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Exploring the driving factors of haze events in Beijing during Chinese New Year holidays in 2020 and 2021 under the influence of COVID-19 pandemic

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HIGHLIGHTS

- Elevated PM2.5 was observed after Chinese New Year during two intensive observation periods.
- Unfavorable meteorology accounted for 11.0% and 16.9% of PM2.5 in both campaigns.
- Formation of secondary aerosols contributed to notably elevated PM2.5 in haze days.
- Regional transports facilitated increases of K⁺ and Cl⁻ due to fireworks setting off.
- Nocturnal chemistry contributed notable increase of secondary components in winter.

ABSTRACT

Unexpected outbreak of the 2019 novel coronavirus (COVID-19) has profoundly altered the way of human life and production activity, which posed visible impacts on PM2.5 and its chemical species. The abruptly emergency reduction in human activities provided an opportunity to explore the synergistic impacts of multi-factors on shaping PM2.5 pollution. Here, we conducted two comprehensive observation measurements of PM2.5 and its chemical species from 1 January to 16 February in Beijing 2020 and the same lunar date in 2021, to investigate temporal variations and reveal the driving factors of haze before and after Chinese New Year (CNY). Results show that mean PM2.5 concentrations during the whole observation were 63.83 and 66.86 μg/m³ in 2020 and 2021, respectively. Higher secondary inorganic species were observed after CNY, and K⁺, Cl⁻ showed three prominent peaks which associated closely with fireworks burnings from suburb Beijing and surroundings, verifying that they could be used as two representative tracers of fireworks. Further, we explored the impacts of meteorological conditions, regional transportation as well as chemical reactions on PM2.5. We found that unfavorable meteorological conditions accounted for 11.0% and 16.9% of PM2.5 during CNY holidays in 2020 and 2021, respectively. Higher secondary inorganic species were observed after CNY, and K⁺, Cl⁻ showed three prominent peaks which associated closely with fireworks burnings from suburb Beijing and surroundings, verifying that they could be used as two representative tracers of fireworks. Further, we explored the impacts of meteorological conditions, regional transportation as well as chemical reactions on PM2.5. We found that unfavorable meteorological conditions accounted for 11.0% and 16.9% of PM2.5 during CNY holidays in 2020 and 2021, respectively. Higher secondary inorganic species were observed after CNY, and K⁺, Cl⁻ showed three prominent peaks which associated closely with fireworks burnings from suburb Beijing and surroundings, verifying that they could be used as two representative tracers of fireworks. Further, we explored the impacts of meteorological conditions, regional transportation as well as chemical reactions on PM2.5. We found that unfavorable meteorological conditions accounted for 11.0% and 16.9% of PM2.5 during CNY holidays in 2020 and 2021, respectively. Higher secondary inorganic species were observed after CNY, and K⁺, Cl⁻ showed three prominent peaks which associated closely with fireworks burnings from suburb Beijing and surroundings, verifying that they could be used as two representative tracers of fireworks.
and aqueous-phase reaction. Additionally, nostrullary chemistry facilitated the formation of secondary components of both inorganic and organic. This study promotes understandings of PM$_{2.5}$ pollution in winter under the influence of COVID-19 pandemic and provides a well reference for haze and PM$_{2.5}$ control in future.

1. Introduction

Fine Particulate Matter (PM$_{2.5}$, particles with aerodynamic equivalent diameter smaller than 2.5 $\mu$m) and its chemical components are well recognized as an important air pollutant that has adverse impacts on human health (Cohen et al., 2017), visibility (Huang et al., 2014; Luo et al., 2021), and climate change (Feng et al., 2019; Zhang et al., 2020). Heavy pollution episodes which mainly featured with high PM$_{2.5}$ concentrations are frequently occurred during the winter season in Northern and Eastern China. Especially, severe, and long-lasting haze frequently occurred at the beginning of the year in Beijing and its surroundings (Han et al., 2016; Zhang et al., 2019). While starting from the beginning of 2020, the whole country has unexpectedly experienced an emergency lockdown state in order to quickly restrain the spread of 2019 novel coronavirus (COVID-19), and the abrupt reduction in human activities was observed which has profoundly shifted the way of public daily life, and the whole economic and society normal works (Tian et al., 2020; Laughner et al., 2021), and in turn, further alternated the characteristics of PM$_{2.5}$ and its chemical species constitutions (Huang et al., 2020b; Le et al., 2020). Despite all these above notable reductions, severe PM$_{2.5}$ pollution events were still occurred and witnessed during the 2020 extended Chinese New Year (CNY) holidays in Beijing unexpectedly (Chang et al., 2020; Huang et al., 2020b).

Extensive studies have focused on exploring the characteristics of haze at the beginning of the new year in Beijing (Sun et al., 2014; Gao et al., 2015; Shao et al., 2018). With respect to the beginning and the CNY of 2020, some researches have been carried out to explore the impacts of lockdown on PM pollution. Sources of emission (such as vehicular sources) have changed significantly due to the lockdown lowered human activities during CNY in urban Beijing (Liu et al., 2020; Feng et al., 2022). Liu et al. (2020) reported that ambient concentration of NO$_2$ over China has decreased about 48% compared with days before CNY in 2020. Feng et al. (2022) reported that secondary organic aerosol elevated due to NO$_3$ radical nocturnal chemical process during the lockdown in Beijing. Previous studies had also explored the responses to different cities during CNY. Compared with the characteristic of PM$_{2.5}$ pollution in 2019, Chang et al. (2020) reported that the heavy haze during CNY were mainly attributed to the secondary inorganic components originated from regional transportation. Huang et al. (2020b) indicated that secondary aerosol formation was the main factor leading to the heavy haze in eastern China.

Previous studies had focused on PM$_{2.5}$ pollution during the COVID-19 compared with that in the previous year of 2019. However, the impact of the COVID-19 on human activities is hardly temporary and the pandemic has continued for nearly three years, restrictions and epidemic prevention measures became more and more regular. For example, Beijing governments encouraged both local permanent and migrant residents to celebrate the CNY holidays in situ and as far as possible to avoid large volume of personnel gathering and trans-cities travels during the CNY holidays in 2021. As we all known, fine particulate pollutions were not only directly released from various primary emission sources (like coal combustion, industrial manufacturing plants, road transportation, etc.), but also indirectly produced from atmospheric secondary formation, as well as simultaneously affected by meteorological conditions, regional transports and so on (An et al., 2019). However, the resulting reductions in anthropogenic activity due to COVID-19 pandemic represent a passive lowered level of primary emissions that highlights the significance view into the impacts of secondary formation, regional transportation, and meteorological conditions where haze still occurred during CNY holiday periods.

In order to investigate the continuously impacts of COVID-19 on air quality variations during traditional CNY holiday periods of 2020 and 2021 in Beijing, we conducted two intensive observation campaigns at Beijing Normal University (BNU) site by using three online monitoring instruments during 1 January to 16 February 2020 and 19 January to 6 March 2021 (both campaigns lasted in the same Chinese lunar date period). Characteristics of PM$_{2.5}$ and its chemical species were highly focused on to investigate into the similarity and differentiation variations between pre-lockdown and lockdown during CNY holidays. Further, we quantitatively explored the contribution and impacts of meteorological conditions, regional transportation as well as secondary formation on PM$_{2.5}$ pollution. Our results highlight the importance of different roles to key factors to PM$_{2.5}$ pollution formation and provide a reference for future control during winter season, not only confined to CNY in Beijing, but also for some traditional festivals in other cities and countries around the world, such as the grand festivals like Holi and Diwali in South Asia.

2. Materials and methods

2.1. Site descriptions

Our observation site is located on the building roof of the School of Environment (~20 m above the ground) in the central campus of Beijing Normal University (BNU, 116.37° E, 39.96° N). BNU represents a typical urban downtown site, which located between the 2nd ring-road in South and the 3rd ring-road in North, Beijing. Observation campaigns were conducted from 1 January to 16 February 2020 and from 19 January to 6 March 2021, respectively, to explore not only before and after CNY but also before and during the lockdown. The whole observation campaigns were divided into two phases by the corresponding Chinese lunar date: Phase 1 (P1, 1 to 24 January 2020 and 19 January to 11 February 2021, 23-day before CNY), Phase 2 (P2, 25 January to 16 February 2020 and 12 February to 6 March 2021, 23-day after CNY). The geographical location map of sampling site is shown in Fig. S1. Detailed site descriptions are available in our previous studies (Shao et al., 2018; Luo et al., 2021).

2.2. Online monitoring observations

Five water-soluble gases and eight inorganic ions of PM$_{2.5}$ were measured at BNU site by a semi-continuous monitoring instrument for Aerosols and Gases in ambient Air (MARGA 2060; Metrhom Applikon B.V.; Delft, the Netherlands) at hourly resolution. Total ions and gases are listed as following: Mg$^{2+}$, NH$_4^+$, K$^+$, NO$_3^-$, Cl$^-$, SO$_4^{2-}$, Na$^+$, Ca$^{2+}$, NH$_3$, NO$_3$, HONO, SO$_2$, and HCl, respectively. Mass measurements of organic carbon (OC) and elemental carbon (EC) in PM$_{2.5}$ were monitored hourly by a semi-continuous OC/EC analyzer (Model 4; Sunset Laboratory Inc., USA) based on the thermal-optical technique and the NIOSH 5040 protocol (Chang et al., 2017). Organic Matter (OM) can be calculated by 1.6 × OC in Beijing (Turpin and Lim, 2001). Simultaneously, mass concentrations of 19 elements (Al, Si, Zn, V, Ga, Fe, Se, Mn, Pd, Cd, Sb, Pb, Ba, Cu, As, Ni, Cr, Ag, and Co) in PM$_{2.5}$ at BNU site were also measured hourly by using non-destructive X-ray fluorescence from a multi-metal online monitor Xact 625 (Cooper Environmental Services, LLC, USA). More detailed descriptions of Xact 625 can be found in previous study (Zhao et al., 2021). All the online monitoring instruments were configured to collect particulate size <2.5 μm with quality assurance controls. In addition, hourly PM$_{2.5}$ mass concentration data and hourly mass concentration of several criterion air pollutants (PM$_{10}$, CO, SO$_2$, O$_3$, and CO) were downloaded from the official website
of China National Environmental Monitoring Centre (http://106.37.208.233:20035/). Meteorological parameters including relative humidity (RH), air temperature (T), wind speed (WS) and wind direction (WD) were obtained by the Olympic Sports Center monitoring station (39.98° N, 116.40° E) nearby BNU campus (http://data.cma.cn/).

2.3. Data analysis

The concentrations of pollutants are generally affected by both emissions and meteorology. In order to evaluate the contribution of meteorology, machine learning based on RF algorithm was conducted due to its better performance in data set analysis (Vu et al., 2019; Shi et al., 2021). Random forest (RF) is a statistical-based machine learning theory. The overall idea of the RF algorithm includes first predicting the concentration of pollutants based on a set of characteristic values (including meteorological data and other time variables) and training a reliable RF model. Then, the RF model can be used to predict the concentration of pollutants under a series of meteorological conditions, and the relevant average value is called the normalized time series of meteorology. In the algorithm, the concentration of a certain pollutant at a specific time point can be predicted by resampling meteorological data. This process was repeated 1000 times, and 1000 predicted concentrations were averaged to calculate the final weather standardized concentration for that hour, day, and year. In the process, only the weather data, not the time variable, was normalized and resampled from the entire study periods (Breiman, 2001; Lv et al., 2022; Lv et al., 2023).

In this study, the RF model was developed from January to December in 2020 and 2021 independently (Grange et al., 2018; Lv et al., 2022). The estimations of meteorological conditions during the whole observation campaigns were conducted by “normalweather” package in R model (Vu et al., 2019). The de-weathered concentration predicted by the RF model was the meteorological normalized concentration. The model is examined and validated with high R² values, which are generally higher than 0.8. More details about the RF model configuration and performance were shown in supplementary materials.

3. Results and discussion

3.1. Variations of PM$_{2.5}$ and its chemical species during CNY in 2020 and 2021

As shown in Fig. S2, average PM$_{2.5}$ concentrations were 63.83 and 66.86 μg/m$^3$ during the two whole study periods of 2020 and 2021, respectively, which were slightly higher than historical average value (61.60 μg/m$^3$). While, PM$_{10}$ were 75.94 and 78.22 μg/m$^3$ separately lower and higher than historical value (77.91 μg/m$^3$), and notable gaps on the ratio of PM$_{2.5}$/PM$_{10}$ suggesting the different contribution of primary sources and secondary formation mechanism of PM$_{2.5}$ between the two similar period of 2020 and 2021. During the whole observation periods, haze events were obviously observed especially after CNY. To further investigation, the whole observation campaign was divided into two phases, which were called as P1 and P2 in 2020 and the same lunar date in 2021, respectively. It should be noted that P2 in 2020 coincided with first-level emergency response in Beijing (Chang et al., 2020; Le et al., 2020).

Fig. 1 shows the overall variations of meteorological conditions, hourly PM$_{2.5}$ mass concentration and its chemical compositions from 1 January (19 January) to 16 February (6 March) in 2020 (2021). Average PM$_{2.5}$ concentrations showed increase during P1-P2 in both two years, which had increased from 41.39 ± 37.27 and 58.50 ± 46.78 μg/m$^3$ to 86.89 ± 75.90 and 75.57 ± 68.62 μg/m$^3$, respectively. This phenomenon is unexpectedly observed both in 2020 and 2021, which can be attributed to the following reasons: a large volume of population has left from Beijing and returned to their hometown for celebrating CNY holidays during P1; as for 2020, intensive quarantine and lockdown policies were taken to minimize the spread of COVID-19 especially during P2. By contrast, instead of abruptly enforced strict lockdown in 2020, Beijing government encouraged both the local-permanents and external-floating residents to celebrate 2021 CNY holidays in situ, as to minimize the risk of spread of COVID-19 associated with large volume population mitigation among cities. As shown in Fig. S3, P1 exhibited higher mitigation and emigration index in 2020, while P2 exhibited higher human activities in 2021. Unexpectedly increased PM$_{2.5}$ concentrations could be attributed to different levels of...
human activities and other factors (such as weather conditions and regional transportation). Despite the different levels of reductions in primary emissions, haze still showed the occurrence during COVID-19 lockdown both in 2020 and 2021. Overall, the unexpected pollution of PM$_{2.5}$ occurred during CNY holidays in two years, whereas the following variation patterns on mass concentrations and chemical compositions implied that it could be attributed to different reasons.

Chemical compositions of PM$_{2.5}$ are more complex. As presented in Fig. S4, among all the chemical compositions, organic matter (OM) accounted for the major contribution of mass PM$_{2.5}$, average concentrations were 15.31 ± 11.05 and 10.84 ± 7.25 μg/m$^3$ (accounted for 27.0 % and 25.2 %, respectively). Due to the well control of SO$_2$ (precursor of SO$_4$$^{2-}$) emissions from fuel combustion sources in past years, NO$_3$ within 0.45 μg/m$^3$ to 2.88 ± 2.30 μg/m$^3$ has surpassed SO$_4$$^{2-}$ and become the major inorganic species in PM$_{2.5}$ (Fu et al., 2020), followed by SO$_2$$^{2-}$ (10.07 ± 10.61, 8.45 ± 11.26 μg/m$^3$), and NH$_4^+$ (9.68 ± 11.35, 8.64 ± 9.52 μg/m$^3$) in the corresponding period of 2020 and 2021, respectively. Sulfate, nitrate, and ammonium (SNA) presented the dominant components and accounted for 56.6 % and 52.1 % of PM$_{2.5}$ mass concentrations during the whole observation campaigns in 2020 and 2021, respectively, indicating the important role of secondary inorganic species on shaping the situation of PM$_{2.5}$ pollution. Mean concentrations of K$^+$, Mg$^{2+}$, and Ca$^{2+}$ ranged within 0.45–1.97 μg/m$^3$ and collectively accounted for about 5 % of total PM$_{2.5}$ mass, showing higher concentrations during P2 and suggesting the important role of fireworks burning during CNY holidays. Overall, mean mass concentrations of 19 elemental species in PM$_{2.5}$ measured by the Xact 625 monitor were 1.24 ± 1.92 and 2.13 ± 2.73 μg/m$^3$ and accounted for 2.0 % and 5.3 % of the total PM$_{2.5}$ in the corresponding period of 2020 and 2021, respectively.

PM$_{2.5}$ as well as its chemical species showed continuously increases from P1 to P2 in 2020 and 2021. As the major chemical components, OM accounted for 28.9 % (11.73 ± 8.44 μg/m$^3$) and 25.2 % (18.90 ± 12.14 μg/m$^3$) during the two periods in 2020, respectively. Ignoring the decreases of NO$_2$ and SO$_2$ during P2 in 2020, concentrations of NO$_2$ varied from 11.01 ± 9.99 μg/m$^3$ to 28.27 ± 26.48 μg/m$^3$, showing significant increases (from 24.5 % to 29.69 %) during P1-P2. This enhancement exhibited a notably nonlinear relationship between NO$_2$ increase and NO$_2$ precursor reduction and suggesting the role of chemical reactions. Concentrations of SO$_4$$^{2-}$ and NH$_4^+$ also changed from 5.35 ± 5.54 and 5.30 ± 5.78 μg/m$^3$ to 15.05 ± 12.28 and 14.17 ± 13.67 μg/m$^3$, while their contributions only exhibited a little increase from 14.3 % and 11.3 % in P1 to 16.5 % and 13.2 % in P2, respectively. EC was more stable and closely associated with primary emission (Ji et al., 2019; Luo et al., 2021), and its concentrations have changed from 1.59 ± 1.59 μg/m$^3$ to 2.88 ± 2.30 μg/m$^3$, also showing increases from P1 to P2, which could be mainly attributed to pollution accumulation and regional transportation under unfavorable meteorological conditions (Diamond and Wood, 2020). While during FW1 and FW3, the increase of K$^+$ concentration lasted to the following day except insignificant variation during FW2 in 2021. Before FW2 in 2021, we observed high WS from northwest which conducive to the accumulation of pollutants. Since our observation site surroundings hardly existed fireworks burns, pollutants during different FWs mainly came from regional transportation, and there should also be a time lag in the time series of concentration variations. Contributions of K$^+$ and Cl$^-$ were 15 % and 10 %, 7 % and 5 %, 5 % and 5.41 % in the total PM$_{2.5}$ during FW1-FW3 in 2020, respectively, reflecting that the burning quantity of fireworks is largest on the eve night, next by the 5th day and the 15th (Lantern Festival) day. During the three FWs, mass of PM$_{2.5}$ and SNA also increased synchronously, indicating the visible impact of regional transportation for the prohibition of fireworks burning in urban Beijing (Jiang et al., 2015). With respect to the situations in 2021, except the insignificant peak value during FW2, concentrations of K$^+$ and Cl$^-$ showed similar patterns to those in 2020. The insignificant variation could be attributed to the clean air clusters blowing from northeast and northwest before and during FW2 in 2021. Mg$^{2+}$ and Na$^+$ also showed enhancements during the firework burnings intervals, which are closely associated with the combustion sources and chemical components of fireworks.

For the high concentrations of some chemical species during P2, we explored the time series variations of chemical species. Higher concentrations of K$^+$ and Cl$^-$ were observed during P2, which increased from 0.85 (0.81), 2.29 (2.15) μg/m$^3$ during P1 to 3.27 (1.98) and 3.72 (2.72) μg/m$^3$ during P2 in 2020 (2021), respectively. These chemical species considered to be closely related to combustion sources (Luo et al., 2021; Zhao et al., 2021). As shown in Fig. S3, concentrations of K$^+$ and Cl$^-$ exhibited three prominent peaks on the CNY’s eve (24–25 January 2020 and 11–12 February 2021, respectively), the fifth day of CNY (29 January 2020 and 16 February 2021, respectively), and the Lantern Festival (8 February 2020 and 26 February 2021, respectively), representing the three traditional festival days during which Chinese people would focused on setting off a lot of fireworks on a special time intervals generally lasting several hours. Traditionally, most fireworks are highly concentrated setting off on 23:00–1:00 of the eve night to celebrate the starting of CNY, and fireworks are focused on setting off on the morning hours (5:00–10:00) of the fifth day of CNY to celebrate the normally re-opening of restaurant, shopping mall and market, whereas most fireworks are set off on the evening hours (18:00–24:00) of the Lantern Festival for celebrating the end of traditional CNY holidays. It should be noted that setting off fireworks has been forbidden in downtown (inside of 5th ring road) of Beijing city for several years, thus the hourly variations of fireworks tracer elements in our monitoring site at BNU campus are largely influenced by the windblown transportation from suburb Beijing and surrounding provinces. These three enhancement periods were defined as fireworks burning periods (FW1, FW2, and FW3), accompanied with changes in WS and WD, which is consistent with previous studies (Jiang et al., 2015). As shown in Fig. S6, K$^+$ concentration began to increase at about 0:00, reached to the peak value at about 5:00, and recovered to flat at about 12:00 during FW2. While during FW1 and FW3, the increase of K$^+$ concentration lasted to the following day except insignificant variation during FW2 in 2021. Before FW2 in 2021, we observed high WS from northwest which conducive to the accumulation of pollutants. Since our observation site surroundings hardly existed fireworks burns, pollutants during different FWs mainly came from regional transportation, and there should also be a time lag in the time series of concentration variations. Contributions of K$^+$ and Cl$^-$ were 15 % and 10 %, 7 % and 5 %, 5 % and 5.41 % in the total PM$_{2.5}$ during FW1-FW3 in 2020, respectively, reflecting that the burning quantity of fireworks is largest on the eve night, next by the 5th day and the 15th (Lantern Festival) day. During the three FWs, mass of PM$_{2.5}$ and SNA also increased synchronously, indicating the visible impact of regional transportation for the prohibition of fireworks burning in urban Beijing (Jiang et al., 2015). With respect to the situations in 2021, except the insignificant peak value during FW2, concentrations of K$^+$ and Cl$^-$ showed similar patterns to those in 2020. The insignificant variation could be attributed to the clean air clusters blowing from northeast and northwest before and during FW2 in 2021. Mg$^{2+}$ and Na$^+$ also showed enhancements during the firework burnings intervals, which are closely associated with the combustion sources and chemical components of fireworks.

The intensity level of human activity declined due to the impacts of the COVID-19 pandemic, which reduced primary emissions to a certain extent, haze still occurred during the CNY holidays. This unexpected phenomenon could be attributed to complex contributions from other factors such as meteorological conditions, regional transportation, and secondary transformation (An et al., 2019).

3.2. Impacts of disturbance of meteorological conditions on PM$_{2.5}$ during CNY in 2020 and 2021

Haze formation during CNY holidays could be influenced by multiple factors. Firstly, we tried to explore and evaluate the impacts of disturbance meteorological conditions on PM$_{2.5}$ in 2020 and 2021. Meteorological parameters were summarized in Table S2. During the whole periods, average WS and RH in 2020 were 1.86 ± 1.24 m/s and 47 ± 22 %, which were higher than those (1.50 ± 1.16 m/s and 48 ± 24 %) in 2021. While average ambient T in 2020 was −1.86 ± 3.88 °C, which was lower than that
(2.62 ± 6.11 °C) in 2021. Considering the more severe pollution occurred during P2, higher WS, RH as well as higher surface temperature inversion intensity could facilitate transportation of pollutants and formation of secondary chemical components (Li et al., 2021). Discrepancies in meteorological parameters for the study periods in two years may lead to different magnitude of effects on PM$_{2.5}$ concentrations. Hence, in order to quantitatively assess the contributions of disturbance meteorological conditions, we applied the random forest to decouple the weather factors with R model.

As shown in Fig. 2, average PM$_{2.5}$ concentrations by decoupled weather factor were 56.81 and 54.54 μg/m$^3$, which were 7.02 and 11.32 μg/m$^3$ lower than the apparently observed concentrations during the whole observation campaign in 2020 and 2021 respectively. This phenomenon suggested that disturbance meteorological conditions were generally unconducive for pollutants dispersion in Beijing during the two years observation periods, however, showing different situations with respect to P1 and P2 phases, respectively. Compared with whole observed concentrations, the contributions of disturbance meteorological factor accounted for about 11.0 % and 16.9 % in 2020 and 2021, respectively, which were consistent to the results reported in Su et al. (2020). As for P1 and P2, de-weathered PM$_{2.5}$ concentrations in 2020 Beijing were 56.12 μg/m$^3$ and 57.54 μg/m$^3$, respectively. For further analysis, we also evaluated the impacts of disturbance on meteorological conditions before (P1) and after (P2) the CNY. Compared with the observed concentrations, the contributions of meteorological conditions disturbance accounted for −34.3 % and 51.6 % during P1 and P2, respectively. This phenomenon indicated that well favorable meteorological conditions for pollution dispersion during P1 whereas unfavorable meteorological conditions during P2. While in 2021, de-weathered PM$_{2.5}$ concentrations also showed insignificant changes from 53.48 μg/m$^3$ during P1 to 55.47 μg/m$^3$ during P2, compared with the observed concentrations, contributions of disturbance meteorological conditions accounted for −11.0 % and 18.6 %, respectively, both showing a relatively smaller positive or negative contributions compared with those in 2020.

The de-weathered PM$_{2.5}$ concentrations in 2020 still increased from P1 to P2 which could be largely attributed to regional transportation and/or secondary formation (Huang et al., 2020b) under the pre-conditions of significantly lowered human activities compared with P1 before the outbreak of COVID-19. While de-weathered PM$_{2.5}$ concentrations at the corresponding period in 2021 showed insignificant increase, which could be attributed to the relatively stable human activities because that people were suggested to celebrate CNY in situ and as far as possible to avoid trans-city travels by the Beijing Municipal government. In addition, we found that meteorological conditions before CNY could be more conducive to the dispersion whereas relatively unfavorable meteorological dispersion conditions after CNY. This implied that winter meteorological conditions needed to take into more comprehensive consideration on future PM$_{2.5}$ pollution control, especially for the half month following the CNY day with thinking about the notable add-on effects of celebrating fireworks burning.

### 3.3. Impacts of regional transportation on PM$_{2.5}$ during CNY in 2020 and 2021

In addition to local primary emissions, regional transportation is another important factor to PM$_{2.5}$ and haze pollution in megacities. According to Chen et al. (2020), different magnitude of pollutants dispersion could be determined by different WS, when WS was higher than 2 m/s indicating a scale of transportation near surface from surrounding areas. As presented in Fig. 3, PM$_{2.5}$ concentration in 2020 was likely to be affected by regional transportation since its hotspot areas were always associated with high WS, such as southwest (2−3 m/s), northeast and southeast (2−3 m/s), and east (3−6 m/s). Compared with 2020, PM$_{2.5}$ in 2021 were more likely to be affected by local sources during the two whole observation periods, with its hotspot areas mostly ranged within WS < 2 m/s.

During P1 in 2020, PM$_{2.5}$ was slightly affected by potential sources from southeast and southwest. However, with the abrupt decreases of gaseous precursors (SO$_2$ and NO$_2$) during P2 in 2020, the concentrations of NO$_3^−$ and SO$_4^{2−}$ still increased which could be partially attributed to the regional transportation. Because these components were also closely associated fireplace works setting off, and they had shown co-enhancement with characteristic tracer elements (like K$^+$, Mg$^{2+}$, and Cl$^-$) at the same time. The potential origination of regional transportation sources during P2 located in the southwest (such as Baoding, Shijiazhuang (Luo et al., 2021), east and southeast (such as Tangshan and Tianjin) of Beijing. During P1 in 2021, PM$_{2.5}$ was mainly affected by the potential sources from northwest accompanied with high WS ranging from 2 to 8 m/s, and less affected from the direction of southeast and southwest as well as northeast of Beijing with WS ranging about 2 m/s. By contrast, PM$_{2.5}$ during P2 in 2021 was affected by potential sources from southeast and southwest directions with WS about 2 m/s, from which heavier pollution in cities of Hebei provinces (like Langfang, Tangshan, Baoding, Shijiazhuang, etc.) can be transported into Beijing.

In order to further explore impacts of local emission or regional transportation, we separated the whole WS into two ranges, which are WS < 2 m/s and WS ≥ 2 m/s, respectively (Chen et al., 2020). As shown in Fig. S7, impacts of local emissions were evenly distributed in different wind directions in general, while contribution of regional transportations from southwest and southeast played an important role on PM$_{2.5}$ in 2020. In 2021, local emission located in northeast showed prominent effects on PM$_{2.5}$ whereas regional transportations from northwest were also not negligible.

Regional transportation also acted as an important role during firework burning events, since concentrations of K$^+$ were observed to be elevated or decreased with the changes of WD and WS. In 2020, before the first peak value of K$^+$, high WS were observed from southeast and southwest. Then K$^+$ concentration accumulated with low WS to the first peak while decreased along with the WD changing to northeast, and the second peak of K$^+$ concentration accumulated with the decrease of WS from southwest during FW1. Similar to FW1, high WS from northeast and southwest were also observed before the peaks during FW2 and FW3, and K$^+$ values accumulated with low WS and decreased with the WD changed in 2020. The peak values of K$^+$ during the three representing FWs in 2020 suggested the important potential regional transportation sources from southwest and northeast. In 2021, the peak concentration of K$^+$ was accompanied with the change of WS from northeast during FW1. During the FW2 in 2021, there existed an unobvious peak value of K$^+$ indicating the blowing and dispersion role of clean clusters from northeast and northwest. During the FW3, the potential regional sources of K$^+$ were mainly originated from southwest and northeast transportation.
3.4. Impacts of chemical reactions on PM$_{2.5}$ during CNY in 2020 and 2021

In order to explore the impacts of chemical reactions, we further separated P1 and P2 into 7 fractions under different PM$_{2.5}$ concentrations intervals in 2020 and 2021, respectively (Chang et al., 2020). Seven fraction contributions of chemical species under different PM$_{2.5}$ concentrations were shown in Fig. S8 before and after the CNY day. In each fraction of PM$_{2.5}$ pollution, OM and SNA constituted the major components. The contributions of SNA presented more clearly increase from clean to polluted, while the contributions of OM and other primary species showed a decrease trend. Especially during P2 in 2020, due to the emergency lockdown, the contribution of primary species showed significant decreases, while simultaneously secondary species showed significant increases ranging within 82 %–83 %, indicating the prominent role of secondary pollution formation during lockdown. As shown in Fig. S8, the different fraction contributions of OM decreased with the enhancement of PM$_{2.5}$ which were 30 % –28 % (17 %–21 %) during P1, and 35 %–19 % (32 %–15 %) during P2 in 2020 (2021), respectively. While the corresponding contributions of SNA showed increases, indicating the notably important roles of SNA during haze processes (Fu et al., 2020). As shown in Fig. S9, with the enhancement of PM$_{2.5}$ RH and O$_3$ also showed upward patterns which facilitated the formation of SNA by different ways (Zhao et al., 2020). Hence, the high contribution of SNA aerosols to the heavy PM$_{2.5}$ pollution could be the synergetic effects of primary emission, regional transportation, and secondary chemistry reactions.

SNA is found to contribute >50 % to PM$_{2.5}$ mass concentration, to further explore the formation process of secondary inorganic components (Huang et al., 2020a), the ratio of NO$_3^-$/SO$_4^{2-}$ was discussed under different RH and O$_x$ conditions (shown in Fig. 4). Overall, the correlation (R$_2$) of NO$_3^-$ and SO$_4^{2-}$ during entire observation campaigns was 0.82 in 2020 which is relatively higher than that (0.60) in 2021. The strong correlation of NO$_3^-$/SO$_4^{2-}$ indicated that they could be largely emitted from similar sources. When the ratios of NO$_3^-$/SO$_4^{2-}$ were divided into two groups with RH (50 %) as a split line (see Fig. S10), R$_2$ of NO$_3^-$/SO$_4^{2-}$ became 0.88 and 0.78 in 2020 and 2021 separately, showing stronger correlations under low-RH (RH ≤ 50 %) than that (0.73 and 0.47) under high-RH (RH > 50 %). This phenomenon suggested different dominant formation pathways for NO$_3^-$ and SO$_4^{2-}$ during the whole observation campaigns (Huang et al., 2020a; Zhao et al., 2020; Wang et al., 2021). Additionally, very high ratios of NO$_3^-$/SO$_4^{2-}$ (1.99 and 2.68) were found under low-RH conditions in both years. Nevertheless, opposite trends were found when related to O$_3$. The fitted slopes of NO$_3^-$/SO$_4^{2-}$ increased with enhancement of O$_3$. This phenomenon indicated that higher NO$_3^-$ productions were found under RH ≤ 50 % and high O$_3$ concentrations, suggesting photochemical process could acted as a key role in NO$_3^-$ formation while aqueous-phase processes had significant impacts on SO$_4^{2-}$ formation (Duan et al., 2020).

Photochemical oxidation process had been considered to be minor in winter due to the weaker solar radiation and water vapor especially, while several recent studies have reported an increase of photochemical process in winter (Lu et al., 2019; Fu et al., 2020; Ren et al., 2021). With the enhancement of PM$_{2.5}$ during P1-P2, the co-enhancement of O$_x$ was also observed, which indicated the strong photochemistry process (Fig. S9). As shown in Fig. 5, with the decreases of NO$_2$, the significant enhancements of O$_3$ were observed during P2 in both 2020 and 2021. Previous studies have reported that large reduction of NO$_2$ promoted the formation of O$_3$ under Volatile Organic Compounds (VOC)-limited regime in Beijing (Leung et al., 2020). When we further investigate the concentration changes of P2-P1, higher decrease of NO$_2$ were also observed during nocturnal, which facilitated the formation of O$_3$ and oxidation capacity by reducing NO$_2$ titration (NO$_2$ + O$_3$ reactions) (Leung et al., 2020). As we all known, OH radical was one of the key oxidants which also indicated the atmospheric oxidation capacity (Wang et al., 2017), and OH is generally formed by HONO (Fu et al., 2020; Yang et al., 2021). Therefore, we use HONO as a proxy of OH radicals. As shown in Fig. 5, HONO showed a clear diurnal pattern with higher values in the early morning and lower in the evening. The higher increase of HONO concentrations were observed from early morning to about 10:00 during CNY, indicating the reduction of NO$_2$ mitigate OH depletion and in turn to facilitate O$_3$ formation during diurnal. And the enhancement of O$_3$ in day and night will further produce more secondary components in PM$_{2.5}$ (Leung et al., 2020). Simultaneously, the nocturnal OM exhibited prominent enhancement. This phenomenon

![Fig. 3. Polar plots of PM$_{2.5}$ concentrations (unit: μg/m$^3$) during the whole observation campaigns (a, d) and P1-P2 (b, c, e, f) in 2020 and 2021; the gray dash circles represent the WS (m/s).](image-url)
could be attributed to nocturnal chemistry facilitating the formation of secondary organic components (Feng et al., 2022). Due to the abrupt reductions of NO2 and human activities along with the emergency lockdown, prominent increases of pollutants were observed in 2020, indicating the non-linear responses to primary emission and PM2.5 pollution.

In addition, the enhancement of O3 and HONO (OH) further facilitate the formation of NO3⁻. Beijing is thought to subject to the NH3-abundant condition in winter (Huang et al., 2012). The enhancement of NH3 concentrations were observed during P2 in 2020 and 2021 (Fig. S11), which promote the formation of NO3⁻ by HNO3 partitioning (Fu et al., 2020). Due to the enhancement of oxidation capacity by O3 and OH radicals, the formation of HNO3 and NO3⁻ were also increased. The higher increase of HNO3 during nocturnal (shown in Fig. 5) could be attributed to the higher increase of HONO and then hydrolysis by N2O5 consist with higher increase of NO3⁻ (Ren et al., 2021). Higher increase of NO3⁻ in 2020 daytime could also be attributed to regional transportations according to the high WS in daytime. The abrupt reduction of NO2 during lockdown implied that the future scenario for continuously reduction in primary emissions which showed non-linear response to air quality, and need to take synergistic considerations of multiple factors.

4. Conclusions

In this study, by conducting two intensive observation campaigns with three online continuous monitoring measurements of PM2.5, and its chemical species starting from 1 January to 16 February in 2020 and from 19 January to 6 March in 2021 (representing the same Chinese lunar time periods which spanning before (P1) and after (P2) CNY), respectively, we conducted an in-depth investigation on the temporal variations of PM2.5 and its chemical species and revealed the underlined influencing factors with de-weathered machine learning algorithm. Results showed that average PM2.5 concentrations were 63.83 and 66.86 μg/m³ in the whole study periods of two years separately, which were higher than averaged historical values over the same lunar date in 2016–2019. By comparison, PM2.5 showed increases from P1 to P2 in both years. With the significant decrease of SO2 and NO2 due to emergency lockdown, several heavy PM2.5 pollution processes still occurred especially during P2 in 2020, indicating the complex synergetic impacts from meteorological conditions, regional transportation, and chemical reactions. We further assessed the contributions of meteorological conditions by random forest model simulation. We found that unfavorable weathers generally occurred during the two observation periods in 2020 and 2021, and the weather concentration of PM2.5 accounted for 11.0 % and 16.9 %, respectively. Regional transportation from southwest and southeast (south) also played an important role on promoting PM2.5 pollution evolution during the two observation periods in 2020 (2021). Analysis of NO3⁻/SO4²⁻ ratio related to RH and OX indicated that photochemical and aqueous-phase reactions could be the key pathway to NO3⁻ formation and SO4²⁻ formation, respectively. Higher decrease of NO2 facilitated the formation of O3 especially during nocturnal and the enhanced nocturnal chemistry further promoted the formation of both secondary inorganic and organic components of PM2.5.

The impacts of COVID-19 on human health and society activities continues and it also provides a unique opportunity to explore the characteristic of PM2.5. While haze still occurred under visibly lowered human activities. This study implies that although absolutely emission reduction of primary air pollutants from various anthropogenic sources is intrinsically necessary to future continuously mitigation of PM2.5, the synthetically impacts of meteorological conditions, regional transportations as well as atmospheric chemical reactions responses should also be taken into considerations integrally. We believed this study which based on two periods of intensive observation campaigns under the continuously influence.
of COVID-19 pandemic provides a well reference for further promoting pollution control during CNY holidays and winter season, and supply some insights and implication for some traditional festivals in other cities and countries around the world, such as the grand festivals like Holi and Diwali in South Asia.

CRediT authorship contribution statement
Lining Luo: Conceptualization, Sampling, Methodology, Software, Investigation, Writing-Original Draft preparation.
Xiaoxuan Bai: Formal analysis, Writing-Review and Editing.
Yunqian Lv: Formal analysis, Writing-Review and Editing.
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Shuang Zhao: Formal analysis, Visualization.
Yifei Xiao: Validation, Formal analysis.
Hezhong Tian: Conceptualization, Data curation, Formal analysis, Writing - Review & Editing, and Supervision.
All authors contributed to interpretation of the data and provided comments on the manuscript.

Data availability
Data will be made available on request.

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.

Acknowledgements
This work was funded by the National Natural Science Foundation of China (22176014, 21777008, and 51678056). We thank the editors and
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