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Variation of pollution sources and health effects on air pollution before and during COVID-19 pandemic in Linfen, Fenwei Plain

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ABSTRACT

Stringent pollution control measures are generally applied to improve air quality, especially in the Spring Festival in China. Meanwhile, human activities are reduced significantly due to nationwide lockdown measures to curtail the COVID-19 spreading in 2020. Herein, to better understand the influence of control measures and meteorology on air pollution, this study compared the variation of pollution source and their health risk during the 2019 and 2020 Spring Festival in Linfen, China. Results revealed that the average concentration of PM$_{2.5}$ in 2020 decreased by 39.0% when compared to the 2019 Spring Festival. Organic carbon (OC) and SO$_4^{2-}$ were the primary contributor to PM$_{2.5}$ with the value of 19.5% (21.1%) and 23.5% (25.5%) in 2019 (2020) Spring Festival, respectively. Based on the positive matrix factorization (PMF) model, six pollution sources of PM$_{2.5}$ were indicated. Vehicle emissions (VE) had the maximum reduction in pollution source concentration (28.39 µg·m$^{-3}$), followed by dust fall (DF) (11.47 µg·m$^{-3}$), firework burning (FB) (10.39 µg·m$^{-3}$), coal combustion (CC) (8.54 µg·m$^{-3}$), and secondary inorganic aerosol (SIA) (3.95 µg·m$^{-3}$). However, the apportionment concentration of biomass burning (BB) increased by 78.7%, indicating a significant increase in biomass combustion under control measures. PAHs-lifetime lung cancer risk (ILCR) of VE, CC, FB, BB, and DF, decreased by 44.6%, 43.2%, 34.1%, 21.3%, and 2.0%, respectively. Additionally, the average contribution of meteorological conditions on PM$_{2.5}$ in 2020 increased by 20.21% compared to 2019 Spring Festival, demonstrating that meteorological conditions played a crucial role in located air pollution. This study revealed that the existing control measures in Linfen were efficient to reduce air pollution and health risk, whereas more BB emissions were worthy of further attention. Furthermore, the result was conducive to developing more effective control measures and putting more attention into unfavorable meteorological conditions in Linfen.

1. Introduction

Atmospheric particulate matter (PM) pollution is one of the most significant environmental issues because it can influence human health and environmental climate (Cao et al., 2021). Due to the frequent occurrence of haze episodes, three key regions for the “Blue Sky Protection Campaign” in China are designated, including the Beijing-Tianjin-Hebei (BTH) region, the Fenwei Plain, and the Yangtze River Delta (YRD) region (Cao and Cui, 2021; Li et al., 2021; Zhao et al., 2021b). Especially, air pollution during Spring Festival is generally concerned due to the strong human activities (e.g., firework burning) (Dai et al., 2021b; Lorenzo et al., 2021). Yao et al. (2019) demonstrated that PM$_{2.5}$ concentration presented a reducing trend in Shanghai during Spring Festival, indicating the high efficiency of regulatory measures. Meanwhile, Zheng et al. (2019) revealed that long-term pollution control measures were performed to improve air quality in Nanjing. However, there are differences in the formation mechanism of haze between the YRD region and North China due to differences in energy structure (Hong et al., 2021; Wang et al., 2021a). Therefore, whether control policies are effective to decrease air pollution is worth being further investigated in North China.

In early 2020, a novel coronavirus disease (COVID-19) spread...
rapidly in China, resulting in a significant reduction in human activities (restriction measures) (Du et al., 2021; Wang et al., 2022; Zheng et al., 2020). Air quality was noteworthy improved across the country (Du et al., 2021). Correspondingly, Hong et al. (2021) revealed that the reduction effects of pollution emissions may be counteracted by unfavorable geographical and meteorological conditions in the BTH region. Liu et al. (2022) also showed PM$_2.5$ concentration may be increased by 8.8% in Hubei province under unfavorable meteorological conditions. Consequently, it is necessary to investigate the variation of pollution sources before and after COVID-19 epidemic and the influence of meteorological conditions on air pollution under emission reduction.

It is crucial to characterize sources of atmospheric PM$_2.5$ by chemistry species, which can provide more significant information for local air pollution control (Li et al., 2020; Zheng et al., 2019). Based on the positive matrix factorization (PMF) model, water soluble ions (WSIs), carbon components (organic carbon (OC) and elemental carbon (EC)), and polycyclic aromatic hydrocarbons (PAHs) are generally used in pollution source apportionment of PM$_2.5$ (Ali-Taleshi et al., 2021; Cheng et al., 2021; Hong et al., 2021; Kong et al., 2018; Zheng et al., 2020). Linfen is a heavy industrial base of Shanxi Province and is located in the north of Fenwei Plain. In recent years, the air pollution of Linfen is gradually concerned (Li et al., 2020; Liu et al., 2021a; Wang et al., 2022). Meanwhile, more control measures were formulated to improve local air quality, such as “limited firework”, “limited coal combustion”, and “Burning gas instead of coal” (http://sthjj.linfen.gov.cn/). However, the information is still lacking about the quantitative variation of main pollution sources under control measures during different periods. And investigating the chemistry species concentration and risks is also instructive for the future policymaking of Linfen (Kong et al., 2018).

Herein, to better understand the influence of control measures and meteorology on air pollution, we collect the PM$_2.5$ samples during the 2019 and 2020 Spring Festival and analyze the chemistry species concentration (WSIs, OC/EC, and PAHs). PMF model is applied to identify the main pollution sources. And regional transport and contribution of secondary aerosol (such as SO$_2^-$ and NO$_3^-$) depends on the level of gaseous precursors pollutants (SO$_2$ and NO$_2$). The conversion efficiency of SO$_2$ and NO$_2$ were quantitatively described by SOR and NOR

$$\text{SOR} = \frac{\text{SO}_2^-}{\text{SO}_2^- + \text{SO}_2} \quad (1)$$

$$\text{NOR} = \frac{\text{NO}_3^-}{\text{NO}_3^- + \text{NO}_2} \quad (2)$$

It is generally confirmed that the critical value of SOR and NOR is 0.1, and the high SOR and NOR level indicate that the secondary transformation processes are dominant in the formation of secondary aerosols (Zhao et al., 2020). The balance of anions and cations by using ion balance calculations was investigated for further understanding the heterogeneous reactions and toxicity of the atmospheric particle. The calculation included the anion ion equivalent (AE) and cation ion equivalent (CE) ratios, which were calculated by the following formula:

$$\text{AE} = \frac{[\text{Cl}^-] \times 35.5 + [\text{SO}_4^{2-}] \times 84 + [\text{NO}_3^-] \times 62}{(3)}$$

$$\text{CE} = \frac{[\text{NH}_4^+] \times 18 + [\text{Na}^+] \times 23 + [\text{Ca}^{2+}] \times 20 + [\text{K}^+] \times 39}{(4)}$$

where [Cl$^-$], [SO$_4^{2-}$], [NO$_3^-$], [NH$_4^+$], [Na$^+$], [Ca$^{2+}$], and [K$^+$] was the concentration of seven WSIs (unit: $\mu$g m$^{-3}$).
2.5. PMF model

As confirmed by previous studies, Positive Matrix Factorization (PMF) model is generally applied to PM2.5 source apportionment (Dai et al., 2021a; Kong et al., 2018). Based on factor analysis, PMF model decomposes a matrix of speciated sample data into two matrices: factor contributions (G) and factor profiles (F). In this study, PMF model (5.0) of the US EPA (Environmental protection agency) was used to source apportionment. More detailed PMF running explanation was shown in Supplementary materials (Text S3 and Table S4-S6).

2.6. Backward trajectory, CWT analysis and de-weathered model

The HYPLIT trajectory model was used to calculate air mass trajectory and concentration-weighted trajectory (CWT) (Wang et al., 2009). Meanwhile, meteorological data was downloaded from the Global Data Assimilation System (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1). The model was run with a starting height of 500 m above the ground level. And the clustering analysis of air mass trajectory was based on air mass space resemblance to track all the combinations. Based on source regions of all air mass trajectory, a grid layer (0.5° × 0.5°) was created for CWT analysis. CWT can calculate the average weight concentration of each grid to provide a relative contribution of each trajectory. More detailed information was shown in Supplementary materials (Text S4).

Air pollution is usually influenced by pollution emissions and meteorological conditions. To compare to the variation of emissions and meteorological conditions during the 2019 and 2020 Spring Festival, we used an R package “normalweather” to isolate the contribution of meteorological conditions (Chen et al., 2018; Dai et al., 2021b). The package built the random forest model based on the variables and pollutant concentrations. Variables included meteorological parameters and time variables. More detailed information of model was shown in Text S5. And meteorological parameters were obtained by an R package “worldMet” (https://github.com/daviddarrawl/worldmet). In general, the contribution of meteorology is the difference between the observations and de-weather values (Zheng et al., 2020). To capture the contribution change of emission reduction and meteorology conditions, we used the followed formula (Liu et al., 2022; Shi et al., 2021; Vu et al., 2019):

$$E_r = \frac{C_{2020} - C_{2019}}{C_{2019}} \times 100\% \quad (5)$$

$$M_{w} = \frac{(C_{2019}^{w} - C_{2020}^{w}) - (C_{2019} - C_{2020})}{C_{2019} \times 100\%} \quad (6)$$

where \(E_r\) and \(M_{w}\) were the contribution change of emission reduction and meteorology conditions; \(C_{2019}^{w}\) and \(C_{2020}^{w}\) were the observed concentration of air pollution in 2019 and 2020; \(C_{2019}\) and \(C_{2020}\) represent the de-weather concentration of air pollution in 2019 and 2020.

2.7. Health risk assessment

Incremental lifetime cancer risk (ILCR) was used to express potential health risks of 16 USEPA priority PAHs via inhalational exposure (Hu et al., 2020), and calculated by followed formula:

$$\text{ILCR} = \text{UR}_{\text{BaP}} \times \sum \text{BaP}_{\text{eq}} \quad (7)$$

where \(\text{UR}_{\text{BaP}}\) is the unit inhalation cancer risk (8.7 × 10⁻⁵ (ng m⁻³)⁻¹) (Kong et al., 2018; Zhang et al., 2019), and \(\sum \text{BaP}_{\text{eq}}\) is the BaP equivalent concentration. \(\sum \text{BaP}_{\text{eq}}\) was calculated by the product of 16 PAHs concentration and their toxic equivalent factors (TEF) (as shown in Table S7).

3. Result and discussion

3.1. Air quality variation during the Spring Festival from 2019 to 2020

3.1.1. p.m.2.5, WSIs and carbon components

The concentration of PM2.5, seven major WSIs (SO²⁻, Cl⁻, NO₃⁻, Na⁺, Ca²⁺, K⁺, and NH₄⁺) and carbon components (OC and EC) in all samples during the 2019 and 2020 Spring Festival were presented in Fig S3. The mean values of PM2.5 during the 2019 and 2020 Spring Festival were 199.00 ± 96.48 μg m⁻³ and 121.36 ± 70.79 μg m⁻³, respectively, with a reduce rate of 39.0%. The increased pattern of WSIs (SO²⁻ and K⁺) was still evident due to fireworks burning on the Chinese New Year Eve. Significantly, SO²⁻ had the highest proportion among all WSIs in this study, while the nitrate wan dominant during the haze periods in previous studies in Table S9, indicating that the formation mechanisms of haze in Linfen may differ from other cities and Linfen was characterized by heavy industry (Cheng et al., 2021; Fu et al., 2020). During the Spring Festival, the average OC and EC concentrations were 38.78 ± 20.88 μg m⁻³ and 8.27 ± 4.86 μg m⁻³ in 2019, 39.94 ± 16.29 μg m⁻³ and 6.00 ± 2.95 μg m⁻³ in 2020, respectively. When compared to the average in 2019, the mean values of Ca²⁺, K⁺, SO²⁻, Cl⁻, NO₃⁻, and EC in 2020 decreased by 91.1%, 81.3%, 38.3%, 68.8%, 44.4%, and 27.4%, respectively, while Na⁺, NH₄⁺, and OC concentration increased by 15.5%, 4.7%, and 3.0%, respectively. As shown in Fig. 1, the significant decrement of K⁺, SO²⁻, Cl⁻, and NO₃⁻ revealed the enhanced control measures during the Spring Festival. Overall, the decreased tendency of 2020 may be mainly caused by the influence of control measures during Spring Festival and lockdown polities during the COVID-19 periods (Zheng et al., 2020).

In general, K⁺ is used as a powerful indicator of fireworks (Yao et al., 2019). The K⁺ concentration increased rapidly during Fireworks periods, revealing the great contribution of fireworks to K⁺. As shown in Fig. S4, the variation trend (p < 0.05) of K⁺ in 2020 was different from that in 2019. And the average concentration of K⁺ in fireworks periods in 2019 and 2020 increased by 14.0 times and 11.3 times, respectively, when compared to the pre-fireworks periods. Whereas the average contents of K⁺ in post-fireworks periods decreased by factors of 7.2 (2019) and 15.6 (2020), when compared to the fireworks periods. It indicated that the more active firework in firework periods of 2019, while the less firework burning in post-firework periods of 2020.

As stated earlier, the Linfen government strengthened the implementation to limit the use of fireworks during the winter of 2019 (http://www.linfen.gov.cn/contents/2588/482361.html). Noteworthily, there was still higher K⁺ (4.00 ± 3.68 μg m⁻³) observed in this study during the 2020 Spring Festival (Table S8) compared to other previous studies. We inferred that there were two possible reasons. Firstly, although fireworks were prohibited in urban areas of Linfen, fire still occurred in suburban areas. It was unavoidable that K⁺ would be transported from suburban areas to urban areas (Yao et al., 2019). Furthermore, a policy of “Burning natural gas instead of coal” (BGIC) was implemented to improve air quality in Linfen during the winter of 2019 (http://sthj.linfen.gov.cn/contents/2588/482361.html). Some studies revealed that PM2.5 concentration decreased significantly after coal-gas replacement (Xie et al., 2020). But this may increase the more biomass combustion for heating and cooking due to the prohibition of coal combustion.

3.1.2. The difference of PAHs levels

The total concentration of ∑16 PAHs during Spring Festival ranged from 20.55 ng m⁻³ to 809.14 ng m⁻³ in 2019, 21.71 ng m⁻³ to 237.42 ng m⁻³ in 2020, respectively. The average concentrations of ∑16 PAHs were 134.81 ± 86.04 ng m⁻³ (2019) and 73.06 ± 39.55 ng m⁻³ (2020), respectively. It was significantly higher than the average value of ∑16 PAHs in other cities during the winter, such as Tehran (49.8 ± 33.0 ng m⁻³) (Ishtiaq et al., 2021), Lahore (2340 ± 408 pg m⁻³) (Xie et al., 2021), Zhengzhou (53 ± 19 ng m⁻³) (Luo et al., 2021),
Huanggang (15.5 ± 6.86 ng·m⁻³) (Xu et al., 2021a), Shanghai (30.02 ± 12.25 ng·m⁻³) (Jia et al., 2021), Hangzhou (38.84 ± 19.09 ng·m⁻³) (Cao et al., 2021), which was mainly attributed to less coal consumption for heating in these cities. However, the mean concentration of PAHs in this study was similar to many studies of northern cities in China, such as Harbin (216 ± 27.6 ng·m⁻³) and Jiamusi (139 ± 14.4 ng·m⁻³) (Gao et al., 2019). It may be related to many activities associated with the coal combustion of northern cities, such as heating, cooking, and heavy industries. Simultaneously, He et al. (2021) also showed that the higher contribution of coal-burning and biomass burning to PAHs in outdoor. The total concentration of ∑16 PAHs decreased by about 45.81% in Fig. 2. And the values of PAHs in 2019 differed significantly (p < 0.05) from those in 2020. Meanwhile, the average concentration of 2-ring, 3-ring, 4-ring, 5-ring, and 6-ring PAHs reduced by 0.11 ng·m⁻³, 17.12 ng·m⁻³, 34.38 ng·m⁻³, 8.54 ng·m⁻³, and 1.59 ng·m⁻³, respectively. Middle-PAHs (MPAHs, 4-rings) and Low-PAHs (LPAHs, 2-rings and 3-rings) dropped by 42.9% and 86.6%, respectively. LPAHs was usually associated with non-combusted petroleum emission (e.g., vehicle emissions) (He et al., 2014; Jia et al., 2021), inferring higher traffic restrictions in 2020.

3.2. Chemical characterization

3.2.1. Transformation mechanism from gaseous to inorganic components

In this study, the NOR mean value (0.49 ± 0.19) was higher than the SOR average (0.46 ± 0.24) in 2019, indicating that the secondary formation of NO₃⁻ was stronger than the SO₄²⁻. While the SOR (0.38 ± 0.16) was higher than the NOR (0.29 ± 0.12) in 2020, revealing the higher secondary formation of SO₄²⁻. Furthermore, relative humidity (RH) only presented the significant positive correlation with NOR (r = 0.368, p < 0.05) in 2019, while RH showed the positive correlation with SOR (r = 0.396, p < 0.05) and NOR (r = 0.406, p < 0.001) in 2020, as shown in Fig. 3. And NOR showed the significant correlation with temperature (T) (r = 0.354, p < 0.05) in 2020. This result indicated that the secondary formation of NO₃⁻ was related to the homogenous/heterogeneous reaction of their gaseous precursor in 2020, while the conversion of NO₂ to NO₃ was only connected with heterogeneous oxidation in 2019 (Guo et al., 2020; Ma et al., 2021).

3.2.2. Acidity of particles and chemical forms of WSIs

As shown in Fig. S5, the significant correlation between the cations and anions equivalents in the 2019 and 2020 studying periods was presented (2019: R² = 0.992; 2020: R² = 0.885). Generally, PM₂.₅ is acidic when the linear regression slope of fitting curve between CE and AE is lower than 1; on the contrary, the particles are neutral or alkaline (Khan et al., 2021). For 2019 sampling periods, cations were insufficient for anion neutralization (r = 0.895x-0.022), revealing that the particle may be acidic. However, the linear regression slope of fitting curve was 1.085 during studying periods in 2020, demonstrating that the cations were enough and therefore the particle was neutral. NH₄ reacts preferentially with H₂SO₄ to cause the formation of NH₄HSO₄ or (NH₄)₂SO₄ (Zhou et al., 2018). In Fig. S5, the linear regression slopes of [NH₄⁺] and [SO₄²⁻] were 0.724 (R² = 0.726, 2019) and 0.949 (R² = 0.956, 2020). The result demonstrated that NH₄⁺ was rich to react with HNO₃ and form NH₄NO₃ in 2020 periods, while NH₃ was insufficient in 2019 and nitrate could have other formations (KNO₃ or Ca(NO₃)₂). Combined
with the mass ratios of AE/CE, the PM$_{2.5}$ was acidic probably due to the excess NO$_3$ in the 2019 periods.

The significant correlation of WSIs indicates the similarity of their sources (Zhan et al., 2021). NH$_4$ was also obviously correlated with NO$_3$ and SO$_4^{2-}$ (Fig S6), revealing that (NH$_4$)$_2$SO$_4$, NH$_4$HSO$_4$, and NH$_2$NO$_3$ still were dominating form of existence. Furthermore, there was a good correlation between K$^+$, SO$_4^{2-}$ and Cl$^-$, with the correlation coefficient greater than 0.7 in 2019 and 2020 periods. This was attributed to the firework burning. Potassium perchlorate often was used as an oxidizer in fireworks, with the corresponding chemical reactions being KClO$_4 \rightarrow$ KCl + 2O$_2$ (Pang et al., 2021; Wu et al., 2018). The higher ratios of NO$_2$/SO$_4^{2-}$ can reflect the predominance of mobile sources of PM$_{2.5}$ pollution. On the contrary, the lower mass ratios indicate the importance of stationary sources (He et al., 2017b). The ratios of NO$_3$/SO$_4^{2-}$ in 2019 averaged at 0.743, with a range of 0.119–1.773 as shown in Fig S7. However, the ratios of NO$_3$/SO$_4^{2-}$ in 2020 averaged at 0.632, with a range of 0.286–1.263. The mass ratios of NO$_3$/SO$_4^{2-}$ gradually decreased from 2019 to 2020, indicating that the gradual decrease of mobile sources emissions and the gradual increase of combustion sources emissions.

### 3.3. Source apportionment in different periods

#### 3.3.1. Sources identification and interpretation of PM$_{2.5}$

PMF model was used to better evaluate the variation in pollution sources between the two haze events (2019–2020). Separately, 2–8 factors were examined, and the six-factor solution was proved to be the best fit.

The factor profiles of six pollution sources were presented in Fig. 4. The first factor was identified as the coal combustion with high loading of Cl$^-$, OC, EC, MPAHs, and High PAHs (HPAHs). The previous studies found that Cl$^-$ of the fine particle was a tracer of coal combustion (Xu et al., 2021b). Additionally, coal combustion (industrial and residential) was a major contributor to OC during the winter (Xu et al., 2021b; Zheng et al., 2019). But OC and EC were generally considered to be emissions from vehicles (Liu et al., 2020a). Hence, there has been a bitter controversy over this. Meanwhile, Liu et al. (2021b) investigated the carbonaceous components of Taiyuan and found that in summer, OC and EC were primarily affected by motor vehicle emissions, while in winter (December to March) they were generally influenced by coal-fired emissions. The coal consumption in Linfen as the main industrial base of Shanxi Province, was approximately 43 million tons in 2018 (Li et al., 2020). As a result, F1 was associated with coal combustion (CC). Generally, Phe was identified as the tracer of biomass burning, and the high emission ratios of Flu, Pyr, Chr, BbF, and BkF were found to be due to wood burning (Kong et al., 2018; Zhang et al., 2019). Therefore, F2 was identified as a biomass burning source (BB). Ga$^{2+}$ and Na$^+$, which were typically associated with crustal dust, were characteristic of F3 (Cheng et al., 2021; Guo et al., 2020; Liu et al., 2020a). Hence, it was confirmed to be the dust fall source (DF). The dominating species of F4 were NH$_4$+, SO$_4^{2-}$, and NO$_3$–, so this factor was confirmed as secondary inorganic aerosol (SIA). F5 was dominated by NO$_3$ or HPAHs (DaA, BghiP, and IcdP). PM$_{2.5}$-related NO$_3$ was mainly generated by the oxidation of nitrogen oxides (NOx). And NOx emissions are mainly affected by vehicle exhaust (Zhan et al., 2021; Zhao et al., 2020). Similarly, IcdP, DahA, and BghiP originated from gasoline emissions as well (Xu et al., 2021a; Zhang et al., 2019). Thus, F5 was represented as...
vehicle emissions (VE). The growth rate and concentration of $K^+$ were maximized during the Chinese New Year Eve. Since F6 was primarily characterized by $K^+$, it was identified as a firework burning (FB) emission.

Overall, all six pollution sources were identified as CC, BB, DF, SIA, VE, and FB emissions, accounting for 20.5%, 3.8%, 9.4%, 29.8%, 22.7%, and 13.8% in 2019, 22.5%, 9.7%, 4.1%, 39.8%, 9.4%, and 14.5% in 2020, respectively.

3.3.2. Source identification of PAHs

Sources of PAHs was identified by diagnostic ratios. Meanwhile, the diagnostic ratios values and source were shown in Table S10. Four ratios of individual PAHs including Fla/(Fla + Pyr), BaA/(BaA + Chr), Ant/(Ant + Phe) and InP/(InP + BghiP) were presented in this study, as shown in Fig. 5 (Hu et al., 2020; Kong et al., 2018; Xu et al., 2021a). The values of Fla/(Fla + Pyr) and InP/(InP + BghiP) ranged from 0.48 to 0.93 and 0.39–0.53 in 2019, 0.48–0.55 and 0.45–0.54 in 2020, respectively, indicating the obvious contribution of biomass and coal combustion. Similarly, the values of BaA/(BaA + Chr) distributed principally within 0.37–0.80 in 2019 and 0.39–0.63 in 2020, suggesting the importance of non-traffic emissions. Nonetheless, there was no obvious difference in pollution sources between 2019 and 2020. Thus, the contribution of pollution sources was worthy of being further assessed.

The pollution source of PM$_{2.5}$-PAHs was mainly affected by primary emissions. Therefore, PMF model was separately run to accurately identify the PAHs source. As shown in Fig S9, the factor profiles of five pollution sources were presented during 2019 and 2020 Spring Festival. Meanwhile, the pollution source (CC, BB, DF, VE, and FB) correlated well between the two runs ($R^2$ = 0.40–0.70) in Table S11, suggesting the robustness of factors solution (Feng et al., 2022). This result would be helpful for comparison of health risks of PAHs.

3.3.3. Variation of pollution sources during different periods

Compared with the contributions of six pollution source in 2019, the average concentration of source decreased obviously during the 2020 Spring Festival, excluding BB emissions. As shown in Fig. 1, VE had the maximum reduction in pollution source concentration (28.39 μg·m$^{-3}$), followed by DF (11.47 μg·m$^{-3}$), FB (10.39 μg·m$^{-3}$), CC (8.54 μg·m$^{-3}$), and SIA (3.95 μg·m$^{-3}$). The reduction of VE may be predominantly attributed to the lockdown measures with the less motorized traffic of COVID-19 in 2020. Similarly, the traffic restrictions also resulted in the reduction of DF emissions. The average contribution concentration of FF emissions in 2020 was 42.89% lower than the average in 2019. It demonstrated that FF emission was effectively controlled due to the

Fig. 4. Factor profiles of coal combustion (CC), biomass burning (BB), dust fall (DF), secondary inorganic aerosol (SIA), vehicle emissions (VE), and firework burning (FB) derived from PMF model. (A: 2019; B:2020).

Fig. 5. The diagnostic ratios for the pollution source identification of PAHs.
implementation of the “Limited Fireworks” policies, which caused fewer fireworks display activities. However, the average concentration of BB emissions showed the reverse variations with an increasing amount of 5.20 μg·m⁻³. Under the influence of COVID-19, it may be mainly attributed to the larger family sizes in 2020 than those in 2019, resulting in an increase of BB emissions from cooking during Spring Festival (Dai et al., 2021a; Du et al., 2021). Meanwhile, the increase in BB emission may be also attributable to the winter heating using biomass burning rather than chunk coal. Because more strict regulations were implemented in Linfen during the winter of 2019 and “No Coal Zone” and “No Combustion Zone” were delineated (http://sthjj.linfen.gov.cn/content/s/2588/396031.html). And solid fuels, such as bulk coal, was replaced with cleaner energy (Zhao et al., 2021a). Simultaneously, it was evidenced by the reduction of the average contribution concentration of CC sources (23.82%) between 2019 and 2020.

Consequently, many factors, including the “Limited Firework” policy, the lockdown measures, and the switch of energy structure, led to the variation of pollution sources. However, we found that the SIA source proportion increases significantly from 2019 (29.8%) to 2020 (39.8%), indicating that secondary aerosol was dominant about the haze episode during the 2020 Spring Festival. Sun et al. (2020) discovered that the percentage of secondary aerosol to PM pollution was about 70% during haze periods in recent years. And secondary aerosol was not only affected by primary emissions but also controlled by meteorological conditions (Sulaymon et al., 2021). Hence, the contributions of meteorological conditions to PM pollution should be further considered.

3.4. Variation of meteorological conditions

3.4.1. The role of meteorology condition during Spring Festival

We performed de-weather model to isolate the influence of meteorological conditions. Due to the limitation of low-resolution offline sampling, we used 1-hour resolution air pollution data (PM2.5). Meanwhile, there was a good correlation (2019: R² = 0.78, 2020: R² = 0.88) between online and offline PM2.5 concentrations in Fig. S10, suggesting that model result was reasonable. As shown in Fig. S12, the pollutant emissions decreased significantly (~14.94 μg·m⁻³) after the Chinese New Year Eve, and the contribution of emissions decreased by 50.98%, which may be attributable to lockdown measures to contain COVID-19. The contribution of meteorological conditions on air pollution increased the average PM2.5 concentration by 6.12 μg·m⁻³ in comparison to 2019. And the average contribution of meteorological conditions increased by 20.21%. Compared to the pollution emissions, meteorological conditions played a crucial role in located air pollution (Zheng et al., 2020). However, the influence of meteorological conditions on air pollution included two aspects principally. Firstly, stationary and high-humidity weather conditions may facilitate the secondary generation and be not conducive to the pollutant’s diffusion. As shown in Fig. S11, the average values of RH in 2019 and 2020 Spring Festival were 50.21 ± 20.76% and 65.96 ± 20.10%, with the 31.1% growth rate of RH. Secondly, regional transports of pollutants may result in local accumulation (He et al., 2017; Liu et al., 2022). Cao and Cui (2021) demonstrated that the topography of Fenwei Plain had a certain impact on regional air pollution. Therefore, regional transports of pollutants deserve further attention.

We used the polar plot (Fig. S13) to analysis the relationship between PM2.5 concentration and wind speed and direction during the 2019 and 2020 Spring Festivals. Higher PM2.5 concentrations with southwest wind suggested that pollutants were transported from Yuncheng and central Shaanxi (Fenwei Plain) to Linfen. Overall, meteorological conditions may be a vital role in air pollution of Fenwei Plain, and regional transport of pollutants cannot be ignored (Li et al., 2021; Zhao et al., 2021b). However, the current study only discussed the possible transport direction of pollutants in a qualitative manner. A quantitative study was urgently required to accurately determine the contribution of regional and local emissions.

3.4.2. Decreased emission from potential source regions

To assess the impact of regional transport during the 2019 and 2020 Spring Festival, we discussed the of air mass trajectory and potential sources area of pollution sources by CWT analysis. As shown in Fig. S14-S15, air mass trajectories were mainly classified into two clusters, including northwest region and southeast region. Meanwhile, the northwest region covered the eastern Xinjiang, Gansu, Ningxia, and North Central Shaanxi, while the southeast region mainly included Jincheng, Yuncheng, southern Hebei, and northern Henan. Therefore, the direction of air mass did not have significantly variation during the 2019 and 2020 Spring Festival.

As the result of CWT, higher concentration (>90 μg·m⁻³) of CC source was found in northern Henan and the southern Hebei during the 2019 Spring Festival (Fig. 6), but some areas with concentrations greater than 20 μg·m⁻³ were found near Linfen in 2020 (Fig. 7). This may be due to the closure of heavy industry in BTH regions under the influence of COVID-19, reducing the result of CC emissions (Xing et al., 2020). Meanwhile, it was proved that pollutants may be transported from BTH region to Fenwei Plain (Liu et al., 2020b). For BB emissions, high level (>20 μg·m⁻³) was presented in northern Shaanxi during the 2019 Spring Festival, while primary source areas were located near Linfen during the 2020 Spring Festival. Similarly, the increased average concentration of BB emissions from PMF result also explained this. DF source was mainly affected by northwest air mass. High level (>40 μg·m⁻³) of VE was found in northern Henan during the 2019 Spring Festival, while SIA and VE were mainly influenced by local emissions during the 2020 Spring Festival. For FB, the potential source areas included eastern Linfen and central Shaanxi (Baqiao and Xi’an, Fenwei Plain) in 2019. However, the potential sources only covered eastern Linfen in 2020. Therefore, fireworks burning should be further controlled in Linfen.

Overall, in 2019, the regional transport of CC, BB, VE, and FB was obvious, including BTH regions and Fenwei Plain. However, the potential source area was mainly located near Linfen in 2020, resulting that strict control measures may affect the regional transport of pollutants (Kong et al., 2018). Meanwhile, the proportion quantification of regional transmission and local emissions should be further investigated.

3.5. Reduced health effect of PAHs

As shown in previous studies, BaP was an individual PAH with high carcinogenic potential (Zhang et al., 2019). The BaPeq values were generally used to calculate health effect as a quantitative description of toxic PAHs concentration. In this study, the average concentration of BaPeq decreased by about 24.4%, and the mean value of BaP reduced by about 18.4%. Meanwhile, the largest reduction of ILCR in VE emissions was 44.6%, followed by CC (43.2%), FB (34.1%), BB (21.3%), and DF (2.0%), as presented in Fig. 2. It demonstrated that more control measures were effective to reduce both the mass concentration and the health effects of PAHs in Linfen during the 2020 Spring Festival. Combined to the PMF results, PAHs was mainly influenced by CC source, BB, VE, and FF emission. However, the mass concentration of BB sources increased while the ILCR values decreased, indicating that concentration of PAHs related to BB sources decreased gradually while others chemistry species related to BB sources increased (Chen et al., 2017). To conclude, it was obvious that the control measures of coal combustion and vehicle emission were the most effective to decrease the PAHs health risks. However, it still was not clear that the decrease of health risks associated with PAHs was mainly affected by the routine control measures (“Limited Firework” policy and the switch of energy structure) or the strict control measures under the influence of COVID-19 (lockdown measures). Therefore, health risk of PAHs was be paid attention in Linfen.
4. Conclusion and future implications

This study investigated the variation of air quality under the influence of control measures during the 2019 and 2020 Spring Festival. Obviously, the concentration of PM$_{2.5}$ decreased from 199.00 ± 96.48 μg·m$^{-3}$ (2019) to 121.36 ± 70.79 μg·m$^{-3}$ (2020). And the average values of Ca$^{2+}$, K$^+$, SO$_4^{2-}$, NO$_3^-$, Cl$^-$, EC, and 16 PAHs reduced by 1.47 μg·m$^{-3}$, 17.46 μg·m$^{-3}$, 17.92 μg·m$^{-3}$, 13.25 μg·m$^{-3}$, 10.58 μg·m$^{-3}$, 2.27 μg·m$^{-3}$, and 61.65 ng·m$^{-3}$, respectively. Whereas the mean values of NH$_4^+$, Na$^+$, and OC in 2019 were 15.5% (0.25 μg·m$^{-3}$), 4.7% (0.83 μg·m$^{-3}$), and 3.0% (1.16 μg·m$^{-3}$) higher than the average in 2020 in the form of percentage. Meanwhile, the significant decrease of NO$_2^-$ caused neutral particles, indicating that control measures influenced the formation mechanism of haze and the pollution source variation. Combined to the PMF result, the apportioned average concentration of six source had substantially decreased, except for BB emission. Meanwhile, potential source area of BB was disturbed in near Linfen, demonstrating the increase of local BB emissions. Additionally, the contribution of meteorological conditions to PM$_{2.5}$ increased by 20.21%, and regional transport of pollutants in Fenwei Plain was more apparent during COVID-19 outbreak. The largest reduction of PAHs- ILCR in VE emissions was 44.6%, followed by CC (43.2%), FB (34.1%), BB (21.3%), and DF (2.0%).

In this study, it was important to realize that variation of contribution of source emission was significant to insight into the effectiveness of pollution control measures. Furthermore, the increase of BB emission was worthy of further attention in Linfen during the promotion of clean energy and control measures. However, based on the current study, the contribution of the routine control measures (“Limited Firework” policy and the switch of energy structure) or the lockdown measures (the COVID-19 periods) on air pollution still cannot be quantified. And the regional transport of pollutants in Fenwei Plain need to further investigate by reliable emission inventory data and modeling systems in future studies.

Fig. 6. Weighted Concentration Weighted Trajectory (CWT) analysis results for coal combustion (CC), biomass burning (BB), dust fall (DF), secondary inorganic aerosol (SIA), vehicle emissions (VE), and firework burning (FB) during the 2019 Spring Festival.
Author contributions statement

Weijie Liu: Investigation, Formal analysis, Methodology, Software, Visualization, Writing - Original Draft Preparation, Writing - Review and Editing; Yao Mao: Formal analysis, Methodology; Tianpeng Hu: Formal analysis, Methodology; Mingming Shi: Formal analysis, Methodology; Jiaquan Zhang: Data Curation, Supervision; Yuan Zhang: Formal analysis, Data Curation; Shaofei Kong: Data Curation, Supervision; Shihua Qi: Data Curation, Supervision; Project Administration; Xinli Xing: Writing - Review and Editing; Data Curation; Resources; Validation; Supervision, Project Administration; All authors have read and agreed to the published version of the manuscript.

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.

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Appendix A. Supplementary data

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