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An improved decomposition method to differentiate meteorological and anthropogenic effects on air pollution: A national study in China during the COVID-19 lockdown period

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HIGHLIGHTS

- An improved method was outlined to assess meteorological effect on air pollution.
- The cause of haze pollution during the COVID-19 lockdown period was investigated.
- Elevated humidity and weakened airflow greatly increased PM$_{2.5}$ in northern China.
- On excluding the meteorological effects, PM$_{2.5}$ substantially decreased across China.

ARTICLE INFO

Keywords:
Air pollution
Meteorology
Pollutant emission
Humidity
COVID-19

ABSTRACT

Although the effects of meteorological factors on severe air pollution have been extensively investigated, quantitative decomposition of the contributions of meteorology and anthropogenic factors remains a big challenge. The novel coronavirus disease 2019 (COVID-19) pandemic affords a unique opportunity to test decomposition methods. Based on a wind decomposition method, this study outlined an improved method to differentiate complex meteorological and anthropogenic effects. The improved method was then applied to investigate the cause of unanticipated haze pollution in China during the COVID-19 lockdown period. Results from the wind decomposition method show that weakened winds increased PM$_{2.5}$ concentrations in the Beijing–Tianjin area and northeastern China (e.g., by 3.19 $\mu$g/m$^3$ in Beijing). Using the improved decomposition method, we found that the combined meteorological effect (e.g., drastically elevated humidity levels and weakened airflow) substantially increased PM$_{2.5}$ concentrations in northern China: the most substantial increases were in the Beijing–Tianjin–Hebei region (e.g., by 26.79 $\mu$g/m$^3$ in Beijing). On excluding the meteorological effects, PM$_{2.5}$ concentrations substantially decreased across China (e.g., by 21.84 $\mu$g/m$^3$ in Beijing), evidencing that the strict restrictions on human activities indeed decreased PM$_{2.5}$ concentrations. The unfavorable meteorological conditions, however, overwhelmed the beneficial effects of emission reduction, causing the severe haze pollution. These results indicate that the integrated meteorological effects should be considered to differentiate the meteorological and anthropogenic effects on severe air pollution.
1. Introduction

Meteorological factors play significant roles in air pollution formation, transport, and dispersion (Hou et al., 2019; Xu et al., 2020; Zou et al., 2017). Therefore, researchers should be cautious when directly attributing air quality improvement to control measures if the effects of meteorological variations are not accounted for. Specifically, high temperature and humidity promote the formation of secondary aerosols via multiphase reactions (Shao et al., 2018; Zheng et al., 2015). Stagnant air mass, which is associated with low wind speed, inhibits the dispersion of air pollutants and thus increases pollutant concentrations near the surface (Liu et al., 2017a; Sun et al., 2019). Numerous severe haze pollution episodes in China during winter have been attributed to such unfavorable meteorological conditions (Tie et al., 2017; Zhang et al., 2014; Zheng et al., 2015).

Although the effects of meteorological factors on the occurrence of severe air pollution have been extensively investigated, quantitative decomposition of the contributions of meteorology and emission reduction remains a big challenge (Zhong et al., 2018). A common method has relied on Chemical Transport Model (CTM) simulations to estimate air pollutant concentrations based on a few conditional scenarios such as under realistic condition, with fixed emission, and with fixed meteorology (Chen et al., 2019; Kang et al., 2019; Zhang et al., 2018). Then, differences between simulation outputs are quantified to understand the relative contributions of meteorology and emissions reduction (Liu et al., 2017c; Liu et al., 2017b; Wang et al., 2017; Zhang et al., 2020; Zhong et al., 2018). For instance, based on the WRF-CMAQ (Weather Research and Forecasting and the Community Multi-scale Air Quality) simulations, Chen et al. (2019) found that the meteorological factors contributed 20% to the change in PM$_{2.5}$ concentration in Beijing from 2013 to 2017. These CTM-based methods entail heavy computational loads and require a detailed investigation of emission inventory (Xing et al., 2020).

Statistical models have also been used to distinguish the meteorological impacts from emission controls (Gui et al., 2019; Li et al., 2019; Meng et al., 2019; Yang et al., 2018). Using a multiple linear regression model, Zhai et al. (2019) found that meteorological factors contributed 12% to the PM$_{2.5}$ trends across China from 2013 to 2018. Plocoste et al. (2019) applied the Empirical Mode Decomposition (EMD) based method and Time Dependent Intrinsic Correlation method (TDIC) to study the relationship between ozone and meteorological values (e.g., humidity and wind) in the Caribbean Basin. Based on wind and pollution roses, Li et al. (2014) developed an observation-based method to decompose wind and non-wind effects on the year-to-year variation in air quality in Hong Kong. Compared to most statistical methods that were based on empirical regressions, this wind decomposition method was unique in the sense that it was based on physical understanding of the relationship between wind and transport of air pollutants. However, impacts of other important meteorological factors (e.g., temperature and humidity) have not been taken into account in this wind decomposition method.

The novel coronavirus disease 2019 (COVID-19) pandemic caused extensive city lockdown around the world (Azman and Luquero, 2020; He et al., 2020). It affords a unique opportunity to test the methods to differentiate meteorological and anthropogenic effects on air pollution. Across much of China, substantial reduction in emissions during the COVID-19 lockdown period has contributed to the improvement in air quality (Chen et al., 2020; Jia et al., 2020; Shi and Brasseur, 2020). However, during this period, extreme haze pollution episodes with high concentrations of particulate matter were also reported in northern China (Nichol et al., 2020; Zhao et al., 2020). Limited understanding of the cause of this haze pollution led to confusions among the public and government.

There have been some CTM-based studies suggested that meteorological factors could have played a critical role in the formation of the severe haze pollution episodes in China during the COVID-19 lockdown period. Using Weather Research and Forecasting model coupled with Chemistry (WRF-Chem), Le et al. (2020) indicated that this severe haze in China formed due to unfavorable weather conditions that promoted secondary aerosol formation via heterogeneous chemical reactions and hindered pollution dispersion. Similar results were obtained using the WRF-CMAQ model, evidencing that the beneficial effects of emission reduction in northern China were stymied by adverse meteorological conditions (Wang et al., 2020). Investigation of the chemical compositions of particulate matter found that the concentrations of secondary pollutants increased substantially during this haze pollution episodes (Dai et al., 2020; Huang et al., 2020).

The CTM simulations are complicated and require prerequisite information of the emission inventory. It is important to use an observation-based method to differentiate the effects of meteorological variables from anthropogenic factors. In the present study, we outlined an improved decomposition method that was based on the wind decomposition method proposed by Li et al. (2014). Results from the previous method that only considered the impact of wind variation can be misleading. Compared to the previous wind decomposition method, the improved method covered various meteorological parameters and estimated integrated effect of meteorological factors on air pollution. The improved method was then applied in a national study in China to investigate the occurrence of unanticipated haze pollution during the COVID-19 lockdown period.

2. Data and methodology

2.1. Study period

The first COVID-19 case was reported in Wuhan, China, in early December of 2019. In response to the ensuing COVID-19 outbreak, the Chinese government implemented a series of containment measures. Figure S1 shows a timeline of major events in China during the COVID-19 outbreak period. A lockdown was imposed in Wuhan, the epicenter of the outbreak, on January 23, 2020. As the disease spread rapidly, the Chinese New Year (CNY) vacation was extended. From the end of February onward, the country began resuming economic activities in phases. On March 11, Hubei province announced that the outbreak was under control in the region and that work could resume. Accordingly, in this study, the primary focus period of the analysis is from January 23 to March 10, 2020. To assess the impact of the COVID-19 containment measures on air quality, we comparatively evaluated air quality variation during the same period (January 23–March 10) in both 2019 and 2020.

2.2. Meteorological data

Meteorological data (wind speed, wind direction, temperature, and relative humidity) in the greater China region for the period January 23–March 10 of both 2019 and 2020 were obtained from the ground-based monitoring network of the World Meteorological Organization (WMO) global telecommunications system. In the study region, the meteorological parameters were measured at 216 stations, as shown in Figure S2. The temporal resolution of the meteorological data is 3 h, and each pair of stations is approximately 100 km apart.

2.3. Air quality data

We obtained hourly PM$_{2.5}$ concentration data for the study period from ground monitoring networks in mainland China (http://www.cnemc.cn/), Taiwan (https://airtw.epa.gov.tw), Hong Kong (http://www.epd.gov.hk/), and Macau (https://www.smg.gov.mo). To obtain the PM$_{2.5}$ concentration at a meteorological station, we calculated the average of the PM$_{2.5}$ concentrations at all air quality stations within 50 km of that meteorological station. Thus, hourly PM$_{2.5}$ concentrations were obtained at 216 meteorological stations.
2.4. Observation-based meteorology-pollution decomposition method

On the basis of ground observations, Li et al. (2014) used a wind-pollution decomposition method to differentiate the effects of wind and non-wind factors on the yearly variation of PM$_{10}$ concentration in Hong Kong. Temperature and humidity can play a critical role in air pollution mechanisms. In this study, we outlined a 4-dimensional (4D) meteorology-pollution decomposition method to differentiate the complex effects of meteorological factors (namely wind, temperature, and relative humidity) and non-meteorological factors on the variation of the PM$_{2.5}$ concentration. The method was then applied in a national study to investigate the cause of unanticipated haze pollution over China during the COVID-19 lockdown period.

For a given time series of meteorological data, we distribute wind speed, wind direction, temperature, and relative humidity into $N_i, N_j, N_k,$ and $N_p$ intervals, respectively. Then, the 4D normalized frequency distribution of the meteorological factors can be denoted as follows:

$$ F_M = \left\{ f_{i,j,k,p} : i = 1, \ldots, N_i; j = 1, \ldots, N_j; k = 1, \ldots, N_k; p = 1, \ldots, N_p \right\} $$

Here, $f_{i,j,k,p}$ ranges from 0 to 1, with its summation equaling 1. We define $F_M$ as a 4D matrix of $f_{i,j,k,p}$ and call it the “meteorological matrix.”

For the time series of the PM$_{2.5}$ concentration data for the same time period as above, the distribution of the PM$_{2.5}$ concentration as a function of the meteorological factors is

$$ C_M = \left\{ c_{i,j,k,p} : i = 1, \ldots, N_i; j = 1, \ldots, N_j; k = 1, \ldots, N_k; p = 1, \ldots, N_p \right\} $$

Here, $c_{i,j,k,p}$ represents the average PM$_{2.5}$ concentration when the wind speed, wind direction, temperature, and relative humidity fall into the $i$th, $j$th, $k$th, and $p$th intervals, respectively. We define $C_M$ as a 4D matrix of $c_{i,j,k,p}$ and call it the “pollution matrix.” In this study, this pollution matrix represents PM$_{2.5}$ matrix. If the method is applied for other air pollutants, for instance, nitrogen dioxide, the pollution matrix represents nitrogen dioxide matrix. Meteorological variation affects the meteorological matrix $F_M$ but not the pollution matrix $C_M$. The pollution matrix changes only when non-meteorological factors (e.g., pollutant emission) change.

The average PM$_{2.5}$ concentration ($\bar{c}$) during the study period can be expressed as the dot product of the meteorological and pollution matrices:

$$ \bar{c} = \sum_{i=1}^{N_i} \sum_{j=1}^{N_j} \sum_{k=1}^{N_k} \sum_{p=1}^{N_p} f_{i,j,k,p} \cdot c_{i,j,k,p} = F_M \cdot C_M $$

(3)

For measurements in two periods, $t_1$ and $t_2$, the average PM$_{2.5}$ concentrations during the two periods can then be expressed as follows:

$$ \bar{c}_{t_1} = F_{M,t_1} \cdot C_{M,t_1} $$

(4.1)

$$ \bar{c}_{t_2} = F_{M,t_2} \cdot C_{M,t_2} $$

(4.2)

Using the Taylor expansion, the average PM$_{2.5}$ concentration during $t_2$ can be expressed as follow:

$$ \bar{c}_{t_2} = \bar{c}_{t_1} + \frac{\partial \bar{c}}{\partial F_M} \Delta F_M + \frac{\partial \bar{c}}{\partial C_M} \Delta C_M + \frac{\partial \bar{c}}{\partial F_M} \cdot \Delta F_M + \frac{\partial \bar{c}}{\partial C_M} \cdot \Delta C_M + o' $$

(5)

Assuming that the changes in $F_M$ and $C_M$ are generally independent, we obtain $\frac{\partial \bar{c}}{\partial F_M} = C_M$, $\frac{\partial \bar{c}}{\partial C_M} = F_M$, and $\frac{\partial \bar{c}}{\partial F_M} \cdot \Delta F_M$ and $\frac{\partial \bar{c}}{\partial C_M} \cdot \Delta C_M$ are 0. The change in PM$_{2.5}$ concentration from $t_1$ to $t_2$ can then be expressed as

$$ \Delta \bar{c} = C_{M,t_2} \cdot (F_{M,t_2} - F_{M,t_1}) + F_{M,t_1} \cdot (C_{M,t_2} - C_{M,t_1}) = \bar{c}_{t_2} - \bar{c}_{t_1} $$

(6)

We define $(F_{M,t_2} - F_{M,t_1}) \cdot (C_{M,t_2} - C_{M,t_1}) = \Delta \bar{c}$, then $\Delta \bar{c}$ can be expressed as

$$ \Delta \bar{c} = C_{M,t_2} \cdot (F_{M,t_2} - F_{M,t_1}) + F_{M,t_1} \cdot (C_{M,t_2} - C_{M,t_1}) + (F_{M,t_2} - F_{M,t_1}) \cdot (C_{M,t_2} - C_{M,t_1}) $$

(7)

The above equation comprises three parts:

$$ \Delta \bar{c}_{t_2} = C_{M,t_2} \cdot (F_{M,t_2} - F_{M,t_1}) $$

(8.1)

$$ \Delta \bar{c}_{t_2} = F_{M,t_1} \cdot (C_{M,t_2} - C_{M,t_1}) $$

(8.2)

$$ \Delta \bar{c}_{t_2} = (F_{M,t_2} - F_{M,t_1}) \cdot (C_{M,t_2} - C_{M,t_1}) $$

(8.3)

$\Delta \bar{c}_{t_2}$, the dot product of $C_{M,t_2}$ and $(F_{M,t_2} - F_{M,t_1})$, represents the change in PM$_{2.5}$ as a result of the meteorological change if there is no change in the pollution matrix $C_M$. The pollution matrix will not change if the non-meteorological factors (e.g., pollutant emission) are the same in $t_1$ and $t_2$. Therefore, $\Delta \bar{c}_{t_2}$ identifies the effects of meteorological factors on the variation of the PM$_{2.5}$ concentration. $\Delta \bar{c}_{t_2}$, the dot product of $F_{M,t_2}$ and $(C_{M,t_2} - C_{M,t_1})$, represents the change in PM$_{2.5}$ as a result of the change in the pollution matrix $C_M$, assuming there is no meteorological change. Because the change in the pollution matrix $C_M$ is associated with changes in the non-meteorological factors, $\Delta \bar{c}_{t_2}$ identifies the non-meteorological effect. Data at a resolution of 3 h may not be able to adequately reveal high-frequency variation in meteorological values and PM$_{2.5}$ concentration. Accordingly, the residue term $\Delta \bar{c}_{t_2} = \varepsilon_t$ represents the unresolved change in PM$_{2.5}$ due to the small sampling period and the non-linear interactions between the meteorological and non-meteorological factors. To indicate the confidence level of the linear decomposition, a linear index was defined as follows:

$$ L = 1 - \frac{\varepsilon_t}{|C_{M,t_2} \cdot (F_{M,t_2} - F_{M,t_1})| + |F_{M,t_1} \cdot (C_{M,t_2} - C_{M,t_1})| + |\varepsilon_t|} $$

(9)

This linear index ranges from 0 to 1, with a higher linear index indicating a smaller residue term. Assuming that the aforementioned non-linear interaction is not considerable, the change in the PM$_{2.5}$ concentration can be decomposed into two main parts resulting from the meteorological and non-meteorological effects. The confidence levels of linear decomposition are 99%, 95%, and 90% if the linear index is larger than 0.98, 0.93, and 0.87, respectively (Li et al., 2014).

To understand the composition of the meteorological effects, the decomposition method is further applied to quantify the individual effects of wind, temperature, and relative humidity:

$$ \Delta \bar{c}_{t_1} \cdot (F_{t_2} - F_{t_1}) + \frac{\partial \bar{c}}{\partial T} \cdot (T_{t_2} - T_{t_1}) + \varepsilon_t $$

(10.1)

$$ \Delta \bar{c}_{t_1} \cdot (F_{t_2} - F_{t_1}) + \frac{\partial \bar{c}}{\partial C_M} \cdot (C_{t_2} - C_{t_1}) + \varepsilon_t $$

(10.2)

$$ \Delta \bar{c}_{t_1} \cdot (F_{t_2} - F_{t_1}) + \frac{\partial \bar{c}}{\partial F_M} \cdot (F_{t_2} - F_{t_1}) + \varepsilon_t $$

(10.3)

For wind decomposition, the meteorological ($F_{t_2}$) and pollution ($C_{t_2}$) matrices are 2D, representing two wind components (i.e., wind speed and direction). In atmospheric sciences, $F_{t_2}$ (namely wind rose) shows normalized frequency distribution of wind as a function of speed and direction (Kassomenos et al., 1995), and $C_{t_2}$ (namely pollution rose) shows average pollutant concentration as a function of wind speed and direction (Ratto et al., 2006). The temperature- and humidity-based meteorological ($T_{t_2}$) and pollution ($C_{t_2}$) matrices are 1D. With the foregoing matrices, the variation in the PM$_{2.5}$ concentration can be decomposed into the wind and non-wind effects, temperature, and non-temperature effects, and humidity, and non-humidity effects. The confidence levels of these decompositions were obtained by examining their linear indexes. Interactions of three meteorological effects were then analyzed. More details on the single-parameter decomposition framework can be found in Li et al. (2014).
3. Results

3.1. Variations of PM$_{2.5}$ and meteorology

PM$_{2.5}$ concentrations during the same period in 2019 and 2020 (January 23 to March 10) were compared. Panels (a) and (b) in Figure S3 show the spatial distributions of the average PM$_{2.5}$ concentration in China for these two periods. In general, the highest PM$_{2.5}$ concentrations were observed in the North China Plain (NCP). The national average PM$_{2.5}$ concentration decreased from 49.95 ± 14.04 μg/m$^3$ in 2019 to 39.03 ± 10.04 μg/m$^3$ in 2020.

Panel (c) of Figure S3 shows the spatial distribution of the change in PM$_{2.5}$ concentration between 2019 and 2020 during the same period (January 23–March 10). Fig. 1 shows the corresponding provincial averages. City lockdowns substantially decreased PM$_{2.5}$ concentration in the NCP and in central China (e.g., by 32.66 and 29.11 μg/m$^3$ in Henan and Hubei, respectively). Surprisingly, PM$_{2.5}$ concentration remained unchanged or even increased in polluted regions in northern China. In particular, PM$_{2.5}$ concentration increased from 59.69 ± 52.89 μg/m$^3$ in 2019 to 61.28 ± 56.86 μg/m$^3$ in 2020 in Beijing. It is important to understand the cause underlying this haze pollution, which persisted despite the strict restrictions on economic activity.

Meteorological factors could play critical roles in the formation of these air pollution events. Figure S4 shows the yearly variations of key meteorological factors, namely (a) wind speed, (b) temperature, and (c) relative humidity, in Beijing during January 23–March 10 for each year from 2017 to 2020. The temperature in 2020 was in the typical range, but wind speed has been showing a decreasing trend over the past few years. In contrast, relative humidity in Beijing was anomalously high during the COVID-19 outbreak period.

We now focus on the meteorological variation from 2019 to 2020. Figure S5 shows the spatial distribution of the change in wind speed, temperature, and relative humidity in China during January 23–March 10 in 2019 and 2020. The wind speed increased sharply in the NCP (e.g., by 0.40 m/s in Shandong) but decreased in the Beijing–Tianjin–Hebei area and northeastern China (e.g., by 22.83% and 23.37% in Beijing and Heilongjiang, respectively). The national average relative humidity increased from 65.07% ± 17.74% in 2019 to 67.13% ± 13.39% in 2020.

3.2. Effect of wind variation

To illustrate the decomposition method, we use Beijing as an example, following which we extend the method over the entire country. Panel (a) in Figure S6 shows the distribution of the PM$_{2.5}$ concentration as a function of wind speed and direction (i.e., the 2D pollution rose) in Beijing during January 23–March 10 of 2019 ($C_{W1}$) and 2020 ($C_{W2}$). PM$_{2.5}$ data were distributed into three bins for wind speed (0–3 m/s, 3–6 m/s, and >6 m/s) and into four bins for wind direction (0°–90° for northeasterly winds, 90°–180° for southeasterly winds, 180°–270° for northwesterly winds, and 270°–360° for northwesterly winds). High PM$_{2.5}$ concentrations were observed under weak or southerly wind conditions, suggesting the effects of local emissions and regional transport from the south of Beijing. Panel (b) shows the change in the pollution rose between 2019 and 2020 (i.e., $C_{W2} – C_{W1}$). PM$_{2.5}$ concentrations decreased under most wind conditions except weak northerly winds.

Similarly, panel (c) in Figure S6 shows the normalized frequency distribution of wind speed and wind direction (i.e., the 2D wind rose) in Beijing during January 23–March 10 of 2019 ($F_{W1}$) and 2020 ($F_{W2}$). Wind data were distributed into three bins for wind speed (i.e., 0–3 m/s, 3–6 m/s, and >6 m/s) and into four bins for wind direction (i.e., 0°–90° for northeasterly winds, 90°–180° for southeasterly winds, 180°–270° for southwesterly winds, and 270°–360° for northwesterly winds). During the study period, northerly and southeasterly winds were frequent. The average wind speed decreased from 3.15 ± 0.1 m/s in 2019 to 2.95 ± 0.2 m/s in 2020. Panel (d) shows the change in the wind rose between 2019 and 2020 (i.e., $F_{W2} – F_{W1}$). From 2019 to 2020, the frequencies of weak northwesterly winds and southeasterly winds increased, whereas those of moderate northerly and southwesterly winds decreased.

With this background, the effects of wind and non-wind factors on PM$_{2.5}$ variation can be identified as $ΔC_{W1} = C_{W1} – (F_{W2} – F_{W1})$ and $ΔC_{W2} = F_{W1} (C_{W2} – C_{W1})$, respectively. The decreased wind speed and the increased frequency of southeasterly wind increased the average PM$_{2.5}$ concentration in Beijing by 3.19 μg/m$^3$. On excluding the wind effect, the non-wind effect reduced PM$_{2.5}$ concentration by 4.46 μg/m$^3$. The unresolved PM$_{2.5}$ variation was 2.85 μg/m$^3$, with a linear index of 0.73. This could be a result of the small sample size and the non-linear interaction between the wind and non-wind effects.

We now extend the assessment of the effect of wind variation over all of China. Panels (a) and (b) in Figure S7 show the spatial distribution of the change in the PM$_{2.5}$ concentration resulting from the wind and non-wind effects at all stations in China. Panel (c) shows the linear index for the wind decomposition. Nationally, the wind effect increased the PM$_{2.5}$ concentration by 0.16 ± 0.99 μg/m$^3$, whereas the non-wind effect decreased the PM$_{2.5}$ concentration by 10.59 ± 6.05 μg/m$^3$. Low unresolved non-linear interactions were found at 164 stations, with a linear index of >0.87 (i.e., 90% confidence level). The national average linear index was 0.90 ± 0.11, indicating high confidence in the overall wind decomposition.

Fig. 2(a) presents the provincial average of the wind effect. The weakened winds substantially increased PM$_{2.5}$ concentration in the Beijing–Tianjin area and northeastern China, whereas the most substantial increases were in Beijing (by 3.19 μg/m$^3$) and Heilongjiang (by 3.65 μg/m$^3$). In contrast, the strengthened winds greatly decreased PM$_{2.5}$ concentration in the NCP (e.g., by 1.89 μg/m$^3$ in Shandong).
3.3. Effect of temperature variation

Panel (a) in Figure S8 shows the distribution of the PM$_{2.5}$ concentration as a function of temperature (i.e., 1D temperature-based pollution matrix) in Beijing during January 23–March 10 of 2019 ($C_{T1}$) and 2020 ($C_{T2}$). In general, low PM$_{2.5}$ concentrations were associated with low temperatures. Panel (b) presents the change in the temperature-based pollution matrix between 2019 and 2020 (i.e., $C_{T2} - C_{T1}$). PM$_{2.5}$ concentrations increased when the temperature ranged from −10°C to 0°C.

Next, the temperature variation in Beijing from 2019 to 2020 is identified. Panel (c) in Figure S8 shows the normalized frequency distribution of temperature (i.e., 1D temperature matrix) in Beijing during January 23–March 10 of 2019 ($F_{T1}$) and 2020 ($F_{T2}$). The average temperature in Beijing increased slightly from 1.13°C in 2019 to 1.17°C in 2020. In 2019, more than 40% of the temperature readings were in the −10°C to 0°C range, whereas in 2020, more than 50% of temperature readings were in the 0°C–10°C range. Panel (d) shows the change in the temperature matrix (i.e., $F_{T2} - F_{T1}$). From 2019 to 2020, the frequency of moderate temperature readings (0°C–10°C) increased, whereas that of low and high-temperature readings decreased.

With this background, the temperature and non-temperature effects on PM$_{2.5}$ concentration can be identified as $\Delta c_{T1} = c_{T1} - (F_{T2} - F_{T1})$ and $\Delta c_{T2} = c_{T2} - (F_{T2} - F_{T1})$, respectively. The increased frequency of moderate temperature readings slightly increased the average PM$_{2.5}$ concentration in Beijing by 0.59 μg/m$^3$. On excluding the temperature effect, the non-temperature effect still increased PM$_{2.5}$ concentration by 1.57 μg/m$^3$. The unresolved PM$_{2.5}$ variation was −0.58 μg/m$^3$ with a linear index of 0.79, indicating the effects such as the non-linear interaction between the temperature and non-temperature effects.

We then extend the assessment of the effect of temperature variation over all of China. Panels (a) and (b) in Figure S9 show the spatial distribution of the change in the PM$_{2.5}$ concentration resulting from the temperature and non-temperature effects at all stations in China. Panel (c) shows the linear index for the temperature decomposition. Nationally, the temperature effect increased the PM$_{2.5}$ concentration by 0.56 ± 1.19 μg/m$^3$, whereas the non-temperature effect decreased the PM$_{2.5}$ concentration by 10.56 ± 6.04 μg/m$^3$. Low unresolved non-linear interactions were found at 142 stations, with a linear index of >0.87 (i.e., 90% confidence level). The national average of the linear index was 0.88 ± 0.12, indicating high confidence in the temperature decomposition.

Fig. 2(b) presents the provincial average of the temperature effect. In particular, elevated temperatures sharply increased PM$_{2.5}$ concentrations in the NCP and in southern China (e.g., by 2.53 and 3.07 μg/m$^3$ in Shandong and Jiangxi, respectively). In contrast, decreased temperatures substantially decreased PM$_{2.5}$ concentrations in northeastern China (e.g., by 3.40 μg/m$^3$ in Heilongjiang). Moreover, elevated temperatures decreased PM$_{2.5}$ concentration in northwestern China as well (e.g., by 1.91 μg/m$^3$ in Shaanxi). In these regions, high PM$_{2.5}$ concentrations were associated with low temperature, which could be due to the low boundary layer height and intensive biomass burning and heating.

3.4. Effect of humidity variation

Panel (a) in Figure S10 shows the distribution of the PM$_{2.5}$ concentration as a function of relative humidity (i.e., 1D humidity-based pollution matrix) in Beijing during January 23–March 10 of 2019 ($C_{H1}$) and 2020 ($C_{H2}$). PM$_{2.5}$ concentration substantially increased as relative humidity increased. In 2019, the average PM$_{2.5}$ concentration exceeded 100 μg/m$^3$ when relative humidity was 60% or higher, whereas the average PM$_{2.5}$ concentration was around 30 μg/m$^3$ when the relative humidity was 20% or lower. Panel (b) shows the change in the humidity-based pollution matrix between 2019 and 2020 (i.e., $C_{H2} - C_{H1}$). PM$_{2.5}$ concentrations decreased under all humidity conditions.

Next, the humidity variation in Beijing from 2019 to 2020 is identified. Panel (c) in Figure S10 shows the normalized frequency distribution of relative humidity (i.e., 1D humidity matrix) in Beijing during January 23–March 10 of 2019 ($F_{H1}$) and 2020 ($F_{H2}$). The average relative humidity in Beijing greatly increased from 35.41% in 2019 to 58.24% in 2020. In 2019, more than 30% of the relative humidity readings were in the 20%–40% range. In 2020, the most frequent relative humidity readings was higher than 80%. Panel (d) shows the change in the humidity matrix between 2019 and 2020 (i.e., $F_{H2} - F_{H1}$). From 2019 to 2020, the frequency of high relative humidity (≥60%) greatly increased, whereas that of low relative humidity (<40%) decreased.

With this background, the humidity and non-humidity effects on PM$_{2.5}$ concentration can be identified as $\Delta c_{H1} = c_{H1} - (F_{H2} - F_{H1})$ and $\Delta c_{H2} = c_{H2} - (F_{H2} - F_{H1})$, respectively. The drastic increase in humidity greatly increased the PM$_{2.5}$ concentration in Beijing by 28.20 μg/m$^3$. On excluding the humidity effect, the non-humidity effect substantially reduced PM$_{2.5}$ concentration by 23.53 μg/m$^3$. The unresolved PM$_{2.5}$ variation was −3.09 μg/m$^3$ with a linear index of 0.94, indicating a 95% confidence level in the humidity decomposition.

We then extend the assessment of the effect of humidity variation over all of China. Panels (a) and (b) in Figure S11 show the spatial distribution of the change in the PM$_{2.5}$ concentration resulting from the humidity and non-humidity effects at all stations in China. Panel (c) shows the linear index for the humidity decomposition. Nationally, the
humidity effect increased PM$_{2.5}$ concentration by 3.40 ± 3.51 μg/m$^3$, whereas the non-humidity effect decreased PM$_{2.5}$ concentration by 13.44 ± 6.58 μg/m$^3$. Low unresolved non-linear interactions were found at 146 stations with a linear index of >0.87 (i.e., 90% confidence level). The national average of the linear index was 0.88 ± 0.12, indicating high confidence in the humidity decomposition.

Fig. 2(c) presents the provincial average of the humidity effect. In particular, the drastic increase in relative humidity substantially increased PM$_{2.5}$ concentrations in northern China (e.g., by 28.20 and 24.71 μg/m$^3$ in Beijing and Tianjin, respectively). On excluding the humidity effect, PM$_{2.5}$ concentrations greatly decreased in northern China (e.g., by 23.53, 28.56, and 26.70 μg/m$^3$ in Beijing, Tianjin, and Hebei, respectively). These results underscore the impact of humidity on PM$_{2.5}$ pollution in northern China.

3.5. Combined meteorological effect

Finally, the combined effect of wind, temperature, and relative humidity is investigated using the 4D decomposition method. In Beijing, between 2019 and 2020, the combined meteorological effect increased the PM$_{2.5}$ concentration by 26.79 μg/m$^3$ during January 23–March 10. On excluding the meteorological effect, the non-meteorological effects (e.g., restrictions on economic activities) substantially decreased PM$_{2.5}$ concentration by 21.84 μg/m$^3$. The unresolved PM$_{2.5}$ variation was −3.37 μg/m$^3$ with a linear index of 0.94, indicating a 95% confidence level in the 4D decomposition. These results indicate that the strict restrictions on human and economic activities indeed greatly decreased the PM$_{2.5}$ concentration in Beijing. However, the unfavorable meteorological conditions (e.g., the drastically increased humidity and the weakened airflow) substantially increased the PM$_{2.5}$ concentration in Beijing, which offset the effect of emission reduction and caused haze pollution. For comparison, the direct summation of the wind, temperature, and humidity effects increased PM$_{2.5}$ concentration by 31.98 μg/m$^3$, which is larger than the combined meteorological effect by 5.19 μg/m$^3$. This difference is due to the internal interactions between the meteorological factors.

We further extend the assessment of the combined meteorological effect over all of China. Panels (a) and (b) in Figure S12 show the spatial distribution of the change in the PM$_{2.5}$ concentration resulting from the meteorological and non-meteorological effects at all stations in China. Panel (c) shows the corresponding linear indexes for the meteorological and non-meteorological decomposition. Nationally, the combined meteorological effect increased the PM$_{2.5}$ concentration by 1.88 ± 3.11 μg/m$^3$, whereas non-meteorological effects decreased the PM$_{2.5}$ concentration by 11.74 ± 5.62 μg/m$^3$. Low unresolved non-linear interactions were found at 107 stations with a linear index of >0.87 (i.e., 90% confidence level). The national average of the linear index was 0.83 ± 0.14.

Fig. 3 shows the provincial averages of the meteorological and non-meteorological effects in China. The combined meteorological variation drastically increased the PM$_{2.5}$ concentration in northern China: the most substantial increases were in the Beijing–Tianjin–Hebei region (e.g., by 26.79, 22.21, and 17.52 μg/m$^3$ in Beijing, Tianjin, and Hebei, respectively). Excluding the meteorological effect, PM$_{2.5}$ concentrations greatly decreased in northern China (e.g., by 21.84, 23.40, and 21.90 μg/m$^3$ in Beijing, Tianjin, and Hebei, respectively). These results reveal that the strict restrictions on economic activities indeed decreased the PM$_{2.5}$ concentration in northern China. However, the unfavorable meteorological conditions, such as the high relative humidity and stagnant airflow, greatly contributed to the formation of severe haze pollution in northern China during the COVID-19 lockdown period.

The combined meteorological effect estimated using the 4D decomposition method differs from the direct summation of the wind, temperature, and humidity effects, as shown in Figure S13(a). The figure shows a similar spatial pattern to that obtained using the 4D decomposition model. Panel (b) shows the difference between the direct summation of the individual meteorological effects and the combined meteorological effect estimated using the 4D decomposition method. In most provinces, the direction summation was larger than the effect estimated using the 4D decomposition method, suggesting internal interactions between the meteorological effects. Large interactions were observed in northeastern China, such as in Heilongjiang, where the interactions contributed 13.24 μg/m$^3$ to the PM$_{2.5}$ variation. In these regions, the increased humidity could be related to the decreased temperature and stagnant airflow.

Fig. 4 shows the meteorological and non-meteorological effects on PM$_{2.5}$ variations in five key provinces in China: Beijing, Shanghai, Guangdong, Hubei, and Sichuan. The meteorological factors, particularly the elevated relative humidity, drastically increased the PM$_{2.5}$ concentration in Beijing. The combined meteorological effect decreased the PM$_{2.5}$ concentration by approximately 5 μg/m$^3$ in Hubei, the epicenter of COVID-19. The meteorological effects were small (within ±1 μg/m$^3$) in Shanghai, Guangdong, and Sichuan. The non-meteorological effect dominated the PM$_{2.5}$ variation in most regions, except in Beijing. Overall, these results suggest that the reduction in the PM$_{2.5}$ concentration in most regions over China can be attributed to the
resstrictions on human and economic activities. However, in certain regions, such as northern China, attributing PM$_{2.5}$ variation to the aforementioned control measures could be inaccurate if the effects of meteorological variations are not accounted for.

4. Discussion

In response to the COVID-19 pandemic, the Chinese government implemented various containment measures, including lockdown of its cities for extensive periods. These efforts to suppress the spread of COVID-19 in China drastically reduced human mobility and economic activity, leading to an unprecedented reduction in the emission of air pollutants (Huang et al., 2020; Le et al., 2020; Xing et al., 2020). However, extreme haze pollution episodes were reported, which confused the public and government. Particulate matter was the major air pollutant that contributed to the formation of the haze pollution, which went with a great reduction in visibility. Therefore, this study focused on PM$_{2.5}$ concentration to investigate the cause of the haze pollution.

Based on the wind-pollution decomposition method proposed by Li et al. (2014), this study outlined an improved decomposition method to differentiate the integrated meteorological and anthropogenic effects on air pollution, and applied the method to investigate the formation of haze pollution in China during the COVID-19 lockdown period. Our results show that the anthropogenic effect substantially decreased PM$_{2.5}$ concentrations in northern China, revealing that the extensive restrictions on human and economic activities (e.g., city lockdowns) indeed substantially decrease PM$_{2.5}$ concentrations in northern China, consistent with the substantial reduction in emissions across the country (Huang et al., 2020; Li et al., 2020; Xing et al., 2020). However, unfavorable meteorological factors, such as the elevated humidity and stagnant air conditions, greatly increased PM$_{2.5}$ concentrations, resulting in severe haze pollution episodes in northern China even during the lockdown period. The stagnant air mass trapped pollutants near the surface, whereas the increased humidity stimulated multiphase reactions for secondary aerosol formation. This direct observational evidence is in line with results obtained using complicated model simulations (Le et al., 2020).

The government interventions in China during the COVID-19 lockdown period were unprecedented. The huge emission reductions notwithstanding the adverse meteorological conditions resulted in haze pollution. Overall, the results of this study highlight the impact of meteorological factors on air pollution, which is strong enough to overwhelm the beneficial effects of emission reduction. In other words, without taking the effects of meteorological variation into account, attributing air quality change to control measures could be misleading.

A linear index $L$ was used to demonstrate the efficacy of the decomposition. Li et al. (2014) used the Monte Carlo simulations to examine confidence levels that were associated with different $L$ values. 100 different sets of 40,000 numbers were randomly generated. Each set of 40,000 numbers had a different mean value and standard deviation. From each dataset, 10,000 $L$ indices were calculated, which were then used to define a cumulative probability density distribution. It was found that the cumulative probability density distributions for all 100 sets of Monte Carlo simulations were almost identical, with difference for any value $<0.005$. Meanwhile, 1%, 5%, and 10% of the generated $L$ indices had a value $>0.98$, $>0.93$, and $>0.87$, respectively. These Monte Carlo simulations indicated that the $p$ values were 0.01, 0.05, and 0.10 (corresponding confidence levels were 99%, 95%, and 90%) when the $L$ index was larger than 0.98, 0.93, and 0.87, respectively.

A high linear index indicates a low unresolved change in PM$_{2.5}$ concentration, which can be caused by the non-linear interaction between meteorological and non-meteorological factors. In this national study, average linear indices were $0.90 \pm 0.11$, $0.88 \pm 0.12$, $0.88 \pm 0.12$, and $0.83 \pm 0.14$ for the decompositions of effects of wind, temperature, relative humidity, and combined meteorological effect, respectively. Meanwhile, high confidence levels with a linear index of $-0.87$ (i.e., 90% confidence level) were found at 164, 142, 146, and 107 stations for the four decompositions. These results indicate good confidence levels in the overall meteorological decompositions.

Uncertainties of the meteorological values and PM$_{2.5}$ concentration can be transferred to the decomposition results. According to the World Meteorological Organization, the required precision of minute measurements of temperature, relative humidity, and wind were $\pm 0.1 \, ^{\circ}\mathrm{C}$, $\pm 3\%$, and $\pm 0.5 \, \text{m/s}$, respectively. Uncertainties of hourly measurements should be much lower than these values. Meanwhile, precision of hourly measurement of PM$_{2.5}$ concentration using the tapered element oscillating microbalance (TEOM) technique or beta attenuation monitors (BAM) was around $\pm 2 \, \mu\text{g/m}^3$. We applied the Monte Carlo simulations to explore how the uncertainties of meteorological and PM$_{2.5}$ data were transferred to the decomposition results. We used Beijing as the study region. In 1000 tests, we added random fluctuations on the hourly temperature, relative humidity, wind, and PM$_{2.5}$ with a standard deviation of $\pm 0.1 \, ^{\circ}\mathrm{C}$, $\pm 3\%$, $\pm 0.5 \, \text{m/s}$, and $\pm 2 \, \mu\text{g/m}^3$, respectively. The 4-D decomposition results show that the combined meteorological effect increased PM$_{2.5}$ concentration by 26.31 $\pm$ 1.74 $\mu\text{g/m}^3$, with an absolute percentage deviation of 6.6%. Meanwhile, anthropogenic effect reduced PM$_{2.5}$ concentration by 21.93 $\pm$ 1.03 $\mu\text{g/m}^3$, with an absolute percentage deviation of 4.7%. These results indicate that the decomposition uncertainty resulted from the meteorological values and PM$_{2.5}$ concentration was well within 10%.

Our decomposition method has several strengths. First, the decomposition method is based only on the ground measurements of meteorological factors and air pollutant concentrations. Thus, this approach does not require complicated model simulations. The decomposition method can be applied in any region as long as the required data are available. Second, a linear index is introduced to demonstrate the validity of the linear decomposition. A high linear index indicates low interaction between the influence factors (e.g., meteorological factors and emission controls). The linear index can help assign confidence levels to the decomposition results and minimize bias arising from the interaction between the influence factors.

The lockdown affected not only PM$_{2.5}$ but also various gaseous pollutants, such as nitrogen dioxide (NO$_2$), sulfur dioxide (SO$_2$), carbon monoxide (CO), and ozone (O$_3$). Figure S14 shows spatial distributions of the changes in (a) NO$_2$, (b) SO$_2$, (c) CO, and (d) O$_3$ concentrations at all stations in China during January 23–March 10 between 2019 and 2020. In Beijing, NO$_2$, SO$_2$, CO, and O$_3$ concentrations changed by $-6.4$ ppb, $-0.83$ ppb, 0.025 ppm, and 1.64 ppb, respectively. In overall, NO$_2$, SO$_2$, and CO concentrations extensively declined over China,
particularly in the North China Plain. The increases in ozone concentrations could be caused by the reduction of nitrogen oxide due to the non-linear ozone chemistry.

The formation of gaseous pollutions (e.g., NO₂, SO₂, CO, and O₃) were also determined by an interaction of meteorological factors and the control of human activity. Contributions of meteorological and anthropogenic effects on the formation of gaseous pollution could greatly differ from PM₂.₅ pollution. Future studies can use our decomposition method to examine the effects of different influence factors on other air pollutants in China during the COVID-19 lockdown period. In addition, most countries have implemented a series of large-scale emergent interventions to contain the ongoing pandemic. The observational decomposition method outlined in this study can be applied in other countries to track the corresponding air quality response to different influence factors.

By design, this study focused on the effects of three key meteorological factors: wind, temperature, and relative humidity. The combined meteorological effect was investigated using a 4D decomposition scheme. Our model is highly flexible and can be extended to a higher dimension to include the effects of more meteorological factors. Future studies could consider the effects of other meteorological factors, such as pressure, cloud cover, and precipitation, to strengthen the estimation of the overall meteorological effect. Other indicators of moisture include absolute humidity, dew point, and mixing ratio. In this study, we used relative humidity to represent the moisture in the air.

Mixing layer height is suggested to play an important role in the formation of severe air pollution episodes. Significant association between the surge of surface air pollutant concentrations and a shallow mixing layer height has been frequently documented (Miao et al., 2019; Su et al., 2020). Wang et al. (2019) studied the evolution of a severe haze episode in central-east China using radiosonde measurements of upper-air meteorology, and concluded that temperature inversion within boundary layer was the most important trigger factor for the haze. However, detailed information of mixing layer height over a large region is much more limited than common meteorological values applied in this study. Radiosondes are regularly launched at 8:00 a.m. and 8:00 p.m. every day at a limited number of stations in China. Aerosol LIDAR system is capable of detecting the mixing layer height at a high temporal resolution (Yang et al., 2013). However, operation of aerosol LIDAR is expensive and sparse. In numerical prediction models, the mixing layer height is characterized by parameterization schemes, and is an important yet difficult parameter to be accurately simulated (Xie et al., 2012). In addition, the mixing layer height is largely governed by other meteorological variables. Therefore, there could be an interaction and overlap effect between the mixing layer height and other meteorological values. In this study, we relied on ground measurements of several common meteorological parameters (e.g., temperature, humidity, and wind) to assess the meteorological effects over entire China. The lack of mixing layer height data over a large domain is one of the limitations of our study. Future assessments can directly consider the impact of the mixing layer height if reliable measurements are available over a large region.

Most of previous studies tested their models in a small region (Plocost et al., 2019). In the present study, we tested the method for different regions in China. Given a vast territory, China has great spatial variance in meteorological conditions, land-use patterns, and emission scenarios. Average temperature during the COVID-19 lockdown period ranged from 10.4 °C to 23.5 °C. Therefore, the climates in China were diverse, covering both warm and cold weather. Meanwhile, our national study covered diverse land-use patterns, such as highly populated city clusters in eastern China (e.g., the Beijing-Tianjin-Hebei, Yangtze River Delta, and Pearl River Delta region) and sparsely populated areas in western China. Results from the national study provide a clue for us to understand the good ability of our decomposition method in various meteorological and land-use conditions.

Simulation-based methods for differentiating meteorological and anthropogenic effects require a detailed investigation of emission inventory. Future studies can also investigate the causes underlying air pollution in China during the COVID-19 lockdown period by using complex modeling systems coupled with reliable emission inventory data. Results from observation- and simulation-based analyses can be compared and integrated to generate a more comprehensive report of the effects of government interventions and meteorological factors on the formation of the aforementioned haze pollution episodes.

5. Conclusion

Based on a wind decomposition method, this study outlined an improved method to differentiate complex meteorological and anthropogenic effects. The improved method was then applied to investigate the cause of unanticipated haze pollution in China during the COVID-19 lockdown period. Results from the wind decomposition method show that weakened winds increased PM₂.₅ concentration by 3.19 μg/m³ in Beijing. However, after the effect of other meteorological factors were taken into account, the combined meteorological effect (e.g., drastically elevated humidity levels and weakened airflow) substantially increased PM₂.₅ concentration by 26.79 μg/m³ in Beijing. On excluding the integrated meteorological effects, anthropogenic effect substantially decreased PM₂.₅ concentrations by 21.84 μg/m³ in Beijing, evidencing that the strict restrictions on human activities indeed decreased PM₂.₅ concentrations. A linear index L was used to demonstrate the efficacy of the decomposition. In this national study, the average linear indices were 0.90 ± 0.11, 0.88 ± 0.12, 0.88 ± 0.12, and 0.83 ± 0.14 for the decompositions of effects of wind, temperature, relative humidity, and combined meteorological effect, respectively. High confidence levels with a linear index of >0.87 (i.e., 90% confidence level) were found at 164, 142, 146, and 107 stations for the four decompositions.

CRediT authorship contribution statement

Yushan Song: Analysis and draft. Changqing Lin: Analysis and draft. Ying Li: Analysis and draft. Alexis K.H. Lau: Supervision. Jimmy C.H. Fung: Review and editing. Xingcheng Lu: Review and editing. Cui Guo: Review and editing. Xiang Qian Lao: Review and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This work was supported by the Research Grants Council of Hong Kong (Project No. GRF 16202120 and T24/504/17), and NSFC/RGC (Grant No. N.HKUST638/19). We thank the Institute for the Environment (IENV) and Environmental Central Facility (ENVF) of Hong Kong University of Science and Technology (HKUST) for providing atmospheric and environmental data. The authors declare that they have no actual or potential competing financial interests.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2021.118270.

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