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Impact of meteorological condition changes on air quality and particulate chemical composition during the COVID-19 lockdown

Jing Ding\textsuperscript{1,3}, Qili Dai\textsuperscript{2,3}, Yafei Li\textsuperscript{2,3}, Suqin Han\textsuperscript{1,3,*}, Yufen Zhang\textsuperscript{2,3,*}, Yinchang Feng\textsuperscript{2,3}

\textsuperscript{1}Tianjin Environmental Meteorological Center, Tianjin 300074, China
\textsuperscript{2}State Environmental Protection Key Laboratory of Urban Ambient Air Particulate Matter Pollution Prevention and Control, College of Environmental Science and Engineering, Nankai University, Tianjin 300350, China
\textsuperscript{3}CMA-NKU Cooperative Laboratory for Atmospheric Environment-Health Research, Tianjin 300074, China

\textbf{A B S T R A C T}

Stringent quarantine measures during the Coronavirus Disease 2019 (COVID-19) lockdown period (January 23, 2020 to March 15, 2020) have resulted in a distinct decrease in anthropogenic source emissions in North China Plain compared to the parallel period of 2019. Particularly, 22.7% decrease in NO\textsubscript{2} and 3.0% increase of O\textsubscript{3} was observed in Tianjin, nonlinear relationship between O\textsubscript{3} generation and NO\textsubscript{2} implied that synergistic control of NO\textsubscript{2} and VOCs is needed. Deteriorating meteorological condition during the COVID-19 lockdown obscured the actual PM\textsubscript{2.5} reduction. Fireworks transport in 2020 Spring Festival (SF) triggered regional haze pollution. PM\textsubscript{2.5} during the COVID-19 lockdown only reduced by 5.6% in Tianjin. Here we used the dispersion coefficient to normalize the measured PM\textsubscript{2.5} (DN-PM\textsubscript{2.5}), aiming to eliminate the adverse meteorological impact and roughly estimate the actual PM\textsubscript{2.5} reduction, which reduced by 17.7% during the COVID-19 lockdown. In terms of PM\textsubscript{2.5} chemical composition, significant NO\textsubscript{3}\textsuperscript{−} increase was observed during the COVID-19 lockdown. However, as a tracer of atmospheric oxidation capacity, odd oxygen (O\textsubscript{3} = NO\textsubscript{2} + O\textsubscript{3}) was observed to reduce during the COVID-19 lockdown, whereas relative humidity (RH), specific humidity and aerosol liquid water content (ALWC) were observed with noticeable enhancement. Nitrogen oxidation rate (NOR) was observed to increase at higher specific humidity and ALWC, especially in the haze episode occurred during 2020SF, high air humidity and obvious nitrate generation was observed. Anomalously enhanced air humidity may response for the nitrate increase during the COVID-19 lockdown period.

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lite images from NASA show how pollution has cleared over China, NO2 emission reduction was particularly remarkable (https://earthobservations.nasa.gov, Airborne Nitrogen Dioxide Plummets Over China). After the extended Spring Festival holiday, industries in China began to resume production in an orderly manner from mid-March.

Many studies have assessed the air quality variation across China during this period, concentrations of different atmospheric pollutants varied with substantial differences. Silver et al. (2020) found that the largest reductions occurred in NO2, with concentrations 27.0% lower on average across China during the lockdown period. Mean PM2.5 and PM10 across China were respectively 10.5% and 21.4% lower during the lockdown period, but there were no significant impacts on O3. Result in work of Kim et al. (2020) also showed that the reduction in NO2 concentrations across China in lockdown period was deeper and longer than in normal years, and PM2.5 reduced by 30% compared with normal years, by contrast, SO2 emissions had not be affected significantly by the pandemic. In Eastern China, satellite observation displayed that CO and NO2 showed the most obvious decrease (20% and 30%), since they were closely associated with energy consumption and transport restrictions (Filonchyk et al., 2020). In the Chinese “epicenter” of COVID-19, Wuhan, compared with the period before the lockdown, NO2 and PM2.5 during the lockdown period decreased by approximately 53.3% and 36.9%, whereas O3 increased by 116.6% (Lian et al., 2020). Apparently, as an indicator of emissions in the transportation sector, large NO2 emission reduction demonstrated that quarantine measures were well implemented.

Although enormous pollutants emission has been reduced, the severe haze episodes still occurred during the lockdown period. The emission reduction control measures were questioned by the public. Actually, in the North China Plain (NCP), pollutants emissions largely exceed the atmospheric environment capacity. Except for SO2, the actual emissions of other pollutants exceeded the environmental capacity by more than 50%, the emission intensity reached twice to five times the national average, and increased by 30% during the heating season (http://www.mee.gov.cn/xzgk2018/). Therefore, haze episodes occur once the adverse synoptic system covers the NCP, which obscures the actual emission reduction effect. Most particularly, the atmospheric pollution sources associated livelihood did not shut down during the COVID-19 lockdown period, especially in northern China, such as heating. Dai et al. (2020) investigated the changes in source contributions to ambient PM2.5 after the outbreak of COVID-19 in Tianjin using the dispersion normalized positive matrix factorization (DN-PMF) model, which can help to reduce the influence of meteorology. Compared to the conventional constrained PMF results, the constrained DN-PMF highlights the contribution of traffic emissions, coal combustion, firework and residential burning to PM2.5.

In addition to the drop in pollutants concentration, the particulate chemical composition has also significantly altered. In Shanghai, Chen et al. (2020) found that PM2.5 reduction was mainly attributed to decreasing concentrations of nitrate. However, another sight showed that much higher secondary aerosol fraction in PM2.5 were observed during the Spring Festival holiday of 2020 (73%) than 2019 (59%) in Shanghai, the synergistic effects of long-range transport and atmospheric chemistry resulted in the efficient conversion of NO2 to particulate nitrate (Chang et al., 2020). With the WRF-Chem simulation, Huang et al. (2020) found that in eastern China, the near-surface O3 was enhanced due to the emission reduction during the COVID-19 lockdown, which enhanced the atmospheric oxidation capacity and further facilitated the secondary aerosol formation. In Beijing, primary aerosol species associated with traffic, cooking and coal combustion emissions during the Spring Festival holiday reduced by 30%-50% on average, whereas secondary aerosol species decreased by a much small part (5%-12%) (Sun et al., 2020). These results preliminarily point out that current emission reduction measures can exert distinct particulate chemical composition, yet in general terms, the reduction measures during the COVID-19 lockdown obviously reduce the primary species but may not suppress secondary aerosol formation efficiently.

In this work, we compared the meteorological parameters, gaseous pollutants and PM2.5 chemical composition during the COVID-19 lockdown period of 2020 (January 23, 2020 to March 15, 2020) with the same period of 2019 in Tianjin, and the possible reasons for the changes were analyzed. Dispersion coefficient normalized PM2.5 concentration was calculated to show the actual reduction during the lockdown period of 2020.

1. Measurements and methodology

1.1. Observation sites and instruments

Tianjin is located in the eastern region of the NCP, and faces Bohai Bay in the east (Fig. S1). The sampling site involving PM2.5 chemical components, as well as meteorological parameters and atmospheric pollutants matched PM2.5 chemical components in the Section 3.3 is the Air Quality Research Supersite at Nankai University (NKUS, 38°59′N and 117°20′E), which is located in the south of Tianjin, is an urban-rural marginal area approximately 20 km from downtown Tianjin and 50 km from Bohai Bay. Vertical meteorological parameters were obtained at the Atmospheric Boundary Layer Observation Station of the China Meteorological Administration, which is located south of Tianjin (39°04′N, 117°12′E).

Ground-based temperature (T), relative humidity (RH), wind speed (WS) and wind direction (WD) were observed by an automatic weather station. Total solar radiation was measured by a pyranometer (CMP21, Kipp & Zonen, Netherlands), respectively. The mixing layer height (MLH) was simulated by Weather Research and Forecasting Model (WRF, V3.8). Vertical distribution of T, RH, WS were measured with an automatic weather station installed on a meteorological tower at 15 platform heights (5, 10, 20, 30, 40, 60, 80, 100, 120, 140, 180, 200, 220 and 250 m). The real-time mass concentration of PM2.5, PM10, SO2, NO2, CO, and O3 used for statistics were obtained from China National Environmental Monitoring Center (http://106.37.208.233:20035/), averaged value of all stations of each city was used for statistics.

Real-time water-soluble ions (SO42-, NO3-, Cl-, NH4+, Na+, K+, Mg2+, Ca2+) in PM2.5 and reactive gaseous precursors (HCl, HNO3, NH3) were measured by online ion chromatography
based studies will be due to the COVID-19 lockdown period, as shown in Fig S2. The predicted and measured \( \text{NO}_2^- \), \( \text{NH}_4^+ \) values are in good agreement, the \( R^2 \) values of linear regressions for \( \text{NO}_2^- \), \( \text{NH}_4^+ \) are all higher than 0.97, and the slopes are approximately 1. It should be noted that when RH is low, ALWC becomes very small, and \( \text{PM}_{2.5} \) pH is subject to considerably more uncertainty. Therefore, only aerosol pH with corresponding RH > 30% was used for statistics.

2. Results and discussion

2.1. Changes in air quality during the COVID-19 lockdown

During the COVID-19 lockdown period in 2020 (January 23, 2020 to March 15, 2020), \( \text{PM}_{10} \), \( \text{SO}_2 \) and \( \text{NO}_2 \) mass concentration in Tianjin and surrounding cities all exhibited a significant drop (Fig 1). By contrast, \( \text{O}_3 \) increased by different degrees, \( \text{O}_3 \) mass concentration in Shijiazhuang, Jinan and Zhengzhou increased by larger than 10%, whereas increased little in Tianjin, Taiyuan and Beijing. Specifically, in Tianjin, \( \text{PM}_{10} \), \( \text{SO}_2 \), \( \text{CO} \) and \( \text{NO}_2 \) mass concentration reduced by 18.3%, 32.7% 17.8% and 22.7%, respectively, and \( \text{O}_3 \) mass concentration only increased by 3.0%. Although massive gaseous pollutants were reduced during the COVID-19 lockdown period, haze episodes still occurred frequently, generating the small part reduction in \( \text{PM}_{2.5} \). Particularly, \( \text{PM}_{2.5} \) in Tianjin reduced by only 5.6%, and even increased by 14.4% in Beijing, indicating that extra factors contributed to the \( \text{PM}_{2.5} \) growth. By comparison, \( \text{PM}_{2.5} \) reduction in Shijiazhuang, Jinan and Zhengzhou was obviously larger than other cities, which may explain the \( \text{O}_3 \) increase in these cities.

In Tianjin, \( \text{NO}_2 \) and \( \text{SO}_2 \) mass concentration all showed a drop at different polluted levels during the COVID-19 lockdown period than the same period in 2019 (Fig S3). On the contrary, \( \text{O}_3 \) displayed a slight increase at all polluted levels during the COVID-19 lockdown period and \( \text{PM}_{2.5} \) was comparable in these two periods. The wind dependent map showed that \( \text{SO}_2 \) in the northeast obviously decreased (Fig S4), which may due to the diminution of pollutant transport in the northeast, such as from Tangshan, a city owns heavy industry. \( \text{NO}_2 \) and CO reduction mainly occurred accompanied by the wind speed lower than 2.0 m/sec, implying that \( \text{NO}_2 \) and CO reduction mainly came from local emission reduction such as vehicle emission.

Lockdown among global cities provided an unprecedented opportunity to investigate environmental restoration process, global analysis in air quality changes in the pandemic has drawn much attention (Table S1). Compared to the same period in 2019, \( \text{NO}_2 \) decrease and \( \text{O}_3 \) increase during the COVID-19 lockdown were reported across most of China (Chu et al., 2021), yet the variation degrees were distinguished. \( \text{NO}_x \) reduction in northern China was less than southern China (Chu et al., 2021; Le et al., 2020; Pei et al., 2020), whereas \( \text{O}_3 \) showed an opposite variation (Chu et al., 2021; Pei et al., 2020). In northern China, emissions from residential heating and major industries were uninterrupted during the COVID-19 lockdown, which may offset the \( \text{NO}_x \) reduction from traffic emission. \( \text{PM}_{2.5} \) also varied by different degrees with much reduction in southern China while less reduction or even increase in northern China (Chu et al., 2021).

1.2. Dispersion coefficient and normalized \( \text{PM}_{2.5} \) concentration

The dispersion coefficient (DC) began to be used to assess air quality changes in the 1970s (Leahey, 1972, Kleinman et al., 1974, 1976). The dispersion coefficient (m²/sec) was defined as the product of the MLH height and the mean wind speed in the MLH (Eq. (1)). Theoretically, the mean wind speed in the MLH should be calculated by integrating wind speed at different heights of the MLH (Huang et al., 2018). In this work, wind speed at the surface was used to replace the mean wind speed in the MLH in the Eq. (1) due to the absence of wind speed at different height. At a given dispersion coefficient, the normalized \( \text{PM}_{2.5} \) concentration can be calculated by Eq. (2) and could be used to assess the impact of dispersion condition on \( \text{PM}_{2.5} \) growth.

\[
\text{DC}_i = \text{MLH}_i \times \text{WS}_i
\]

\[
\text{CDNI}_i = \frac{C_i \times \text{DC}_i}{\text{DC}_{\text{mean}}}
\]

where, \( \text{CDNI}_i \) (\( \mu \text{g/m}^3 \)) and \( C_i \) (\( \mu \text{g/m}^3 \)) is the dispersion coefficient normalized \( \text{PM}_{2.5} \) mass concentration and the measured \( \text{PM}_{2.5} \) mass concentration during period \( i \), DCi (m²/sec) is the corresponding dispersion coefficient during period \( i \), and DCmean (m²/sec) is the average dispersion coefficient for a long period, DCmean during the COVID-19 lockdown period in 2020 and the same period in 2019 was 806 and 862 m²/sec. Noted that surface wind speed and mean wind speed in MLH can generate large discrepancy in dispersion coefficient, however, in Eq. (2), both DCi and DCmean are calculated using the surface wind speed, thus, it will not have a significant impact on the CDNI.

The normalized particulate concentration was originally used to eliminate the meteorological effects, thereby making the relative impact of the various emission sources observable (Kleinman et al., 1976). At polluted period when DCi is low, the measured \( \text{PM}_{2.5} \) mass concentration is generally much high due to the poor dispersion. Thus, we scale the concentrations down to that they would have had if the MLH and wind speed were equal to the mean DC value. During the clean period when DCi is high, the normalized \( \text{PM}_{2.5} \) mass concentration will be scaled upward.

1.3. \( \text{PM}_{2.5} \), pH, and ALWC calculation

In this work, the thermodynamic model ISORROPIA-II (Nenes et al., 1998) running in forward mode was used to calculate ALWC and predict the \( \text{PM}_{2.5} \) pH. More detailed information about \( \text{PM}_{2.5} \) pH prediction can be referred in previous studies (Ding et al., 2019; Guo et al., 2017; Song et al., 2018). Comparison of measured and predicted \( \text{NO}_3^- \), \( \text{NH}_4^+ \) Cl⁻ based on real-time ion chromatography data were showed in
The United States (US), India, and Europe are the regions with much higher COVID-19 infections, recommended rather than mandatory quarantine measures were implemented in these areas. In India, the maximum reduction was in particulate matters (Mahato et al., 2020; Sharma et al., 2020). By the contrary, in US, NO₂ was witnessed the most significant decline, whereas PM₂.₅ was observed with little decrease (Berman and Ebisu, 2020; Zangari et al., 2020), indicating the different atmospheric polluted characters in India and US. Emission reduction in Brazil and Spain was similar with US, CO and NO₂ displayed the most significant reductions, which closely associated with vehicle emission (Baldasano, 2020; Dantas et al., 2020). In areas where mass concentrations of particulate matter are already low, such as US and some Europe countries, it is difficult to largely reduce them even over a long period of lockdown.

As mentioned above, nationwide NO₂ decrease and O₃ increase during the COVID-19 lockdown suggested O₃ formation is highly nonlinear with NO₂. Previous studies generally attributed the anticorrelation between PM₂.₅ and O₃ to the solar radiation variation induced by aerosol radiative effect (Zhang et al., 2015; Wu et al., 2020), and the particle sink for hydroperoxy radicals was also weakened as the PM₂.₅ mass concentration decreased (Li et al., 2019). Moreover, Sicard et al. (2020) and Le et al. (2020) proposed that reduction of fresh NO emissions due to the absence of traffic alleviated O₃ titration, thereby the reduction of NOₓ resulted in the O₃ increase. However, according to the result from Sicard et al. (2020), NOₓ reductions in European cities were comparable to or higher than Wuhan, yet the O₃ reduction was lower than Wuhan, a lower titration of O₃ by NO seems to be contradictory with the observation. The O₃ formation depends on the VOCs-NOₓ ratio, higher VOCs-NOₓ ratio due to the large NOₓ reduction may be the primary reason of O₃ increase. Nationwide O₃ increase and NO₂ decrease implied that O₃ generation in most cities

Fig. 1 – Changes in mean mass concentration of atmospheric pollutants in main cities of North China Plain during the COVID-19 lockdown (2020*) compared to the same period in 2019 (2019*).
is not NOx-limited, synergetic control of NOx and VOCs is needed.

Odd oxygen (O₃ = NO₂ + O₂) was wildly utilized to represent the atmospheric oxidation capacity (Chang et al., 2020; Duan et al., 2020; Ma et al., 2019). As shown in Fig. 2, O₃ during the COVID-19 lockdown was 9.2% averagely lower than the same period in 2019, specifically, almost no difference in O₃ was observed between these two periods when PM₂.₅ ≤ 75 μg/m³. Whereas when PM₂.₅ > 75 μg/m³, O₃ was 19.9% averagely lower during the COVID-19 lockdown, and was much significant in the daytime (21.5%–27.3%). Another key factor in photochemistry, solar radiation, was also observed with no obvious difference during these two periods, and was only slightly lower when PM₂.₅ > 75 μg/m³ in the COVID-19 lockdown. In the case of large drop in NO₂ and little increase in O₃, the atmospheric oxidation capacity did not enhance that much.

### 2.2. Changes in meteorology during the COVID-19 lockdown

Little change in PM₂.₅ mass concentration in Tianjin during the COVID-19 lockdown period was partly attributed to adverse meteorological condition. During the COVID-19 lockdown period, mean MLH in daytime was 647 m, slightly lower than that in the same period in 2019 (691 m) (Fig. 3), whereas nocturnal MLH during the COVID-19 lockdown period was higher than that in the same period in 2019. The frequency of breeze (WS < 1.5 m/sec) during the COVID-19 lockdown period increased by 16.9% while the frequency of WS with 1.5–3.5 m/sec reduced by 12.4%. In addition, WS above 50 m was also lower during the COVID-19 lockdown period, with the mean decrease was 7.8% (Fig. 4). Accordingly, mean dispersion coefficient during the COVID-19 lockdown period (806 m²/sec) was lower than the same period in 2019 (862 m²/sec). The most noticeable change was in RH, ground mean RH during the COVID-19 lockdown period was 62% ± 23%, significantly higher than the 43% ± 22% during the same period in 2019, with the mean increase below 250 m was 49.2% (Fig. 4). The temperature was 9.7% higher during the COVID-19 lockdown period. Higher temperature and RH suggested that the water vapor content was much more abundant during the COVID-19 lockdown period than the same period in 2019, which exerted an impact on the aerosol chemical composition and will be discussed in Section 3.3.

The mean measured PM₂.₅ mass concentration and dispersion coefficient normalized PM₂.₅ (DN-PM₂.₅) mass concentration were 67.5 and 45.6 μg/m³ during the COVID-19 lockdown period (Fig. 5), indicating that unfavorable meteorological condition generated approximately 22 μg/m³ growth of PM₂.₅. By comparison, mean measured PM₂.₅ mass concentration and DN-PM₂.₅ mass concentration was 71.6 and 55.4 μg/m³ during the same period of 2019. DN-PM₂.₅ mass concentration during the COVID-19 lockdown period was 17.7% lower than the same period in 2019, which can roughly reflect the actual emission reduction since the adverse meteorological impact was stripped. Reduction ratio of DN-PM₂.₅ in this work was much close to the result (14% for Tianjin) estimated using the bottom-up inventory model of Multi-resolution Emission Inventory for China (MEIC) (Huang et al., 2020).

### 2.3. Changes in PM₂.₅ chemical composition during the COVID-19 lockdown period

In addition to the changes in gaseous pollutants mass concentration, PM₂.₅ chemical composition also showed an obvious change (Fig. 6a). The most obvious change was the increase of NO₂⁻, mean NO₂⁻ fraction in PM₂.₅ was 20.2% dur-

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**Fig. 2** – Diurnal variation of odd oxygen (O₃) and solar radiation at different PM₂.₅ levels during the COVID-19 lockdown (2020*) compared to the same period in 2019 (2019*).
Fig. 3 – Changes in mixing layer height, wind speed frequency and dispersion coefficient during the COVID-19 lockdown (2020*) compared to the same period in 2019 (2019*).

Fig. 4 – Changes in vertical distribution of temperature (T), relative humidity (RH) and wind speed (WS) during the COVID-19 lockdown (2020*) compared to the same period in 2019 (2019*).

In 2019. By contrast, SO$_4^{2-}$ and Cl$^-$ mass fraction in total ions both dropped during the COVID-19 lockdown period compared to the same period in 2019, with SO$_4^{2-}$ decreased to 21.4% from 25.1%, and Cl$^-$ decreased to 6.5% from 11.6%. Accordingly, NO$_3^-$/SO$_4^{2-}$ ratio (mass ratio) during the COVID-19 lockdown period largely elevated, with median value was 2.3
Fig. 5  - (a), (b) Time series and (c) mean value of measured PM$_{2.5}$ mass concentration (city averaged) and dispersion coefficient normalized PM$_{2.5}$ (DN-PM$_{2.5}$) mass concentration during the COVID-19 lockdown (2020*) and the same period in 2019 (2019*).

Fig. 6  - (a) Mean PM$_{2.5}$ chemical composition during the COVID-19 lockdown (2020*) and the same period in 2019 (2019*). (b) Mean fractional contribution of each ion accounting for total ions, (c) NO$_3^-$/SO$_4^{2-}$ (mass ratio) and (d) PM$_{2.5}$ pH at different PM$_{2.5}$ loading levels during the COVID-19 lockdown (2020*) and the same period in 2019 (2019*). At each PM$_{2.5}$ level, the left column was the data during 2019*, the right column was the data during 2020*.

during the COVID-19 lockdown period, while it was 1.5 during the same period in 2019. SO$_4^{2-}$ and Cl$^-$ are generally recognized as tracers for coal combustion (Bi et al., 2019), similar reduction of SO$_4^{2-}$ and Cl$^-$ demonstrated that the contribution of coal combustion to PM$_{2.5}$ was reduced. In the work of Dai et al. (2020), the contribution of coal combustion estimated from constrained DN-PMF before SF was 37.6% in Tianjin, while it reduced to 12.5% during the SF and 13.3% after the SF.

Differences in PM$_{2.5}$ chemical composition at different polluted levels was further investigated. PM$_{2.5}$ mass concentration was divided into three groups, with PM$_{2.5}$ mass concentration bins were < 75 $\mu g/m^3$, 75–150 $\mu g/m^3$, > 150 $\mu g/m^3$, the corresponding air quality is clean, slightly & moderately pol-
luted and severely polluted. Compared with the same period in 2019, the most obvious \( \text{NO}_3^- \) increase during the COVID-19 lockdown mainly emerged on clean days, while the \( \text{NO}_3^- \) fractional contribution to total ions was slightly higher on polluted days. Differences in \( \text{SO}_4^{2-} \) fractional contribution at different polluted levels between COVID-19 lockdown period and same period in 2019 were obviously lower than \( \text{NO}_3^- \). As a consequence, the \( \text{NO}_3^-/\text{SO}_4^{2-} \) during the COVID-19 lockdown period was obviously higher than the same period in 2019. In contrast, only slight raise in \( \text{O}_3 \) mass concentration on clean days during the COVID-19 lockdown period was observed, and \( \text{O}_3 \) in the daytime was even slightly lower during the COVID-19 lockdown on clean days. Huang et al. (2020) reported that \( \text{O}_3 \) increase can enhance the nighttime \( \text{NO}_2 \) radical and further facilitate secondary aerosol formation. Given the fact that no significant change was observed for \( \text{O}_3 \), the interpretation proposed by Huang et al. (2020) cannot explain the nitrate increase in Tianjin. In summary, if atmospheric oxidation was to be the main factor governing \( \text{NO}_3^- \) production in Tianjin, the \( \text{NO}_3^- \) fractional contribution in \( \text{PM}_{2.5} \) should not increase significantly, but that other factors must have an important effect on the \( \text{NO}_3^- \) formation.

Air humidity may be an important factor on nitrate formation. Specific humidity \( q \), one of the indexes of absolute humidity was calculated by the equation recommended by the extrapolated Wexler’s formula (Wexler, 1976; Bolton, 1980). As shown in Fig. 7a, both specific humidity and aerosol liquid water content (ALWC) increased during the COVID-19 lockdown, with mean value was \( 2.8 \pm 1.1 \text{ g/kg} \) and \( 40 \pm 64 \mu \text{g/m}^3 \), respectively, comparing to the \( 1.9 \pm 1.0 \text{ g/kg} \) and \( 18 \pm 30 \mu \text{g/m}^3 \) during the same period of 2019. Correlation between nitrogen oxidation rate (\( \text{NOR} = n[\text{NO}_3^-]/[n[\text{NO}_3^-] + n[\text{NO}_2^-]) \)) sulfur oxidation rate (\( \text{SOR} = n[\text{SO}_4^{2-}]/[n[\text{SO}_4^{2-}] + n[\text{SO}_2^-]) \)) and \( \text{O}_3 \), \( \text{NO}_2 \) (or \( \text{SO}_2 \)), \( \text{T} \), \( q \), ALWC was examined, respectively (Figs. S5 and S6). We found that during the same period in 2019, NOR responded to \( \text{O}_3 \), \( q \), and ALWC more obviously, higher NOR was observed accompanied by higher \( \text{O}_3 \), \( q \) and ALWC. Nevertheless, NOR correlated well with only \( q \) and ALWC during the COVID-19 lockdown period, higher NOR occurred even with low \( \text{O}_3 \). At a given \( \text{O}_3 \), NOR raised significantly with increasing \( q \) and ALWC (Fig. 8) during these two periods, whereas at a constant \( q \) or ALWC, elevation of \( \text{O}_3 \) did not enhance the NOR significantly during the COVID-19 lockdown. Compared with NOR, SOR has a stronger dependency on ALWC, which is in line with mainstream understanding of the sulfate formation that is aqueous reaction is the main pathway for sulfate generation. Responses of SOR to \( \text{O}_3 \), \( q \) and ALWC were much similar with NOR (Fig. S7). Collectively, these results demonstrated that anomalously high air humidity played a key role in nitrate formation during the COVID-19 lockdown.

Formulation mechanism of nitrate has drawn much of concern in recent years, yet the explicit pathways for nitrate formation were still controversial. Current consensus about nitrate formation was that: in the daytime, photochemistry dominates the formation of nitrate. Recently, several works have demonstrated that photochemistry was active in winter polluted condition (Fu et al., 2020; Lu et al., 2019; Tan et al., 2018), \( \text{O}_3 \) and \( \text{OH} \) productions are sufficiently high to facilitate fast gas-phase and heterogeneous conversion of \( \text{NO}_x \) to nitrate. In the nighttime, \( \text{N}_2\text{O}_5 \) heterogeneous hydrolysis is an important source of particulate nitrate (Wang et al., 2017a,b; Wang et al., 2018). Liu reported that in NCP, \( \text{N}_2\text{O}_5 \) heterogeneous hydrolysis dominates the nighttime \( \text{HNO}_3 \) production (83.6%) and also contributed 10.1% of \( \text{HNO}_3 \) production during daytime (Liu et al., 2020). Air humidity can affect nitrate formation by both photochemistry and aqueous pathway. On the one hand, \( \text{O}_3 \) photoysis \( (\text{O}_3 + \text{H}_2\text{O} \rightarrow 2\text{OH}) \) can contribute to the addition of \( \text{OH} \), thereby the raise in specific humidity during the COVID-19 lockdown may increase the \( \text{OH} \) concentration in the atmosphere. On the other hand, increased RH during the COVID-19 lockdown period resulted in the elevation of ALWC, thereby facilitated the nitrate formation though aqueous reaction.

Increased ALWC induced by 1 \( \mu \text{g/m}^3 \) water-soluble salts (donated as ALWC/Ion\(_{\text{mass}}\)) aided more hydrogen ion (H\(_{\text{air}}^-\)) was released, thereby enhances the aerosol acidity (Fig. S8). Mean \( \text{PM}_{2.5} \) pH during the COVID-19 lockdown period was 4.5 ± 0.4, slightly lower than the 4.8 ± 0.6 during the same period in 2019. Reaction rates of \( \text{SO}_4^{2-} \) production though aqueous oxidation pathways depend on the initial aerosol acidity. Sulfate production under acidic conditions is largely limited by the amount of \( \text{SO}_2 \) that can partition to the aqueous phase (Pye et al., 2020; Seinfeld and Pandis, 2016). \( \text{PM}_{2.5} \) pH drop during the COVID-19 lockdown period may inhibit the reaction rate of \( \text{SO}_4^{2-} \) production more or less. Considering the competition between sulfate and nitrate formation for ammonia, once the sulfate formation is suppressed, the nitrate formation will be conspicuous. Aerosol pH has been reported as a major factor affect the partitioning of \( \text{HNO}_3 \rightarrow \text{NO}_3^- \), at
Fig. 8 – Nitrogen oxidation rate (NOR) at different levels of odd oxygen \( (O_3) \), temperature \( (T) \), specific humidity \( (q) \) and aerosol liquid water (ALWC). The upper and under row is the result during the COVID-19 lockdown (2020*) and the same period in 2019 (2019*), respectively.

A constant ALWC, more \( NO_3^- \) was measured at higher pH (Guo et al., 2017), similar results were also found in Tianjin of China (Shi et al., 2019).

2.4. Changes in pollution characters during the SF

Setting off fireworks in some northern cities are not strictly regulated, instantaneous large emission superimposes adverse meteorological conditions will generate regional air pollution. During the 2020SF, atmospheric dispersion condition was much worse than that in 2019 SF. In Tianjin, mean WS and MLH in daytime in 2020 SF was 0.8 m/s and 443 m, respectively, comparing to that 1.9 m/s and 647 m in 2019 SF, atmospheric stratification tended to be more stable, which was extraordinarily conducive to the persistent of air pollution. The polluted period (period wherein \( PM_{2.5} \) mass concentration was continuously larger than 75 \( \mu g/m^3 \)) in 2020SF and 2019SF lasted for 88 and 24 h, respectively, with mean \( PM_{2.5} \) mass concentration was 186 ± 61 \( \mu g/m^3 \) (peak: 333 \( \mu g/m^3 \)) and 189 ± 76 \( \mu g/m^3 \) (peak: 369 \( \mu g/m^3 \)). The averaged T, RH and q was 1.0 °C, 72% and 2.8 k/\( kg \) in 2020SF; comparing to that −3.2 °C, 44% and 1.4 k/\( kg \) in 2019SF (Fig. 9), which was closely associated with secondary aerosol formation.

Compared to the sharp raise and drop of \( PM_{2.5} \) on New Year’s Eve of 2019 (4 February, 2019), no obvious \( PM_{2.5} \) increase was observed on 2020 New Year’s Eve (24 January 2020), the air quality deteriorated begin from noon of 25 January 2020 and the increase tendency was much mild (Fig. 9). In early morning of 25 January, 2020, \( PM_{2.5} \) mass concentration in large area was higher than 500 \( \mu g/m^3 \), especially in northeastern China (Fig. S9), the simultaneous air quality in Tianjin, however, was clean. Fig. S10 showed that air masses arriving in Tianjin had passed the northeastern China, affected by the pollutants transport, air quality in Tianjin deteriorated subsequently. Apparently, the \( PM_{2.5} \) increase on 2019 New Year’s Eve was mainly attributed to the local fireworks setting offs, whereas the air pollution on the following days after 2020 New Year’s Eve was mainly affected by the regional transport.

In the 2019SF, \( PM_{2.5} \) was dominated by OM, \( SO_4^{2-} \), \( NO_3^- \), \( Cl^- \) and EC, average fractional contribution of \( SO_4^{2-} \), \( NO_3^- \), \( Cl^- \) in total ions was 29.7%, 25.6% and 14.6%. Whereas in the 2020SF, main contributors to \( PM_{2.5} \) were \( NO_3^- \), OM, \( SO_4^{2-} \), \( Cl^- \) and EC, average fractional contribution of \( SO_4^{2-} \), \( NO_3^- \), \( Cl^- \) in total ions was 43.7%, 22.9% and 10.7%. \( PM_{2.5} \) pH in 2019SF and 2020SF was 5.5 ± 1.0 and 4.3 ± 0.4. Considering the much long-lasting air pollution in 2020SF, here, we more focus on the analysis of \( PM_{2.5} \) chemical characters in 2020SF.

As a nonreactive gas, the variation of \( SO_4^{2-}/CO \), \( NO_3^-/CO \) and \( Cl^-/CO \) can be utilized to indicate the relative increasing rate of \( SO_4^{2-} \), \( NO_3^- \) and \( Cl^- \) compared to CO. 5-hour moving average value of \( SO_4^{2-}/CO \), \( NO_3^-/CO \) and \( Cl^-/CO \) was used to eliminate the short-term meteorological disturbance, denoted as \( SO_4^{2-}/CO_5h \), \( NO_3^-/CO_5h \) and \( Cl^-/CO_5h \). During the first accumulation stage (AS-I) of 2020SF (2020/1/25 00:00-2020/1/26 12:00), \( SO_4^{2-}/CO_5h \), SOR, and contribution of \( SO_4^{2-} \) in total ions all exhibited a steady increase (Fig. 9). In the meanwhile, \( Cl^-/CO_5h \) and contribution of \( Cl^- \) also showed an obvious upward tendency. In contrast, \( NO_3^-/CO_5h \) only slightly increased and NOR increased to 0.36 from 0.15. \( NO_3^- \) contribution in ions was observed to keep constant compared to the obvious \( SO_4^{2-} \) increase, implying that sulfate raised...
Fig. 9 – Time series of (a,b) T, RH, WS, specific humidity (q), solar radiation (SR), and (c–e) mass concentration of atmospheric pollutants including SO\(_2\), NO\(_x\), O\(_3\), CO, PM\(_{2.5}\) as well as SOR, NOR. (f) 5-hour moving average value of SO\(_2\)/CO, NO\(_x\)/CO and Cl\(^-\)/CO, (g) mass concentration of PM\(_{2.5}\) chemical components in 2019 Spring Festival (2019/2/4–2019/2/10) and 2020 Spring Festival (2020/1/24–2020/1/29).

faster than nitrate. In the light of the obvious pollution regional transport induced by fireworks, the growth of sulfate may result from both the primary emission and aged process during the transport.

During the second accumulation stage of 2020SF (AS-II, 2020/1/25 20:00–2020/1/28 10:00), water vapor content was obviously higher than AS-I, the specific humidity and RH was higher than 3 g/kg and 85%, respectively, resulting in the ALWC increase (112 ± 45 μg/m\(^3\)) compared to the 80 ± 66 μg/m\(^3\) during AS-I. NO\(_3\)\(^-\)/CO\(_5h\) was observed to steadily step up, NOR further increased to 0.43, NO\(_3\)\(^-\) fractional contribution to total ion climbed to 49.0% from 30.0%, demonstrating the efficient conversion of nitrate formation, yet no obvious diurnal variation pattern was observed. If we scale the hourly variation rate of NO\(_3\)\(^-\) mass concentration to the CO variation rate, approximately 25% NO\(_3\)\(^-\) growth was attributed to the secondary transformation during the AS-II, which reached 40% in the early morning of 28 January, 2020, wherein the corresponding NO\(_2\) mass concentration, ALWC and specific humidity were all higher. Therefore, higher air humidity may elevate the contribution of aqueous reaction to nitrate formation in AS-II. SO\(_2\)\(^2-\)/CO\(_5h\) during the AS-II stage was observed to change slightly, variation of Cl\(^-\)/CO\(_5h\) was much similar to the SO\(_2\)\(^2-\)/CO\(_5h\). Average fractional contribution of SO\(_2\)\(^2-\) in total ions was 25.4% ± 1.5%, the smaller standard deviation also indicated a mild change in SO\(_2\)\(^2-\), implying that the growth of SO\(_2\)\(^2-\) mass concentration largely resulted from the accumulation.

3. Conclusions

During the COVID-19 lockdown (January 23, 2020 to March 15, 2020), PM\(_{10}\), SO\(_2\), NO\(_2\) reductions in NCP was much pronounced than PM\(_{2.5}\) and CO, whereas O\(_3\) showed an upward tendency in varying degrees. Specifically, PM\(_{10}\), SO\(_2\), NO\(_2\), CO
mass concentration in Tianjin reduced by 18.3%, 32.7%, 22.7% and 17.8% during the COVID-19 lockdown period compared to the same period in 2019. In contrast, PM$_{2.5}$ mass concentration only reduced by 5.6% and O$_3$ increased by 3.0%. Slight O$_3$ increase and NO$_2$ decrease implied that O$_3$ generation may be VOCs-limited in Tianjin, synergetic control of NO$_x$ and VOCs is needed. The deteriorating meteorological condition during the COVID-19 lockdown period obscured the actual reduction of PM$_{2.5}$. Compared to the 2019SF, air quality in 2020SF of Tianjin was much deteriorated due to the stable meteorological conditions and regional transport induced by fireworks. Here the dispersion coefficient was used to normalize the measured PM$_{2.5}$, aiming to eliminate the adverse meteorological impact and roughly estimate the actual PM$_{2.5}$ reduction, the normalized PM$_{2.5}$ reduced by 17.7% during the COVID-19 lockdown period.

As a tracer of atmospheric oxidation capacity, O$_3$ during the COVID-19 lockdown was 9.2% averagely lower than the same period in 2019, and it was 19.9% averagely lower at polluted condition. Solar radiation was observed with no obvious difference during these two periods. However, air humidity elevated notably, mean RH, specific humidity and ALWC was 62%, 2.8 ± 1.1 g/kg and 44 ± 66 μg/m$^3$, respectively, increased by 44%, 47% and 144% than the same period in 2019. Accordingly, PM$_{2.5}$ composition during the COVID-19 lockdown period was characterized by the increased secondary species in aerosols. NO$_3^-$ accounted for 20.2% in PM$_{2.5}$ during the COVID-19 lockdown period compared to the 14.5% in the same period of 2019, mass fractional contribution of SO$_4^{2-}$ total ions also increased. Whereas OM, EC, Cl$^-$ all showed a drop tendency. NOR was observed to increase at higher specific humidity or ALWC, anomalously high air humidity facilitated nitrate formation. In the 2020SF haze episode, high air humidity and obvious nitrate generation was observed. The increased air humidity may facilitate nitrate formation though aqueous reaction. Moreover, increased ALWC lowered PM$_{2.5}$ pH, which may inhibit the production rate of sulfate formation and thus favored the formation of nitrate.

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Appendix A. Supplementary data

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2021.02.022.

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