discussing the results and revision. L.M. contributed to proof reading and revision. Z.W. and N.H. contributed to the figures production.

Competing interests

The authors declare no competing interests.

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