Puzzling Haze Events in China During the Coronavirus (COVID-19) Shutdown

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Abstract It is a puzzle as to why more severe haze formed during the New Year Holiday in 2020 (NYH-20), when China was in an unprecedented state of shutdown to contain the coronavirus (COVID-19) outbreak, than in 2019 (NYH-19). We performed a comprehensive measurement and modeling analysis of the aerosol chemistry and physics at multiple sites in China (mainly in Shanghai) before, during, and after NYH-19 and NYH-20. Much higher secondary aerosol fraction in PM$_{2.5}$ were observed during NYH-20 (73%) than during NYH-19 (59%). During NYH-20, PM$_{2.5}$ levels correlated significantly with the oxidation ratio of nitrogen ($r^2 = 0.77$, $p < 0.01$), and aged particles from northern China were found to impede atmospheric new particle formation and growth in Shanghai. A markedly enhanced efficiency of nitrate aerosol formation was observed along the transport pathways during NYH-20, despite the overall low atmospheric NO$_2$ levels.

Plain Language Summary In China, there are multiple cases (e.g., the 2008 Summer Olympics in Beijing and the 2010 World Expo in Shanghai) when combustion-related emissions (e.g., NO$_x$) were actively, and successfully, reduced to transiently improve air quality. During the extended Chinese Lunar New Year holiday in 2020 (between 24 January and 10 February), whole China was in an unprecedented state of shutdown, because most people were contained in their homes to reduce the spread of the novel coronavirus disease (COVID-19). Mobility, energy demand, and industrial output remained far below their normal levels. Nevertheless, widespread haze pollution still occurred over Eastern China. To elucidate haze formation mechanisms, we performed comprehensive and continuous measurements of aerosol chemistry and physics in and out of Shanghai before, during, and after the Chinese New Year Holiday in 2019 and 2020, respectively. We argue that the synergistic effects of long-range transport and atmospheric chemistry leading to the efficient conversion of NO$_x$ to particulate nitrate were the key of haze formation during the Chinese New Year Holiday of the COVID-19 outbreak in Shanghai.

1. Introduction

In the first half December 2019, several cases of pneumonia with unknown etiology were reported in Wuhan City, Hubei Province, China (Wu, Zhao, et al., 2020; Zhu et al., 2020). On 7 January 2020, a novel coronavirus called 2019-nCoV was identified as the cause of such pneumonia by the Chinese Center for Disease Control (Huang, Wang, et al., 2020; Wu & McGoogan, 2020). The World Health Organization (WHO) declared the outbreak to be a Public Health Emergency of International Concern and on 11 February 2020 announced “COVID-19” to be the official name of the novel coronavirus disease (Sohrabi et al., 2020). On 23 January, a day before the Lunar New Year of 2020, China’s central government imposed a lockdown in Wuhan and other cities in the Hubei province in an effort to quarantine the epicenter of the COVID-19 outbreak (Wu, Gamber, & Sun, 2020). However, the epidemic outbreak in the Hubei province happened concurrent with the Spring Festival travel rush, with ~5 million people migrating to other regions, just before the lockdown of Wuhan, aiding the spread of COVID-19 over most of China (Chen et al., 2020).

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Facing the great challenge of the fast-spreading virus, all local governments raised their infectious disease response activities to the highest alert levels. The 2020 Chinese New Year Holiday (NYH-20) was planned originally for the week from 24 to 30 January. To better control the epidemic, and to extenuate the transportation demands, China expanded NYH-20 to 10 February (18 days totally) (China, 2020). Quarantines, blocked roads, and checkpoints throughout the country’s urban and rural areas forced most people to stay at home. The measures taken during NYH-20 were, and still are, dramatically affecting the country’s economy. The energy demand and industrial output remained far below the average levels of the same periods in preceding years. For example, energy production by coal-fired power plants was reduced by one third. Oil refineries and steel industries reached the lowest activity level of the past 5 years. Also, vehicular transportation and domestic flights were reduced by >70% during NYH-20.

The reduction in industrial coal and oil combustion was associated with a significant decrease in the emissions of combustion-derived air pollutants (e.g., NOx and SO2). Quite surprisingly, severe haze pollution with high PM2.5 mass concentrations were nevertheless observed over Eastern China (Wang, 2020). As one of the largest megacities in China, Shanghai pioneered the implementation of a series of control measures against the spread of infections during the COVID-19 outbreak in early 2020 (Perman, 2020), providing an unprecedented opportunity to understand this haze paradox, that is, the elevation of atmospheric PM2.5 concentrations despite lower emissions. In this study, we present results from analyses of aerosol chemistry and physics performed at an urban site in Shanghai before, during, and after the New Year Holiday in 2019 (NYH-19) and 2020 (NYH-20), and we compared the chemical composition and formation processes of the aerosols, as well as meteorological conditions, between the two periods in order to gain clues on the haze event puzzle in NYH-20.

2. Materials and Methods

2.1. Site Description

The Pudong site (PD; 31.233°N, 121.545°E; 15 m a.g.l) represents an urban setting in central Shanghai (Chang et al., 2016). The measurements were conducted before (from 12 January to 3 February 2019 and from 1 January to 23 January 2020), during (from 4 February to 10 February 2019 and from 24 January to 10 February 2020), and after (from 11 February to 26 February 2019 and from 11 February to 26 February 2020) the New Year Holidays in 2019 (NYH-19) and 2020 (NYH-20). During NYH-20, additional, more sporadic, measurements conducted at other sampling sites were included to support our main campaign at PD: Dongtang Eco-park (DT; 31.524°N, 121.958°E) and Dianshan Lake (DSH; 31.096°N, 120.988°E) in rural Shanghai, Caocangmen in urban Nanjing City (32.057°N, 118.749°E; Jiangsu Province), Shanxi in rural Jiaxing City (30.835°N, 120.899°E; Zhejiang Province), and the Tieta branch of the Institute of Atmospheric Physics in urban Beijing City (39.974°N, 116.371°E). Detailed site descriptions are available in our previous work (Chang et al., 2020; Jiang et al., 2015).

2.2. Field Measurements

Inorganic components of PM2.5 (NH4+, NO3−, SO42−, Cl−, Na+, K+, Ca2+, and Mg2+) and water-soluble gases (NH3, SO2, HCl, HONO, and HNO3) were assessed at the sites in Shanghai, Jiaxing, and Nanjing using a semi-continuous Monitoring device for AeRosols and Gases in ambient Air (MARGA 2060; Metrohm Applikon B.V.; Delft, the Netherlands) at hourly time resolution (Chang et al., 2020). At the PD site in Shanghai, PM2.5 inorganic species, NH3 concentrations, temperature, and relative humidity (RH) were used to calculate the aerosol water content (AWC) using the ISORROPIA-II model (Fountoukis & Nenes, 2007). In Beijing, an Aerodyne Time-of-Flight-Aerosol Chemical Speciation Monitor (ToF-ACSM) was used, equipped with a PM2.5 aerodynamic lens and capture vaporizer that can measure nonrefractory PM2.5 (NR-PM2.5) aerosol species in real-time, including organics, NH4+, NO3−, SO42−, and Cl− (Sun et al., 2020). Simultaneously, mass concentrations of organic carbon (OC) and elemental carbon (EC) in PM2.5 at PD were measured hourly, using a semi-continuous OC-EC analyzer (model RT-4; Sunset Laboratory Inc.; Tigard, USA) based on the thermal-optical technique and the NIOSH 5040 protocol (Chang et al., 2017). Organic matter (OM) was found to have an average OM/OC ratio of 1.5 in Shanghai (Huang, He, et al., 2012). The Minimum R Squared (MRS) method was used to estimate secondary organic carbon (SOC) concentrations based on the measured OC and EC data (Wu & Yu, 2016). Besides, mass concentrations of 19 elements (Si, Fe, K, Ca, Zn, Mn, Pb, Ba, V, Cu, Cd, As, Ni, Cr, Ag, Se, Hg, Co, and Au) in PM2.5 at PD were
determined every hour by a multi-metal monitor (Xact™ 625; Cooper Environmental Services, LLT; Portland, USA) (Chang et al., 2018). To investigate atmospheric new particle formation at DSH and DT, two instruments, one nano-scanning mobility particle sizer (nano-SMPS), and one long-SMPS were operated simultaneously at each site to measure number size distributions of 3–736 nm particles at a time resolution of 5 min (Yao et al., 2018). The performance/accuracy of these online instruments was validated in our previous work (Chang et al., 2016, 2017, 2018, 2019).

Additional measurements included the bulk PM$_{2.5}$ mass concentrations using a tapered element oscillating microbalance (TEOM) and a filter dynamics measurement system (FDMS) (TEOM-FDMS 1405-F; Thermo Fisher Scientific™; Waltham, USA). RH, air temperature ($T$), atmospheric pressure, and wind speed and direction were recorded by a series of Vaisala® weather sensors (WXT530 Weather Transmitter Series; Vaisala®; Vantaa, Finland) at 10 min time resolution. The acquired original backscatter signals from ceilometer (CL51; Vaisala®; Vantaa, Finland) measurements were used to retrieve the cloud-base and planetary boundary layer height (PBLH) up to 15.4 km, at a time and space resolution of ~10 s and 10 m, respectively (Peng et al., 2017; Zhang et al., 2012). It needs to be noted that the PBLH values measured with the CL51 ceilometer in our study are relatively low, likely due to high aerosol loadings that affect the backscatter signals (Tang et al., 2015). As detailed in the supporting information Text S1, large biases have been found for PBLH values measured by different methods, including radiosonde, ground-based lidar, and Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) (Guo et al., 2019; Ho et al., 2015; Kim et al., 2008; Korhonen et al., 2014; Liu et al., 2015, Liu et al., 2019; Su et al., 2018; Zhang et al., 2016).

### 2.3. Air Quality Modeling

Air quality in Shanghai and the surrounding Yangtze River Delta region during 1 January to 29 February 2020 was simulated using the Community Multiscale Air Quality Model (CMAQ) version 5.2. The anthropogenic air pollutant emissions were estimated by the Shanghai Academy of Environmental Sciences, and the emission reductions during the lockdown period were estimated using the online continuous stack emission monitoring data in Shanghai. Meteorological conditions were simulated with the Weather Research and Forecasting (WRF) Model version 4.0. The horizontal grid resolution was 4 km. The model configurations of WRF and CMAQ, respectively, were equivalent to the ones used in the study by Hu et al. (2016).

### 3. Results and Discussion

Time series data of major chemical components of PM$_{2.5}$ and their fractional contribution before, during, and after NYH-19 and NYH-20 are shown in Figure 1. The average PM$_{2.5}$ concentrations decreased from 55.1 µg m$^{-3}$ before NYH-19 to 29.8 µg m$^{-3}$ during NYH-19, and then back to 42.2 µg m$^{-3}$ after NYH-19 (Figure 1a). This temporal pattern is expected, as it is consistent with the typical fluctuation of energy demand before, during, and after the New Year Holidays. China relies on coal for over 60% of its energy consumption (Tang et al., 2019). The Chinese New Year Holidays represent officially a 1-week national celebratory event. However, coal consumption typically starts to decline before the Chinese New Year, generally reaching its minimum during the holiday period before returning to its peak around half a month after the weeklong break (Huang, Zhuang, et al., 2012). In early 2020, the measures taken to minimize the spread of COVID-19 during NYH-20 have resulted in unprecedented and longer-lasting reductions in terms of social movement, transportation, energy demand, and emissions. According to the urban travel intensity determined in Shanghai, the pre-holiday travel activity in 2020 was 26% higher than the year before but was more than 50% lower after the New Year of 2020 (Figure S1). The nationwide reduction in automotive mobility and energy production following the COVID-19 outbreak was accompanied by plummeting atmospheric NO$_2$ levels. More precisely, satellite observations show that NO$_2$ emissions during the economic slowdown were reduced all over China to a record low, that is, not only with respect to the pre-holiday period but also when compared to the same holiday week period following the 2019 Lunar New Year (Figure S2), which was consistent with the result of surface NO$_2$ measurement (Figure S3).

The decline in NO$_2$ levels clearly coincided with the large reduction of coal consumption. NO$_2$ is a key contributor to smog and haze in the atmosphere. Therefore, if NO$_2$ emissions were to be the main factor governing air quality, the PM$_{2.5}$ concentrations during NYH-20 should also have been at minimum levels, yet this was not observed. Most curiously, frequent PM$_{2.5}$ spikes were observed during NYH-20 despite lower NO$_2$
and other emissions from industry and vehicles, and the overall PM$_{2.5}$ level during NYH-20 was more than 2.5 times higher than during NYH-19 (excluding a 12-hr episode during the eve of Chinese New Year in 2019) (Figure 1). Moreover, such apparently paradox haze events during NYH-20 also emerged in other megacities like Beijing (Wang, 2020).

In order to understand the mechanisms and conditions that may have resulted in the observed unusual low-emission/high-PM$_{2.5}$ scenario, we first compared the meteorological parameters during NYH-19 and NYH-20 and tested whether the meteorological conditions during NYH-20 were more conducive to haze pollution. PBLH plays a vital role in determining the vertical dispersion of air pollutants that are emitted from the Earth’s surface (Su et al., 2018). We observed one period of high PM$_{2.5}$ pollution (PM$_{2.5}$ > 75 μg m$^{-3}$) during NYH-19, and three of these events during NYH-20. The average PBLH (mean ± 1σ) for the three haze events during NYH-20 were 565 ± 189 m (from 4:00 [28 January] to 12:00 [30 January]), 565 ± 91 m

![Figure 1](image-url). Time series of the major chemical components of PM$_{2.5}$ and fractional contributions before, during, and after the New Year Holiday in 2019 (a) and 2020 (b) measured at the Shanghai station PD. The numbers in the single panels (in μg m$^{-3}$) refer to the average PM$_{2.5}$ concentrations measured during each period. The fractional contributions as a function of the PM$_{2.5}$ bins before, during, and after NYH-20 are also shown (c). Note the uniform increase in the relative abundance of secondary aerosols with the increase of PM$_{2.5}$. 

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As discussed above, the SOA concentrations during NYH-19 (528 ± 87 m; from 0:00 to 10:00 [4 February]). Similar to the observations by Liu et al. (2019), the PM$_{2.5}$ concentrations were weakly anti-correlated with PBLH (see Figure S4). Yet, as discussed below, PM$_{2.5}$ concentrations can be, and likely were, also affected by regional transport and secondary aerosol formation. Other parameters (e.g., $T$ and RH) indicate that the overall meteorological conditions during NYH-19 (6.5 ± 3.8°C; 80.5 ± 12.2%) were also similar to those during NYH-20 (6.7 ± 3.0°C; 75.8 ± 22.1%). As becomes evident in Figure S5, there was no significant correlation between wind speed and PM$_{2.5}$ mass concentration, neither during NYH-19 nor during NYH-20. Therefore it is reasonable to conclude that wind speed is not a determinant factor to influence PM$_{2.5}$ pollution. As a consequence, we argue that it was not the meteorology that can explain the observed differences in haze pollution between NYH-19 and NYH-20 but that other factors must have an important effect on the increased aerosol particle formation during NYH-20.

We further investigated the chemical composition of PM$_{2.5}$ to understand the haze puzzle during NYH-20. As shown in Figures 1a and 1b, the fraction of secondary aerosols during NYH-20 (72.5%) was significantly higher ($p < 0.01$) than that during NYH-19 (59.1%). More specifically, during NYH-19, OM was the dominant component in PM$_{2.5}$, while in other periods (including NYH-20), nitrate consistently represented the largest fraction. The sum of sulfate, nitrate, and ammonium (SNA) aerosols during NYH-20 (24.1 μg m$^{-3}$) was significantly higher ($p < 0.01$) than during NYH-19 (14.7 μg m$^{-3}$). Figure 1c clearly shows that the fractional contributions of secondary aerosol increased with increasing PM$_{2.5}$ mass concentrations for all three periods in 2020 (up to 88%), and a generally similar pattern was observed for the three periods in 2019 (see supporting information). Compared to the lowest-concentration PM$_{2.5}$ bins, the secondary fraction in the highest-concentration PM$_{2.5}$ bins increased steadily (by up to 27%), suggesting major contribution specifically of secondary aerosols in driving particulate pollution (Huang et al., 2014; Xin et al., 2015). Moreover, particulate pollution was largely driven by the formation of SNA aerosols. For example, during NYH-20, the fractional contribution of SNA increased from 41% in the lowest-concentration PM$_{2.5}$ bin to 72% in the highest-concentration PM$_{2.5}$ bin, whereas the corresponding contribution of secondary organic aerosols (SOA) decreased from 11% to 5%. The high contribution of SNA aerosols to high particulate pollution in the studied urban atmosphere could be due to the synergistic effects of emission, chemistry, and transport (An et al., 2019; Guo et al., 2014).

As discussed above, the SOA concentrations during NYH-20 (3.8 μg m$^{-3}$) and NYH-19 were quite similar (4.3 μg m$^{-3}$) (Figure 2a). In contrast, the SNA aerosol concentrations were over 1.6 times higher during NYH-20 (24.1 μg m$^{-3}$) than during NYH-19 (14.7 μg m$^{-3}$) (see Figure 2a), despite the fact that their precursors were 26.2% (NO$_2$), 32.1% (SO$_2$), and 9.3% (NH$_3$) lower after NYH-19 than after NYH-19. This observation is consistent with the significantly higher sulfur oxidation ratio (SOR = n[SO$_4^{2-}$]/($n$[SO$_2$]$^2^{-}$+$n$[SO$_2$])) and nitrogen oxidation ratio (NOR = n[NO$_3$]/($n$[NO$_2$]+$n$[NO$_2$])) (Ji et al., 2018) during NYH-20 (SOR = 0.50 ± 0.15 and NOR = 0.21 ± 0.13) compared to NYH-19 (SOR = 0.25 ± 0.10 and NOR = 0.14 ± 0.11), indicating more efficient formation of SNA aerosols during NYH-20. Both NOR and SOR were positively correlated with PM$_{2.5}$ mass, yet NOR displayed a much higher correlation coefficient than SOR (Figure 2b). During NYH-20, with the overall increase of PM$_{2.5}$ mass concentration, NOR and SOR increased continuously. Most strikingly, for NOR we observed a fivefold increase between low low-PM$_{2.5}$ samples and the high-PM$_{2.5}$ samples (Figure 2c). Low temperatures and high RH during the wintertime are generally favorable for the formation of SNA aerosols. Field measurements by others also revealed that aqueous-phase oxidation of SO$_2$ can be an important formation pathway of sulfate at high RH during haze events (Cheng et al., 2016; Xue et al., 2019). Moreover, the transformation of HNO$_3$ into particle phase is generally enhanced at high RH and low temperature conditions (Huang, He, et al., 2020; Sun et al., 2015). However, a uniform response, neither of NOR nor of SOR, to temperature and RH was not discernable (Figure 2c). For example, with the increase of PM$_{2.5}$ concentration, SOR increased steadily from 0.53 to 0.85; while a corresponding change for temperature and RH was not observed. Similarly, despite a lower temperature, NOR stabilized at 0.55 at PM$_{2.5}$ > 75 μg m$^{-3}$. Finally, if at all, the lower $T$ and higher-RH environment as encountered during NYH-19 should have stimulated the formation of secondary inorganic aerosols and should have led to higher and not lower PM$_{2.5}$ mass concentrations compared to NYH-20.
Here we further elucidate the effects of heterogeneous (aqueous) and photochemical processes on the nitrate and sulfate secondary aerosol formation. The WRF-CMAQ results show that NO2 and OH radical display a non-linear relationship (Figure S6). With the decrease of NO2, OH radical concentration increases (Figure S6). We have also compared the diurnal cycle of OH radicals before and after the COVID-19 shutdown. As shown in Figure S7, the daytime OH concentrations were much higher after the shutdown than before the shutdown, which can partly explain the enhanced formation of nitrate during NYH-20. Correlations between nitrate and sulfate as function of aerosol water content (AWC) or odd oxygen (Ox = NO2 + O3) during NYH-20 were investigated in Figure 2d. While there was generally a good correlation between nitrate and sulfate aerosol mass concentrations (r2 = 0.55), they both were rather independent of AWC and Ox. This finding is further supported by the fact that there was no significant correlation between AWC/Ox and nitrate/sulfate, respectively (Figure S8), and that the diurnal cycles of SNA species did not follow the diurnal patterns of AWC and Ox (Figure S9). These results suggest that other factors (e.g., long-range transport) likely affect the formation of SNA.

Located in the atmospheric outflow from East Asia, Shanghai is a typical receptor city, receiving episodically highly polluted air masses from northern China during wintertime. During NYH-20, there were three distinct PM2.5 pollution episodes (PE) with PM2.5 mass concentration peaks occurring on 28 January (PE-1), 3 February (PE-2), and 9 February (PE-3) (Figure 3a). For each PE, a 2-day back trajectory cluster analysis at 500 m is provided in Figure 3c. The majority of the polluted air masses are derived from northern China. This is particularly true for PE-1, when an aerosol layer (white rectangle in Figure 3a) between 1,500 and 2,500 m, detected by Lidar ceilometer, was transported from the upper to the lower...
atmospheric layer. During periods with transiently low PM$_{2.5}$ concentrations and chemical speciation (i.e., nitrate and sulfate) (e.g., 26 January: PM$_{2.5}$ = 5 μg m$^{-3}$; 6 February: PM$_{2.5}$ = 12 μg m$^{-3}$), air masses originated mainly from the open ocean (Figure S10). Performing an air-mass back trajectory analysis for Shanghai, we divided the potential geographical origins into five regions (i.e., local, northern China, southern China, northeast sea, and southeast sea; Figure S11). During NYH-20, 43.3% of the air masses arrived from northern mainland China, a region with intensive industries and miscellaneous air pollution sources (e.g., residential coal combustion for heating and cooling), while the corresponding share was only 4.2% during NYH-19 (Figure 3b). In fact, over 88% of air masses in NYH-19 are derived from the open sea. Typically, the frequent and strong influence of long-range transport (e.g., from northern China) will lead to the input of aged PM that can serve as condensation sinks, hindering atmospheric nucleation, and subsequent growth of newly formed nano-sized particles (Yao et al., 2018). Simultaneously measured at the Dongtan Eco-park (DT) and Dianshanhu (DSH) sites in Shanghai, the number size distributions of 3–736 nm particles during NYH-20 are illustrated in Figure 3d. There is evidence for one new particle formation event that occurred after PE-2 under relative clean-air conditions, yet no continuous and lasting size growth from the atmospheric layer.

![Figure 3](image-url)
nucleation-mode particles was observed before any of the PEs. Although sensitive to ambient conditions (e.g., concentrations of gaseous precursors, temperature, and RH), the lack of nucleation may partly be explained by large condensation sinks of preexisting particles from long-range transport. Collectively, these results provide putative evidence that long-range transport was of critical importance to haze pollution during NYH-20.

We can use the longest-lasting pollution episode (PE-1) as a test case to investigate how long-range atmospheric transport increase SNA secondary aerosol mass concentrations and in turn NOR and SOR. The time series of SNA concentrations and their gaseous precursors before (0:00–19:00 LT [27 January]), during (20:00 LT [27 January]–10:00 LT [31 January]), and after (11:00 LT [31 January]–0:00 LT [1 February]) PE-1 at the PD site in Shanghai are shown in Figure 4a, and NOR and SOR in and out of Shanghai for each period in Figures 4b and S12, respectively. The “apparent” growth rates (mean$_{\text{max}}^\text{min}$ ±1σ) of nitrate aerosol particles before, during, and after PE-1 were 2.51$^{+1.6}_{-0.8}$, 18.3$^{+9.9}_{-8.2}$ ± 4.0, and 6.3$^{+10.0}_{-3.4}$ ± 2.1 μg m$^{-3}$ hr$^{-1}$, respectively. Sulfate, ammonium, and SO$_2$ also reached the highest growth rates during PE-1. In contrast, the average concentration of NO$_2$ during PE-1 (37.5 μg m$^{-3}$) was lower than before (55.7 μg m$^{-3}$) and after (39.0 μg m$^{-3}$) PE-1. The minimum NO$_2$ during PE-1 cannot be explained by low local NO$_2$ emissions. In fact, during PE-1, the NO$_2$ fluctuations correspond well with observed changes in NO$_3^-$ ($r^2 = 0.71$) (Figure 4a), suggesting that the low NO$_2$ observed during PE-1 may be best explained by a highly efficient conversion of NO$_2$ to nitrate. At all the receptor sites (Jiaxing site in Zhejiang and DT, PD, and DSH sites in Shanghai), NOR was the highest during PE-1. Interestingly, during the phase after PE-1, NOR was the lowest at DT (0.19; remote wetland) in the NE, followed by PD (0.26; urban), DSH (0.40; rural), and Jiaxing (0.55; suburban) in the SW. This inter-city NOR gradient is consistent with the transport pathway of air masses and the associated oxidation of NO$_2$, as evidenced at the regional scale during PE-1 (see Figure 4b).

More specifically, following the regional transport pathway from north to south, NOR during PE-1 was the lowest in Beijing (0.24), then increased to 0.36 in Nanjing, and finally reached the highest values in Jiaxing.
and Shanghai. In contrast, NOR in Beijing was the highest before PE-1 and then gradually decreased to 0.24 during PE-1 and to 0.10 after PE-1. Given that Beijing is located in the source region of the North China Plain, the high NOR before the Shanghai PE-1 reflects the efficient conversion of NO₂ to nitrate during longer-term transport from the North. As a consequence, we further argue that the lowest NO₂ concentration observed during PE-1 indicates the highly efficient conversion of NO₂ to nitrate along the transport pathways. During the first day of PE-1, we observed a much higher average RH in Shanghai (76.6%) than that in Beijing (40.5%) (Figure 4b). Moreover, the growth rates of nitrate during PE-1 correlated significantly with RH in Shanghai ($r^2 = 0.56$, $p < 0.01$; Figure S13), highlighting the important role of RH in regulating nitrate formation during transport. SOR during PE-1 in Shanghai (0.73 at DT and DSH, 0.76 at PD) was not only much higher than in Beijing (0.38) and Nanjing (0.68) but also higher than in the southernmost Jiaxing (0.65) (Figure S12), suggesting divergent transport and transformation of sulfate versus nitrate.

4. Conclusions

In this work, we present a timely investigation of China’s haze puzzle in Shanghai in early 2020. Over two-times higher mass concentrations of fine particles were observed during the New Year Holiday and the contemporaneous COVID-19 outbreak, despite the record economic slowdown and associated decline in pollutant emissions from business, transportation, and industry. Fast formation of secondary inorganic (mostly nitrate) aerosols was identified as the main factor contributing to the relatively high atmospheric particle concentrations (>80%). We show that particulate nitrate formation can be largely enhanced (with nitrate formation rates up to 27 μg m⁻³ hr⁻¹) during lasting regional transport, suggesting that differential transport patterns, rather than local emissions, may be responsible for fluctuations in aerosol concentrations (e.g., NYH-19 vs. NYH-20). The results of this study highlight that regional joint management efforts and control strategies are required to effectively clear China’s air.

Conflict of Interest

The authors declare that they have no conflicting interest.

Data Availability Statement

The associated data can be downloaded online (https://doi.org/10.5281/zenodo.3738768).

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