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Air pollutant variations in Suzhou during the 2019 novel coronavirus (COVID-19) lockdown of 2020: High time-resolution measurements of aerosol chemical compositions and source apportionment

Honglei Wang a, *, Qing Miao b, Lijuan Shen a, Qian Yang b, Yezheng Wu b, Heng Wei b

a Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters, Key Laboratory for Aerosol-Cloud-Precipitation of China Meteorological Administration, Nanjing University of Information Science & Technology, Nanjing, 210044, China

b Suzhou Environmental Monitoring Center, Suzhou, 215000, China

Abstract

To control the spread of the 2019 novel coronavirus (COVID-19), China imposed rigorous restrictions, which resulted in great reductions in pollutant emissions. This study examines the characteristics of air pollutants, including PM2.5 (particles with aerodynamic diameters < 2.5 μm), gas pollutants, water-soluble ions (WSIs), black carbon (BC) and elements, as well as the source apportionment of PM2.5 in Suzhou before, during and after the Chinese New Year (CNY) holiday of 2020 (when China was under an unprecedented state of lockdown to restrict the COVID-19 outbreak). Compared to those before CNY, PM2.5, BC, SNA (sulfate, nitrate and ammonium), other ions, elements, and NO2 and CO mass concentrations decreased by 9.9%–64.0% during CNY. The lockdown policy had strong (weak) effects on the diurnal variations in aerosol chemical compositions (gas pollutants). Compared to those before CNY, source concentrations and contributions of vehicle exhaust during CNY decreased by 72.9% and 21.7%, respectively. In contrast, increased contributions from coal combustion and industry were observed during CNY, which were recorded to be 2.9 and 1.7 times higher than those before CNY, respectively. This study highlights that the lockdown policy that was imposed in Suzhou during CNY not only reduced the mass concentrations of air pollutants but also modified their diurnal variations and the source contributions of PM2.5, which revealed the complex responses of PM2.5 sources to the rare, low emissions of anthropogenic pollutants that occurred during the COVID-19 lockdown.

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Positive matrix factorization (PMF) is the most popular and widely used model for source apportionment studies internation-
ally and is used in the study of haze processes and urban PM$_{2.5}$ pollution (Jaeckels et al., 2007; Kim et al., 2018; Li et al., 2020b; Okuda et al., 2010). For example, by using the PMF model, Zheng et al. (2020) suggested that primary emissions have decreased while secondary formation has increased since the lockdown in Wuhan. Moreover, the accuracy of PMF results is highly dependent on the time resolution of the observation data. Yu et al. (2019) reported that 1-h measurement data were preferred over 23-h averages for source apportionments during PMF model simulations due to the limits of inter-sample variability. Numerous studies have been carried out that involve PMF studies on air pollutants in the YRD (Mo et al., 2017; Wang et al., 2020; Tang et al., 2016; Yu et al., 2019), which are crucial to air quality control and subsequent policy formulation. Overall, a previous study demonstrated the great impacts of air pollution control policies, such as the activities of APEC, the 2nd World Internet Conference and the 2016 G20 Summit, on pollutant emissions; these policies also significantly affected the chemical compositions, spatiotemporal variations and formation mechanisms of aerosols (Cai et al., 2017; Shen et al., 2017; Sun et al., 2016; Yu et al., 2018). However, the true contribution of emission controls to particle mass (PM) reduction remains poorly constrained, largely due to variations in meteorological factors and regional atmospheric transport (Sun et al., 2016). Moreover, emission controls have usually targeted certain areas of China for a guarantee of previous events, with a focus on Hangzhou and its surroundings for the G20 Summit, for instance (Yu et al., 2018), which cannot eliminate the impacts of regional transport of air pollutants as a result.

The 2020 Chinese New Year (CNY) holiday was originally scheduled to take place from January 24 to 30, 2020. To control the spread of the 2019 novel coronavirus (COVID-19), the 2020 CNY holiday was delayed until February 10 (a delay of 18 days in total). During the CNY holiday, quarantines and roadblocks were imposed in urban and rural areas, which dramatically influenced the country’s economy. Energy demand and industrial output were reported to decrease sharply during lockdown periods (Le Quéré et al., 2020; Myllyvirta, 2020). These restrictions are believed to have caused large reductions in air pollutant emissions in China (Huang et al., 2020a; Chang et al., 2020; Shi and Brasseur, 2020). During the COVID-19 lockdown, satellite-derived NO$_2$ column density data showed substantial decreases of 40% on average over Chinese cities relative to the same period in 2019 (Bauwens et al., 2020). Bao and Zhang (2020) found that travel restrictions reduced human mobility by 69.85%; reductions in AQI, PM$_{2.5}$ and CO levels were partially mediated by human mobility; SO$_2$, PM$_{10}$ (particles with aerodynamic diameters of <10 μm) and NO$_2$ levels were completely mediated. Primary pollutants were reported to have decreased dramatically due to the lockdown policy, while air oxidation and secondary pollutant levels (O$_3$, secondary aerosols) were reported to have increased (Chang et al., 2020; Huang et al., 2020a; Xu et al., 2020; Zheng et al., 2020). However, the variations in chemical compositions and local and regional transport sources of PM$_{2.5}$ during the lockdown period remain unclear.

As an influential city in the YRD, Suzhou had a permanent residential population of 10.75 million people, and the number of residents who owned vehicles was 4.1774 million in 2019 (http://www.tjcn.org/tjgb/10js/36361_4.html). Numerous investigations of atmospheric pollution in Suzhou have been conducted recently (Costabile et al., 2006; Tian et al., 2016; Wang et al., 2015; Yang et al., 2012). Yang et al. (2012) reported that the health impacts of O$_3$ are more evident in the cooler seasons in Suzhou. Tian et al. (2016) reported that OM, ($\text{NH}_4$)$_2$SO$_4$ and NH$_4$NO$_3$ are major contributors to visibility impairment, but this share differs from that of haze events. Wang et al. (2017) reported a summer aerosol concentration in Suzhou of 12,797 ± 5931 cm$^{-3}$ with a unimodal distribution of the spectrum peaking at 100–110 nm. Due to the several impacts of the CNY and COVID-19 lockdown, emission reduction levels were substantial. As such, investigating the changes in the chemical components of aerosols that occurred during this special period can deepen our understanding of the impact of anthropogenic sources on air pollutants in urban areas. To date, numerous studies have been carried out on the impact of the lockdown on air quality and on source apportionment of aerosols in the NCP and in Hubei Province (Cui et al., 2020; Dai et al., 2020; Sun et al., 2020; Zheng et al., 2020); however, few studies have been conducted on the YRD. Given that emission sources, economic structures and meteorological patterns differ in different regions of China, the impact mechanisms of the lockdown policy on the distributions and sources of air pollutants were distinct in each region. This study examines variations in air pollutants, including PM$_{2.5}$, gas pollutants, water-soluble ions (WSIs), black carbon (BC) and elements, that were measured in the major city of Suzhou in the YRD before, during and after the 2020 CNY holiday. Moreover, the distinct PM$_{2.5}$ sources that were involved in different stages were identified using a 1-h dataset that was obtained via positive matrix factorization (PMF) model analysis to reveal the characteristics sources to which the residents of Suzhou were exposed and to develop better pollution management strategies.

2. Methods and materials

2.1. Observation datasets

The observation site is located at the South Gate Station in Suzhou (31.29°N, 120.63°E), which is a national control point of the China Environmental Monitoring Station; this site is surrounded by residential areas and is located 100 m from the southwest road of Suzhou to the south, which makes it a mixed region of commercial and residential areas. Air pollutants and meteorological data were recorded with a time resolution of 1 h, and the observation period ran from January 1 to February 29, 2020. WSIs in PM$_{2.5}$ were detected using an Ambient Ion Monitor-Ion Chromatograph (AIM-IC, URG-9000D, Thermo Scientific™, USA), which can continuously measure ion mass concentrations, including Na$^+$, NH$_4^+$, K$^+$, Mg$^{2+}$, Ca$^{2+}$, Cl$^-$, NO$_3^-$ and SO$_4^{2-}$ (Malaguti et al., 2015). Quality control experiments, including collection efficiency, flow calibration and air tightness tests, were carried out each month. An internal check standard of lithium bromide injected into each sample was also used to confirm IC analysis accuracy. Additionally, ICS were calibrated by testing concentration gradients of the standard agent (Merck, Germany) using the external calibration method (Tang et al., 2020). Twenty-three elements (e.g., K, Fe, Zn, Ca, Si, Mn, Pb, Cu, Ti, As, V, Ba, Cr, Se, Ag, Cd, Ni, Au, Co, Sn, Sb, Tl, and Hg plus Pd for quality assurance) were monitored by an Xact 625 ambient metals monitor (Cooper Environmental Services (CES), Beaverton, OR, USA) via X-ray fluorescence (XRF) (Furger et al., 2017). For each sample analysis, the detector energy gain was automatically adjusted using pure Pd as an internal standard. The XRF response of the element of interest was calibrated using the standard thin film provided by the manufacturer. The measured mass was in good agreement with the standard mass for each element, and the deviation was less than 5%. BC mass concentrations were observed with a Model AE-31 aethalometer (Mage Scientific, USA), which simultaneously measures the attenuation of light as it passes through a particle laden filter spot and through a particle free filter spot (reference spot) of filter tape at seven wavelengths (e.g., 370, 470, 520, 590, 660, 880 and 950 nm) (Hansen and Schnell, 2005). Although measurements were conducted every 5 min, the data...
were averaged over a period of 1 h to decrease uncertainties originating from instrumental noise, flow rates, filter spot areas and detector responses (Corryman et al., 2006). Silica gel in the drier was used upstream of the AE-31 instrument to change the monitored relative humidity (RH) to ensure that the RH of the sample air was below 40%. Air in the container was released to the area surrounding the drier to reduce the temperature difference between the interior of the container and drier. Temperatures within the container were maintained at approximately 25 °C. PM2.5 concentrations were measured by a Synchronized Hybrid Ambient Real-time Particulate Monitor (SHARP Model 5030L, Thermo Scientific™, USA), and gas pollutants, NO2, SO2, CO and O3 were monitored by a series of monitors (e.g., 42i, 43i, 48i, and 49i, Thermo Scientific™, USA) (Wei et al., 2020). PM2.5 concentrations were measured using the β absorption method. NO2, SO2 and O3 concentrations were measured using chemiluminescence, ultraviolet fluorescence, and UV spectrophotometry methods, respectively. CO was measured using the nondispersive infrared absorption method and gas filter correlation infrared absorption method. Wang et al. (2014b) provide a more detailed introduction to these instruments. Meteorological data (e.g., temperature, relative humidity (RH), wind speed, wind direction, visibility and precipitation) were observed with the CSI-CHR000 (Logan, UT, USA) automatic weather station.

Measurement periods were classified as being before (January 1 to 23, 2020), during (January 24 to February 10, 2020), and after (February 11 to 29, 2020) the CNY holiday of 2020.

2.2 Source apportionment

The U.S. Environmental Protection Agency (EPA) PMF version 5.0 has been widely applied for PM2.5 source apportionment (Bari and Kindzierski, 2018; Callén et al., 2014; Ji et al., 2018; Paatero and Tapper, 1994; Zheng et al., 2020; Ziková et al., 2016). A detailed description of the PMF model can be found in the supplementary material. WSIs, BC, NO2, SO2 and elements were included for PMF analysis. Species were categorized as strong, weak, and bad according to their signal-noise ratios and percentages below detection limits (Callén et al., 2014). Species were categorized as “bad” when the signal-to-noise (S/N) ratio was <0.5, “weak” if the S/N ratio was greater than 0.5 but less than 1 and “strong” if the S/N ratio was greater than 1 (US EPA, 2014). Finally, datasets of (sample number × species number) 552 × 26, 432 × 26 and 456 × 26 were introduced into the model before, during and after CNY periods, respectively. Choosing an optimal factor number is challenging. Considering too many factors results in meaningless outputs, while choosing too few factor numbers may result in mixed sources (Bressi et al., 2014). The PMF model was run with a random seed, and the lowest Q was considered as the base run solution. Q-values, resulting source profiles and scaled residuals were examined. Resolution conditions can be found in the supplementary material. Finally, five factors that were in effect before and after CNY and six factors that were in effect during CNY were considered to be the most reasonable solutions. The uncertainties of the PMF model are usually estimated by bootstrapping (BS), displacement (DISP), and bootstrapping with displacement (BS-DISP). All factors are mapped over 96%–99% in BS. There was no factor swap and no decrease in the Q value in DISP. There were no swaps with DISP, and 97%–98% of the BS-DISP runs were successful.

3 Results and discussion

3.1 Changes in air pollutant loading

PM2.5 concentrations during CNY were noticeably lower than those before CNY (Fig. 1c), whereas their hourly concentrations still exceeded 75 μg m⁻³ several times under periods with rigorous restrictions, such as from 00:00–16:00 on January 30, from 03:00–15:00 on January 31, from 13:00–19:00 on February 3 and from 07:00–11:00 on February 9, when the maximum PM2.5 concentrations varied from 89 to 118 μg m⁻³. According to Chinese air quality standards, a 24 h-mean PM2.5 concentration of more than 75.0 μg m⁻³ indicates the presence of air pollution. These pollution episodes were weaker than those that occurred before CNY but were much stronger than those occurring after CNY. During CNY, PM2.5 had a mean concentration of 38.8 μg m⁻³, which was 36.5% lower than that measured before CNY (61.4 μg m⁻³) and slightly higher than that measured after CNY (31.8 μg m⁻³) (Fig. 2b).

As shown in Fig. 1c, the variations in chemical compositions of PM2.5 were similar to those of PM2.5, while large discrepancies were still found among the different chemical species. Given the impact of fireworks, K⁺ concentrations were mostly greater than 1.0 μg m⁻³ from 22:00 on January 24 to 03:00 on January 25 and peaked at 00:00 on January 25 with a value of 3.1 μg m⁻³, which exceeded the levels measured in non-fireworks periods by more than 10-fold. The mean concentrations of BC, SNA (i.e., sulfate, nitrate and ammonium), other ions and elements during CNY were 1.5, 10.3, 12.2 and 1.6 μg m⁻³, respectively, (Fig. 2a), and were 51.1%, 48.2%, 9.9% and 23.3% lower, respectively, than the concentrations measured before CNY and were 0.5%, 29.7%, 23.6% and 41.7% higher, respectively, than those measured after CNY. Although people started returning to work in China after the Spring Festival (February 10), Jiangsu Province modified its level-I response to a level-II response on February 25, 2020 due to the public health emergency. Hence, the lockdown policy was not fully lifted after CNY, which resulted in relatively low pollutant concentrations. Li et al. (2020a) reported that industrial activity is the dominant PM2.5 contributor with a relative contribution of 32.2%–61.1% in the YRD, while the relative contribution from residential sources was higher during the lockdown period. Moreover, high PM2.5 concentrations were observed on New Year’s Eve (January 24) during CNY, which is attributed to fireworks. A short PM2.5 peak from January 27 to February 1 was also recorded and was due to unfavorable meteorological conditions, which are discussed in detail elsewhere (Shen et al., 2020). Hence, air pollutants reached higher concentrations during CNY than after CNY.

During CNY, NO2 concentrations were significantly lower than they were before and after CNY, and they varied inconsistently with the PM2.5 concentrations, which were different from the changes observed before CNY (Fig. 1b). For instance, NO2 concentrations were relatively high at 70–73 μg m⁻³ from 21:00 on January 21 to 00:00 on February 1, during which the PM2.5 concentrations were only 42–49 μg m⁻³. In contrast, the NO2 variations generally corresponded to the PM2.5 concentrations during the haze period before CNY (i.e., January 11 to 16). In addition, meteorological conditions also influenced PM2.5 concentrations, such as high RHs and low wind speeds, which also led to similar variations in air pollutants (Bressi et al., 2014). As such, the correlation coefficient of NO2 and PM2.5 was 0.57 before CNY and was thus much higher than that during (0.19) and after (0.32) CNY, which is in agreement with other results (Bauwens et al., 2020; Chang et al., 2020; Xu et al., 2020). The correlation coefficient of NO2 and NO3 was 0.47 before CNY and was thus higher than that during (0.16) and after (0.26) CNY. Such high correlations before CNY reveal that vehicle exhaust contributes greatly to NO2 and NO3 and further increases PM2.5 concentrations. Hence, the NO2 variations were mostly similar to the PM2.5 variations. A dramatic reduction in traffic sources was the main driver of decreased NO2 concentrations during CNY and reached a mean of 16.2 μg m⁻³ which was 64.0% and 23.1% lower than before (45.0 μg m⁻³) and after (21.0 μg m⁻³) CNY, respectively (Fig. 2d).
The decreased contributions of traffic sources to PM$_{2.5}$ also led to lower correlation coefficients between NO$_2$ and PM$_{2.5}$ during CNY. SO$_2$ concentrations showed flat variations during the whole observation period (Fig. 1b), and they had mean concentrations of...
5.0, 4.0 and 5.2 μg m⁻³ before, during and after CNY, respectively (Fig. 2c). CO concentrations showed variations that were consistent with those of PM₂.₅, with a mean of 0.7 mg m⁻³ that was measured during CNY, which was 5.2% lower and 16.5% higher than the concentrations measured before and after CNY, respectively (Fig. 2f). The mean O₃ concentrations reached 66.8 and 62.7 μg m⁻³ during and after CNY, respectively, which were 2.0 and 1.9 times higher than that before CNY, respectively (Fig. 2e). Such opposing changes are mainly explained by the unprecedented reduction in NOₓ emissions that led to less O₃ titration by NO (Sicard et al., 2020). Greater decreases in NO₂ concentrations during and after CNY resulted in weaker titration effects and even higher O₃ concentrations that occurred during and after CNY could also have resulted in weakened solar radiation extinction; such conditions favor ozone formation. A detailed investigation of O₃ enhancement during CNY resulted in weaker titration effects and even higher O₃ concentrations (Fig. 2). In addition, the sharp reduction in PM₂.₅ concentrations that occurred during and after CNY could also have resulted in weakened solar radiation extinction; such conditions favor ozone formation. A detailed investigation of O₃ enhancement during CNY resulted in weaker titration effects and even higher O₃ concentrations (Fig. 2). In addition, the sharp reduction in PM₂.₅ concentrations that occurred during and after CNY could also have resulted in weakened solar radiation extinction; such conditions favor ozone formation. A detailed investigation of O₃ enhancement during CNY resulted in weaker titration effects and even higher O₃ concentrations during CNY and ranged from 25.2% (14:00) to 35.1% (08:00) from 07:00–15:00 and exceeded 38% in other time segments, with a maximum of 42.7% occurring at 03:00. Meanwhile, slight changes in PM₂.₅ concentrations were observed when precipitation occurred during CNY (Fig. 3a). Given that light rain dominated during the observation period with hourly precipitation rates mostly remaining below 1 mm h⁻¹, scavenging effects on PM₂.₅ were weak as a result. Meanwhile, these stable weather conditions with high RHs were conducive to pollutant accumulation and to the formation of secondary aerosols, which could be the dominant driver of PM₂.₅ increases. Fig. 4b reveals that wind speeds averaged approximately 3.1 m s⁻¹ with few changes occurring throughout the study period, which suggest that there were few distinctions in diffusion conditions; as such, the discrepancies in air pollutant concentrations that occurred in different stages were caused mainly by changes in emission sources.

Although diurnal PM₂.₅ variations underwent considerable changes in different stages (Fig. 3b), visibility values presented unimodal distributions (Fig. 4d). Moreover, the correlations between PM₂.₅ and visibility weakened with decreases in PM₂.₅ concentrations, with coefficients of −0.51, −0.46 and −0.33 measured before, during and after CNY, respectively. In particular, the differences between PM₂.₅ variations that were measured during and after CNY were much smaller than those of visibility according to comparisons between Figs. 3b and 4d, which show that PM₂.₅ was not the principal factor that affected visibility when its concentrations were low. For instance, PM₂.₅ concentrations during CNY were 1.5–1.6 times higher than those after CNY from 09:00–11:00, during which the PM₂.₅ difference was the greatest, yet the difference in visibility was less than that measured from 13:00–17:00 (Fig. 4d). As the RH after CNY was lower than that during CNY from 13:00–18:00 (Fig. 4c), aerosol water contents probably had greater impacts on visibility when PM₂.₅ concentrations were low.

NO₂ exhibited bimodal distributions during different stages and peaked at 08:00 and 20:00 during and after CNY, whereas NO₂ peaked at 10:00 and 19:00–21:00 before CNY (Fig. 5a). The rush hours may have delayed the morning peak and advanced the evening peak. Diurnal variations in NO₂ were similar in different stages, with the lowest values recorded at noon. High wind speeds at noon (Fig. 4b) were responsible for NO₂ diffusion. Meanwhile, high temperatures (Fig. 4a) and low RHs (Fig. 4c) also encouraged photochemical reactions of NO₂. During the lockdown period,
Fig. 4. Diurnal variations of meteorological factors before, during and after CNY.

Fig. 5. Diurnal variations of trace gases before, during and after CNY.
traffic sources decreased significantly, which resulted in a lower contributions to NO₂, which were also impacted by industrial and natural sources. As such, the diurnal variations in NO₂ were little affected by the lockdown. Unimodal distributions were recorded for diurnal variations in SO₂ that peaked from 10:00–13:00 in different stages with small discrepancies in concentrations (Fig. 5b), which can be attributed to slight variations in SO₂ emission sources that were mainly affected by industrial coal burning. Diurnal variations in CO exhibited a bimodal distribution before CNY, with peaks occurring from 08:00–09:00 and from 19:00–21:00. However, trimodal distributions were observed for CO concentrations during and after CNY and peaked at 09:00, 13:00 and 19:00. CO is generally produced by incomplete combustion and is reported to be mainly generated from industry and vehicle exhaust in urban areas (Parrish et al., 2009; Sahu et al., 2015); as such, the CO diurnal variations showed a similar pattern as those of NO₂. Unimodal distributions were consistently found for O₃ and peaked from 13:00–16:00 in different stages (Fig. 5d), which are consistent with the variations in air temperature (Fig. 4a).

The diurnal variations in BC exhibited a bimodal distribution before CNY and peaked at 07:00 and 19:00 (Fig. 6a), and then changed to a trimodal distribution during CNY with peaks occurring at 07:00, 13:00 and from 17:00–19:00. A flat variation was found for BC after CNY, which was similar to those of SNA for different stages with higher concentrations than those measured before CNY.

Diurnal variations in SO₄²⁻, NO₃⁻ and NH₄⁺ showed minor differences throughout the observation period (Fig. S1), consistent with those of SNA (Fig. 6b). Trajectory lengths represent the transport speeds of air masses. Fig. S2 shows that the 24-h backward trajectories were similar for different periods and mostly exceeded 300 km, which suggested great impacts from the foreign pollutant transport process on air pollutants in Suzhou. Meanwhile, northerly air masses were dominant during the observation period and came mainly from the northwest inland and northeast marine areas. Some southerly air masses were noticed before and after CNY, which were also from marine areas with source characteristics that were similar to the northeast marine air masses. Overall, such long-range transport of air masses was conducive to the aging process of aerosols, which further promoted secondary ion formation, such as SO₄²⁻, NO₃⁻ and NH₄⁺, which showed weak diurnal variations as a result. SO₄²⁻ exhibited a unimodal distribution that peaked at 14:00 during CNY. However, there were few diurnal differences in SO₄²⁻ before and after CNY. SOR exhibited high and low concentrations in the daytime and nighttime, respectively, during CNY, which were consistent with those of SO₄²⁻. In contrast, lower and higher SOR values were recorded in the daytime and nighttime, respectively, before and after CNY (Fig. 5a). Mean SO₄²⁻ concentrations reached 5.3, 3.6 and 1.6 µg m⁻³ before, during and after CNY, respectively, and showed larger discrepancies than those of SO₂ (Fig. 2c). The correlation coefficients between SO₂ and SO₄²⁻ were 0.5, 0.5 and 0.2 before, during and after CNY, respectively. The SOR values after CNY were much lower than those before and during CNY, which revealed a minor contribution of SO₂ conversion to SO₄²⁻ after CNY. Overall, sulfate can be sourced from heterogeneous processes that were distinct from the photochemical reactions of SO₂ during the observation period. Shen et al. (2021) also reported that nitrate and sulfate formation can be more strongly influenced by aerosol water contents, pH values and air oxidation compared with their precursors.

NO₃⁻ exhibited mean concentrations of 9.7, 3.9 and 3.3 µg m⁻³ before, during and after CNY, respectively, with changes occurring in different stages that were similar to those of NO₂ (Fig. 2d). However, these diurnal variations in NO₃⁻ were much weaker than those of NO₂ (Fig. S1b). NOR showed significant diurnal variations which peaked at 04:00 and 15:00 (Fig. S3b) and lagged behind those of NO₂, which is ascribed to the conversion of NO₂ to NO₃⁻. It is worth noting that NO₃⁻ concentrations were much higher before CNY than during and after CNY, while NOR concentrations showed small differences before, during and after CNY, which suggest a

Fig. 6. Diurnal variations of chemical components before, during and after CNY.
more complex source of NO$_3^-$ before CNY. The correlation coefficients between NO$_2$ and NO$_3^-$ were 0.5, 0.2 and 0.3 before, during and after CNY, respectively, which suggested that NO$_3^-$ levels were strongly impacted by vehicle exhaust before CNY when compared with the levels during and after CNY when sharp decreases in traffic emissions were observed. Nitrate, which normally originates from photochemical and heterogeneous reactions (Shen et al., 2021), may be generated mostly by the aerosol aging process during its transportation, and after reaching Suzhou, such effects of pollutant transportation on nitrate cannot be neglected as a result. NH$_4^+$ concentrations underwent diurnal variations similar to those of NO$_3^-$ (Fig. S1) with mean concentrations of 4.8, 2.7 and 2.3 µg m$^{-3}$ measured before, during and after CNY, respectively.

The NO$_3^-$/SO$_4^{2-}$ ratios showed large discrepancies among different stages (Fig. S3c). Similar to NO$_2$, the NO$_3^-$/SO$_4^{2-}$ ratios were high during rush hour and were usually above 1.5 with a mean value of 1.7 before CNY, which revealed the noticeable contributions of K, Mg, Cu and Ba ions and elements. The latter is ascribed to higher wind speeds and low RHs, which were also responsible for their unimodal distributions after CNY, with higher values recorded in the daytime.

3.3. Variations in source contributions

Based on the PMF-modeled results, five factors were derived before and after CNY (Fig. S4 and S5), and six factors were identified as part of the optimal solution for the period during CNY (Fig. 7). Strong correlations were found between the modeled and observed concentrations before, during and after CNY (Fig. S6). The slope of the fitting function reached over 0.98 with R values ranging from 0.98 to 0.99, which indicate that the observations were reasonably represented by the PMF simulation.

The first factor is coal combustion in view of the higher observed loadings of BC, SO$_2$, AS, Se and Pb. Se and Pb have been widely reported to be tracers of coal combustion (Liu et al., 2018; Zheng et al., 2020), while BC is mainly generated from the incomplete combustion of fossil and biomass fuels (Qin and Xie, 2012; Streets et al., 2001), and SO$_2$ is normally produced by industrial coal burning processes (Kharol et al., 2020). The second identified factor refers to industrial processes with larger contributions of the heavy metal elements, Cr, Mn, Fe, Ni and Zn (Querol et al., 2007; Ji et al., 2018). The third factor, which is characterized by the higher loadings of Ca$^{2+}$, Ca and Mg$^{2+}$, pertains to dust sources (Zheng et al., 2020; Sudheer and Rengarajan, 2012). Vehicle exhaust was identified as the fourth factor, with higher NO$_2$ fractions of 62.3%–79.3% observed in the three stages (Chan et al., 2011; Liu et al., 2017). Secondary processes were identified as the fifth factor due to the higher loadings of NO$_3^-$, SO$_4^{2-}$ and NH$_4^+$. Additionally, the sixth factor, fireworks, was also observed during CNY, as large contributions of K, Mg$^{2+}$, K, Cr, Cu and Ba were observed (Kong et al., 2015; Rai et al., 2020; Vecchi et al., 2008; Zheng et al., 2020).

Although source concentrations (112 µg m$^{-3}$) and contributions (32.0%) of vehicle exhaust were higher than those of other sources during CNY, they decreased by 72.9% (39.7%) and 21.7% (19.5%) relative to those measured before (after) CNY (Fig. 8). These results exhibited bimodal distributions for the diurnal variations in vehicle exhaust after CNY (Figs. S7 and S8) with the peak hours corresponding to rush hours, which were consistent with results for NO$_2$ and CO (Fig. 5). However, variations in vehicle exhaust showed subtle peaks before and during CNY. The fraction remained at high values from 12:00 before CNY and peaked slightly at 08:00 and 11:00 during CNY. Figs. S9–S11 show that vehicle exhaust levels showed high values under low wind speeds in different stages, which revealed that its origin was mainly from local emissions in Suzhou, which are ascribed to traffic arteries around the observation site (Fig. S12). As such, high concentrations of vehicle exhaust at night during the lockdown were also observed (Fig. S7d).

Slight changes in the contributions of secondary processes and dust were observed, with values of 20.3% and 16.0% found for the former and values of 12.3%, 10.0% and 6.1% found for the latter before, during and after CNY, respectively. The source concentrations of secondary processes and dust during CNY decreased by 53.2% and 62.9%, respectively, relative to those measured before CNY. The diurnal variations in secondary processes were consistent in different stages (Figs. S7 and S8), which showed that this source type was little affected by the CNY holiday and lockdown policy. High concentrations due to secondary processes were found under light southerly winds before CNY (Fig. S9), westerly winds during CNY (Fig. S10) and northerly after CNY (Fig. S11). High wind speeds were accompanied by low RHs (Fig. 4) and were responsible for the high concentrations and proportions of dust sources during daytime (Figs. S7–S11). Due to human activities, dust concentrations peaked at 10:00, 15:00 and 21:00 before CNY (Fig. S7) and decreased noticeably during and after CNY due to the restrictions.

In contrast, increased contributions of coal combustion (20.7%) and industry (11.1%) were found during CNY, which were 2.9 and 1.7 times higher than those before CNY, respectively. The source concentrations of coal combustion and industry reached 5.5, 7.3, and 5.2 µg m$^{-3}$ and 5.1, 3.9, and 4.4 µg m$^{-3}$ before, during, and after CNY, respectively. Such flat variations in different stages reveal stable emissions from industrial processes, including power generation and iron and steel coking. The observed distributions of coal combustion were largely consistent with those of power plants (Figs. S9–S12). High concentrations of industry sources under northwesterly winds before and after CNY were observed, which highlight the strong impacts of industrial parks. During CNY, high industrial values were found in the northwestern and southwestern regions (Fig. S10), where power plants are distributed (Fig. S12), which indicate the strong influence of power plants on industrial concentrations. The diurnal concentrations of coal combustion showed subtle variations in different stages, which again confirmed a stable emissions source. It is worth noting that coal combustion and sea salt sources exhibited higher concentrations during CNY than before and after CNY from 04:00–15:00, which suggest the strong impacts of sea salt in view of the high wind speeds that occurred during this period (Fig. 4b). The proportions from coal combustion showed flat diurnal variations before and after CNY and reached peak values at 04:00, 09:00 and 13:00 during CNY (Fig. S8), probably due to the decreasing contributions of other sources, e.g., vehicle exhaust.

Finally, fireworks sources were also recognized, with concentrations and fractions of 1.8 µg m$^{-3}$ and 5.3%, respectively, measured during CNY and values being higher at night (Figs. S7 and S8). These high fireworks emission concentrations largely corresponded to high wind speeds from the northwest, which suggest that their origins were from the surrounding suburbs (Fig. S10).
4. Conclusions

In this work, the characteristics of air pollutants (e.g., PM$_{2.5}$, gas pollutants, WSIs, BC and elements) and PM$_{2.5}$ sources in Suzhou of the YRD were investigated before, during, and after CNY. Compared to before CNY, the mass concentrations of PM$_{2.5}$, BC, SNA, other ions, elements, NO$_2$ and CO reached 38.8, 1.5, 10.3, 1.2, 16.2 $\mu$g m$^{-3}$ and 0.7 mg m$^{-3}$, respectively, during CNY (COVID-19 lockdown), which represented decreases of 36.9%, 51.1%, 48.2%, 9.9%, 23.3%, 64.0% and 25.2%, respectively. The extent of PM$_{2.5}$ reduction was greater at night, with a maximum of 42.7% occurring at 03:00. The lockdown policy had strong (weak) effects on diurnal variations in aerosol chemical compositions (gas pollutants). Compared to before (after) CNY, source concentrations and contributions of vehicle exhaust during CNY decreased by 72.9% (39.7%) and 21.7% (19.5%), respectively. However, slight changes in
the contributions of secondary processes and dust were observed during CNY despite large reductions in their source concentrations (by 53.2% and 62.9%, respectively). In contrast, increased contributions of secondary processes and dust were observed during CNY despite large reductions in their source concentrations. 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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.116298.

References

Bao, R., Zhang, A., 2020. Does lockdown reduce air pollution? Evidence from 44 cities in northern China. Sci. Total Environ., 139052 https://doi.org/10.1016/j.scitotenv.2019.139052.

Bari, M.A., Kindzierski, W.B., 2018. Ambient volatile organic compounds (VOCs) in Beijing during COVID-19 shutdown. Geophys. Res. Lett. 47 (12), e2020GL088533 https://doi.org/10.1029/2020GL088533.

Calléon, M.S., Ramanathan, V., Schell, J.J., 2006. Impact of monsoon transitions on the physical and optical properties of aerosols. J. Geophys. Res. 111, 1–15. https://doi.org/10.1029/2005JD006370.

Costabile, F., Berton, G., Desantis, F., Wang, F., Varini, H., Fenglei, L., Allegriini, L., 2020. Preliminary assessment of major air pollutants in the city of Suzhou, China. Atmos. Environ. 40 (33), 6388–6395. https://doi.org/10.1016/j.atmosenv.2016.05.056.

Cui, Y., Y., D., Maenhaut, W., Gao, W., Zhang, R., Wang, Y., 2020. Levels and sources of hourly PM2.5-related elements during the control period of the COVID-19 pandemic at a rural site between Beijing and Tianjin. Sci. Total Environ. 744, 140840 https://doi.org/10.1016/j.scitotenv.2020.140840.

Dai, H., Zhu, J., Liao, H., Li, J., Liang, M., Yang, Y., Yue, X., 2021. Co-occurrence of ozone and PM2.5 pollution in the Yangtze River Delta over 2013–2018: spatiotemporal distribution and meteorological conditions. Atmos. Res. 249, 105363 https://doi.org/10.1016/j.atmosres.2020.105363.

Dai, Q., Liu, B., Bi, X., Wu, J., Liang, D., Zhang, Y., Feng, Y., Hopke, P.K., 2020. Dispersion normalized PMF provides insights into the significant changes in source contributions to PM2.5 after the COVID-19 outbreak. Environ. Sci. Technol. 54 (16), 9917–9927. https://doi.org/10.1021/acs.est.0c02776.

Fenger, M., Cruz Mingüillón, M., Yadav, V., Slowik, J.G., Higlin, C., Fröhlich, R., Petters, M., Prevot, A.S., 2010. Elemental composition of ambient aerosols measured with high temporal resolution using an online XRF spectrometer. Atmos. Meas. Tech. 10 (6) https://doi.org/10.5194/amt-10-2061-2017.

Hansen, A.D.A., Schnell, R.C., 2005. The Aetheralometer. Magee Scientific Company, Berkeley, California, USA, p. 7.

Huang, L., Lin, J., Koo, B., Yarwood, G., Yan, R., Wang, Y., Cheng, H., Li, L., 2019. Sulphate formation during heavy winter haze events and the potential contribution from heterogeneous SO2 + NOx reactions in the Yangtze River Delta region, China. Atmos. Chem. Phys. 19 (22), 14311–14328. https://doi.org/10.5194/acp-19-14311-2019.

Huang, X., Ding, A., Gao, J., Zheng, R., Zhou, D., Qi, X., Tang, R., Wang, J., Ren, C., Nie, W., Chi, X., Xu, Z., Chen, L., Li, Y., Chen, F., Pang, N., Wang, H., Tong, D., Qin, W., Cheng, W., Liu, W., Fu, Q., Liu, B., Chai, F., Davi, S.J., Zhang, Q., He, K., 2020a. Enhanced secondary pollution offset reduction of primary emissions running COVID-19 lockdown in China. Natl. Sci. Rev. nwwaa137 https://doi.org/10.1093/nsr/nwwaa137.

Huang, X., Ding, A., Wang, Z., Ding, K., Gao, J., Chai, F., Fu, C., 2020b. Amplified transboundary transport of haze by aerosol-boundary layer interaction in China. Nat. Geosci. 1–7 https://doi.org/10.1038/s41561-020-0583-4.

Jacquel, J.M., Bae, M.S., Schauer, J.J., 2007. Positive matrix factorization (PMF) analysis of molecular marker measurements to quantify the sources of organic aerosols. Environ. Sci. Technol. 41 (16), 5763–5769. https://doi.org/10.1021/es070253b.

Ji, D., Cui, Y., Li, L., He, J., Wang, L., Zhang, H., Wang, W., Zhou, L., Maenhaut, W., Wen, T., Wang, Y., 2018. Characterization and source identification of fine particulate matter in urban Beijing during the 2015 Spring Festival. Sci. Total Environ. 628, 430–440 https://doi.org/10.1016/j.scitotenv.2018.01.229.

Kang, H., Zhu, B., Gao, J., He, Y., Wang, H., Su, J., Chen, P., Zhu, T., Bu, Y., 2019. Potential impacts of cold frontal passage on air quality over the Yangtze River Delta, China. Atmos. Chem. Phys. 19 (6), 3673–3685. https://doi.org/10.5194/acp-19-3673-2019.

Kharel, S.K., Fioletov, V., McLinden, C.A., Shephard, M.W., Sioris, C.E., Li, C., Krotkov, N.A., 2020. Ceramic industry at Morbi as a large source of SO2 emissions in India. Atmos. Environ. 223, 117243 https://doi.org/10.1016/j.atmosenv.2020.117243.

Kong, S., Kim, T.Y., Yi, S.M., Heo, J., 2018. Source apportionment of PM2.5 using positive matrix factorization (PMF) at a rural site in Korea. J. Environ. Manag. 214, 325–334. https://doi.org/10.1016/j.jenvman.2018.03.027.

Li, L., An, J., Zhou, M., Qiao, L., Zhu, S., Yan, R., Ooi, C., Wang, H., Huang, C., Huang, L., Tao, S., Yu, J., Chan, A., Wang, Y., Feng, J., Chen, C., 2018. An integrated source apportionment method and its application on the potentially affected River Delta region, China. Atmos. Environ. 52 (24), 14216–14227. https://doi.org/10.1016/j.atmosenv.2018.08.011.

Li, L., Liu, Q., Huang, L., Wang, Q., Zhu, A., Xu, J., Liu, Z., Li, H., Shi, L., Li, R., Azan, M., Wang, Y., Zhang, X., Liu, Z., Zhu, Y., Zhang, K., Xue, S., Ooi, M., Zhang, D., Chan, A., 2020a. Air quality changes during the COVID-19 lockdown over the Yangtze River Delta Region: an insight into the impact of human activity pattern changes on air pollution variation. Sci. Total Environ. 139282 https://doi.org/10.1016/j.scitotenv.2020.139282.
