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Evolution of organic carbon during COVID-19 lockdown period: Possible contribution of nocturnal chemistry



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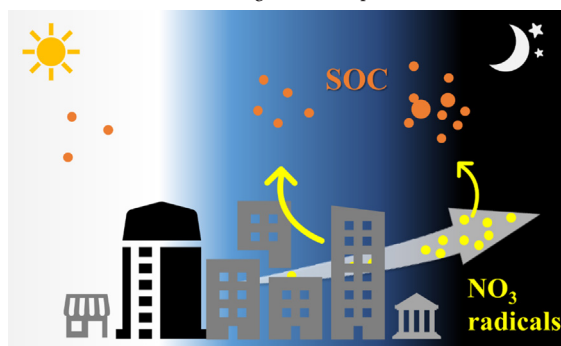
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HIGHLIGHTS

- Higher concentration of SOC was observed during the lockdown period than the normal period.
- Nocturnal chemistry related to NO₃ radicals contributed to increase of SOC.
- The southeast regions of the NCP contributed more to SOC during the lockdown period than the normal period.

GRAPHICAL ABSTRACT

Potential source of SOC during COVID-19 pandemic.



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ABSTRACT

Carbonaceous aerosol is one of the main components of atmospheric particulate matter, which is of great significance due to its role in climate change, earth's radiation balance, visibility, and human health. In this work, carbonaceous aerosols were measured in Shijiazhuang and Beijing using the OC/EC analyzer from December 1, 2019 to March 15, 2020, which covered the Coronavirus Disease 2019 (COVID-19) pandemic. The observed results show that the gas-phase pollutants, such as NO, NO₂, and aerosol-phase pollutants (Primary Organic Compounds, POC) from anthropogenic emissions, were significantly reduced during the lockdown period due to limited human activities in North China Plain (NCP). However, the atmospheric oxidation capacity (Ox/CO) shows a significantly increase during the lockdown period. Meanwhile, additional sources of nighttime Secondary Organic Carbon (SOC), Secondary Organic Aerosol (SOA), and $b_{\text{abs, BrC}}(370 \text{ nm})$ are observed and ascribed to the nocturnal chemistry related to NO₃ radical. The Potential Source Contribution Function (PSCF) analysis indicates that the southeast areas of the NCP region contributed more to the SOC during the lockdown period than the normal period. Our results highlight the importance of regional nocturnal chemistry in SOA formation.

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1. Introduction

Over the past few decades, the poor air quality in China has attracted intensive attentions (Fan et al., 2021; Liu et al., 2020a; Wang et al., 2021b; Zong et al., 2021). To improve air quality, the Chinese government has implemented a series of strict emission control strategies, such as the Air Pollution Prevention and Control Action Plan in 2013 (Cai et al., 2017; Ge et al., 2021). As a consequence, particulate mass concentration with a diameter less than 2.5 μm ($\text{PM}_{2.5}$) had been significantly reduced nationwide (Wang et al., 2017). For example, the annual mean $\text{PM}_{2.5}$ concentration in Beijing dramatically reduced from 89.5 $\mu\text{g m}^{-3}$ in 2013 to 42.0 $\mu\text{g m}^{-3}$ in 2017 (Wang et al., 2020d; Zhang et al., 2019b). However, the average $\text{PM}_{2.5}$ concentration in China still far exceeds the guideline recommended by the World Health Organization (WHO, annual mean of 5.0 $\mu\text{g m}^{-3}$).

Reducing anthropogenic emissions can significantly decrease air pollution levels (Le et al., 2020; Wang et al., 2021a). For example, great improvements in air quality have been observed during the 2008 Olympics (Huang et al., 2010; Wang et al., 2010), the 2014 Asia-Pacific Economic Cooperation (APEC) meeting (Huang et al., 2015; Meng et al., 2015; Sun et al., 2016), the 2016 Hangzhou G20 summit (Li et al., 2018), and the 2015 China Victory Day Parade in Beijing (Ren et al., 2019; Zhao et al., 2017). The outbreak of the COVID-19 was followed by a strict nationwide lockdown in China, which has restricted anthropogenic activities such as transportation, commerce, and industry to prevent the spread of COVID-19 on a national scale. As a consequence, the emissions of primary air pollutants from anthropogenic sources were significantly reduced (Chen et al., 2020a; Le et al., 2020; Liu et al., 2020b; Lu et al., 2021; Ma et al., 2021). This was also observed as the spread of the COVID-19 pandemic worldwide (Baldasano, 2020; Kanniah et al., 2020; Lee et al., 2020; Otmami et al., 2020; Zhang et al., 2021a; Zoran et al., 2020). However, heavy air pollution events dominated by $\text{PM}_{2.5}$ still occurred during the COVID-19 lockdown in China (Huang et al., 2021; Li et al., 2021a; Liu et al., 2020b). This highlights the complexity and the nonlinearity between the primary and secondary pollutants in the atmosphere. The long-term lockdown period due to the COVID-19 pandemic provides us a good chance for understanding the chemistry connecting primary emissions to secondary pollutants formation.

The NCP region is one of the economic zones in China, while most affected by air pollution (Li et al., 2020; Sun et al., 2021b). Carbonaceous aerosol is one of the main components of atmospheric particulate matter, which is of great significance due to its role in climate change, earth's radiation balance, visibility, and human health (Bikkina et al., 2017; Li et al., 2009; Wang et al., 2020b; Wang et al., 2020e). Organic carbon (OC) is the main component (~90%) (Ram et al., 2008) of carbonaceous aerosols, while organic aerosol (OA) is an important contributor (20–90 wt%) to $\text{PM}_{2.5}$ (Cao et al., 2012; Sosa et al., 2017). OC can be directly emitted from various sources, such as traffic exhaust, biomass combustion, and plant respiration, and secondarily formed through photochemical or heterogeneous reactions in the atmosphere (Bae et al., 2004; Turpin and Huntzicker, 1995). Many previous works have discussed the concentration changes of air pollutants, such as O_3 (Kang et al., 2021; Lu et al., 2021), NO_2 and HONO (Liu et al., 2020b), $\text{PM}_{2.5}$ (Chen et al., 2020a; Zheng et al., 2020), heavy metal elements (Cui et al., 2020), light absorption components in $\text{PM}_{2.5}$ (Chen et al., 2020b), the source changes from biomass burning (Metya et al., 2020), and secondary formation (Huang et al., 2021; Sun et al., 2020b) during the lockdown period. However, there were few studies focusing on OC and the possible formation mechanism of SOC during the COVID-19 pandemic in the NCP region.

In this work, carbonaceous aerosols were semi-continuously measured in Beijing and Shijiazhuang, which are the typical cities in the NCP region, before and during the lockdown period of the COVID-19 pandemic. The effects of the lockdown on the concentrations, light absorption properties, formation mechanism, and potential sources of carbonaceous aerosols were discussed. This work will help for understanding the response of secondary formation of OC to emission reductions in the atmosphere.

2. Material and methods

2.1. Field measurements

Ground observations were conducted in Beijing and Shijiazhuang (Fig. S1) from December 1, 2019 to March 15, 2020, which covered the normal (December 1, 2019 to January 22, 2020) and the lockdown (January 23, 2020 to March 15, 2020) periods. The sampling site in Beijing is located at Aerosol and Haze Laboratory of Beijing University of Chemical Technology (AHL/BUCT, about 18 m from the ground, Lat. 39.97 and Lon. 116.42) (Cai et al., 2020; Chu et al., 2021; Liu et al., 2020c; Zhou et al., 2019) and the sampling site in Shijiazhuang is located at Hebei Atmospheric Super Station (HAS/SJZ, about 25 m from the ground, Lat. 38.03 and Lon. 114.61) (Liu et al., 2020b). Both cities are typical urban observation stations surrounded by traffic and residential emissions.

Mass concentration of $\text{PM}_{2.5}$ was measured by a beta attenuation mass monitor (BAM-1020, Met One Instruments) in Shijiazhuang and a Tapered Element Oscillating Microbalance (TEOM, ThermoFisher Scientific, 1405) in Beijing. Trace gases, including NO_x , SO_2 , CO , and O_3 , were measured with corresponding analyzers (Thermo Scientific 42i, 43i, 48i, and 49i) in the two sites. Monitor for Aerosols and Gases in ambient air (MARGA, ADI 2080, Applikon Analytical B.V.) were used to measure water-soluble ions (Cl^- , NO_3^- , SO_4^{2-} , NH_4^+) in $\text{PM}_{2.5}$ in Shijiazhuang and a Time-of-flight-Aerosol Chemical Speciation Monitor (ToF-ACSM) was used to measure non-refractory components (OA , Cl^- , NO_3^- , SO_4^{2-} , NH_4^+) in $\text{PM}_{2.5}$ in Beijing. Detailed information for ToF-ACSM has been described in previous works (Cai et al., 2020; Middlebrook et al., 2012).

Nitrate radical (NO_3) was measured with an iodide-based chemical ionization Atmospheric Pressure Interface Time-of-Flight Mass Spectrometer (CI-API-TOF, Aerodyne Research, Inc.) at the AHL/BUCT station. The API-TOF and CI-inlet has been well described in previous works (Jokinen et al., 2012; Junninen et al., 2010; Kurtén et al., 2011). Briefly, a 10 L min^{-1} of sample of air was drawn into the instrument through a 1/4 in. (O.D.) and 2 m (length) Teflon tubing, with the residence time of sample gas around 380 ms. 2 L min^{-1} of the sample gas was then drawn into the Ion-Molecule Reaction (IMR) unit. The main part of IMR was heated to 30 °C to reduce the wall loss of pollutants on the inner surface. In addition, the pressures in the IMR and Small Segmented Quadrupole (SSQ) were regulated to ~300 and ~2.5 mbar. This instrument can alternately measure gaseous species and particle-phase compounds after thermal desorption using a Filter Inlet for Gases and Aerosols (FIGAERO) (Lopez-Hilfiker et al., 2014). During the gas-phase sampling phase, it is able to detect the dinitrogen pentoxides (N_2O_5) in the cluster form of ($\text{N}_2\text{O}_5\text{I}^-$) (Kercher et al., 2009). The concentration of NO_3 was calculated based on the equilibrium between NO_3 and N_2O_5 (Brown et al., 2006). The calibration of N_2O_5 was performed as same as that described in previous work (Wang et al., 2016), i.e., by injecting known concentration of N_2O_5 produced via mixing O_3 and NO_2 .

OC and element carbon (EC) in $\text{PM}_{2.5}$ were measured at a 1-hour resolution using a semi-continuous thermo-optical transmittance (TOT)-based OC/EC analyzer (Model4, Sunset Laboratory Inc.). The principles and operating procedures have been described in detail elsewhere (Bae et al., 2004; Ji et al., 2018b; Klingshirn et al., 2019). An online parallel carbon stripper capable of removing volatile organic gases was installed upstream of the analyzer. Ambient air with a flow rate of 8 L min^{-1} was sampled through the $\text{PM}_{2.5}$ cyclone separator inlet and 3/8-inch steel tube into the OC/EC analyzer. $\text{PM}_{2.5}$ was collected hourly for 40 min on a baked round quartz fiber filter and then analyzed according to the NIOSH-5040 protocol (Birch and Cary, 1996). All carbons on the filter are thermally volatilized and oxidized to carbon dioxide (CO_2), then quantified sequentially using non-dispersive infrared (NDIR) detectors. The split point between OC and EC was determined by a 660 nm (Rattigan et al., 2010) laser to correct the artifacts in EC produced in the first stage. At the end of each sample, CO_2 was calibrated using methane equilibrated with 95% helium. Additional calibrations were performed monthly with known amounts of sucrose and weekly with zero calibration. Black carbon (BC) was measured

with a seven-band Aethalometer (AE33, Magee Scientific) in both sites. The AE33 has been widely used to continuously measure aerosol light absorption at 370, 470, 520, 590, 660, 880, and 950 nm (Helin et al., 2021; Jing et al., 2019; Lin et al., 2021). The principle and more details of AE33 can be found in previous work (Drinovec et al., 2015).

Meteorological parameters including temperature (T), pressure, relative humidity (RH), wind speed (WS), and wind direction (WD) were measured using a meteorological station (WXT520, Vaisala) in Shijiazhuang and an automatic weather station (QML201C and PWD22 Vaisala) in Beijing. Mixing boundary layer height (MLH) was measured using a Doppler Lidar (EV-Lidar-CAM, Everise Technology Ltd.) in Shijiazhuang and ceilometer (CL51, Vaisala) in Beijing.

2.2. Data treatment and analysis

2.2.1. SOC estimation by minimum R squared (MRS) method

POC is usually emitted from primary emission sources, including vehicular emissions, biomass burning, and cooking emissions. SOC can be formed through the oxidation of POC and volatile organic compounds (VOCs). The EC tracer method (Turpin and Huntzicker, 1995; Zhao et al., 2013) has been widely used to estimate the mass concentration of SOC using the following equation, in which combustion is assumed to be the only source of EC.

$$\text{POC} = (\text{OC}/\text{EC})_{\text{pri}} \times \text{EC} \quad (1)$$

$$\text{SOC} = \text{OC}_{\text{total}} - \text{POC} \quad (2)$$

Thus, the first step is to find the appropriate ratio of primary OC/EC, i.e., $(\text{OC}/\text{EC})_{\text{pri}}$ (Turpin and Huntzicker, 1995). In most of these previous studies, the lowest value (5–20%) of OC/EC was used empirically (Yao et al., 2020; Zhang et al., 2019a). In this study, the appropriate value of $(\text{OC}/\text{EC})_{\text{pri}}$ was determined using the MRS method (Bian et al., 2018; Ji et al., 2018b; Millet et al., 2005; Sun et al., 2020a; Wu and Yu, 2016), which determining the $(\text{OC}/\text{EC})_{\text{pri}}$ when the smallest correlation coefficient (R^2) between SOC and EC achieved by assuming a series of continuous OC/EC ratios. More details can be found elsewhere (Wu et al., 2019; Wu and Yu, 2016; Yao et al., 2020). The actual $(\text{OC}/\text{EC})_{\text{pri}}$ corresponding to the minimum R^2 (SOC vs. EC) are shown in Fig. S2. The monthly $(\text{OC}/\text{EC})_{\text{pri}}$ values in Shijiazhuang were 3.14, 2.43, 4.91, and 3.28, respectively, in December 2019, January 2020, February 2020, and March 2020. The corresponding values in Beijing were 2.70, 2.88, and 2.96 for December 2019, January 2020, and February 2020, respectively. The $(\text{OC}/\text{EC})_{\text{pri}}$ value in March 2020 was absent due to the miss of OC data.

In addition, positive matrix factorization (PMF) was applied to confirm the SOC results in Beijing. An IgorPro program (Wavemetrics, ver. 6.3.7.2) based the Source Finder toolkit (SoFi, ver. 6.8.4) with a multi-linear engine (ME-2) was used to execute the PMF analysis and evaluate the results. Detailed information of SoFi and PMF calculations are described elsewhere (Cai et al., 2020; Canonaco et al., 2013; Daellenbach et al., 2016).

2.2.2. The light absorption data

BC was measured using an aethalometer (AE33, Magee Scientific), which has seven bands at 370, 470, 520, 590, 660, 880, and 950 nm. Brown carbon (BrC) was calculated according to the previous works (Virkkula et al., 2015; Zhang et al., 2021b). Briefly, the mass absorption cross-section (MAC) values were 18.47, 14.54, 13.14, 11.58, 10.35, 7.77, and $7.19 \text{ m}^2 \text{ g}^{-1}$ for the corresponding wavelengths in AE33. The absorption coefficient (b_{abs}) at different λ was calculated according to Eq. (3):

$$b_{\text{abs},\lambda} = \text{BC}_{\text{output},\lambda} \times \text{MAC}_{\text{BC},\lambda} \quad (3)$$

where, $\text{BC}_{\text{output},\lambda}$ is the mass concentration of BC at wavelength λ nm. Absorption Ångström exponent (AAE) has been widely used to describe the wavelength dependence of light absorb aerosol (Lack and Langridge,

2013; Laskin et al., 2015; Liu et al., 2018; Wang et al., 2020c). It can be calculated according to Eq. (4):

$$b_{\text{abs}} = K \times \lambda^{-\text{AAE}} \quad (4)$$

where, K is a constant value. It should be noted that non-BrC coating on BC could also lead to increased AAE (Garg et al., 2016; Lack and Langridge, 2013). However, lots of researches usually suggested that the AAE value of BC is about 1 (Lack and Langridge, 2013; Liu et al., 2018). At long wavelengths, such as 880 and 950 nm, BrC does not absorb light (Kirchstetter et al., 2004; Liakakou et al., 2020). Thus, the absorption of BC and BrC can be calculated according to Eqs. (5) and (6), respectively:

$$b_{\text{abs},\text{BC},\lambda} = b_{\text{abs},\text{BC},880} \times \left(\frac{880}{\lambda}\right)^{\text{AAE}_{\text{BC}}} \quad (5)$$

$$b_{\text{abs},\text{BrC},\lambda} = b_{\text{abs},\lambda} - b_{\text{abs},\text{BC},\lambda} \quad (6)$$

2.2.3. Backward trajectory and PSCF model analysis

To understand the potential source regions, we performed a 48-hour backward trajectory analysis using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. The time interval was set to 1 h. The National Centers for Environmental Prediction (NCEP) Global Data Assimilation System (GDAS) data (<ftp://arlftp.arl.hq.noaa.gov/pub/archives/>) with a spatial resolution of $1^\circ \times 1^\circ$ were input to the HYSPLIT model. 100 m (Dimitriou and Kassomenos, 2016) was used as the arriving height. 2544 trajectories were obtained. The Potential Source Contribution Function (PSCF) analysis was carried out based on the above generated backward trajectories using the TrajStat software (<http://meteothink.org/index.html>) (Wang, 2014, 2019; Wang et al., 2009) to identify the source strength of a geographical area. Weighting functions were used to minimize the uncertainty of PSCF analysis and referred to as WPSCF (Polissar, 1999; Sun et al., 2019a).

3. Results and discussion

3.1. Overview of the air quality during observation

Fig. 1 shows the time series of various pollutants and meteorological parameters in Beijing and Shijiazhuang, including NO, NO₂, O₃, T, RH, WS, WD, and MLH. Because of the lack of OA data in the Shijiazhuang site, the OA was replaced with organic matter (OM, $1.5 \times \text{OC}$) (Countess et al., 1980; Japar et al., 1984; Ren et al., 2021; Xing et al., 2013). During the lockdown period, the mean PM_{2.5} concentrations in Shijiazhuang decreased by 15.2% (from 114.3 ± 84.6 to $97.0 \pm 75.1 \mu\text{g m}^{-3}$) with respect to the normal period. By contrast, the mean PM_{2.5} concentrations in Beijing increased considerably (52.4%, from 41.9 ± 39.7 to $63.9 \pm 58.3 \mu\text{g m}^{-3}$) during the lockdown period compared with that in the normal period (Fig. 1A and E). The mean concentrations of NO and NO₂ decreased sharply compared to these in the normal period in both Shijiazhuang (87.9 and 41.8%, from 22.2 and 26.5 ppb to 2.7 and 15.5 ppb) and Beijing (82.2 and 59.6%, from 20.1 and 31.0 to 3.6 and 12.5 ppb). This is in agreement with the strict controls on anthropogenic emissions during the lockdown period. However, O₃ concentrations increased 176.0% (from 11.7 to 32.2 ppb) in Shijiazhuang and 241.2% (from 7.4 to 25.1 ppb) in Beijing (Fig. 1B and F).

Long-term stagnant meteorological conditions, including high RH, low WS, and low MLH, result in the accumulation of pollutants, subsequently, lead to the high occurrence of haze pollution during the lockdown periods (Chen et al., 2020b; Le et al., 2020; Liu et al., 2020b; Sun et al., 2019b). As shown in Fig. S3, the wind speeds in Beijing are significantly lower than that in Shijiazhuang, which means the air mass in Beijing will be more readily affected by the adverse meteorological conditions than that in Shijiazhuang. This could be the reason why the mean concentration of PM_{2.5} increased at the beginning of the lockdown period compared to the

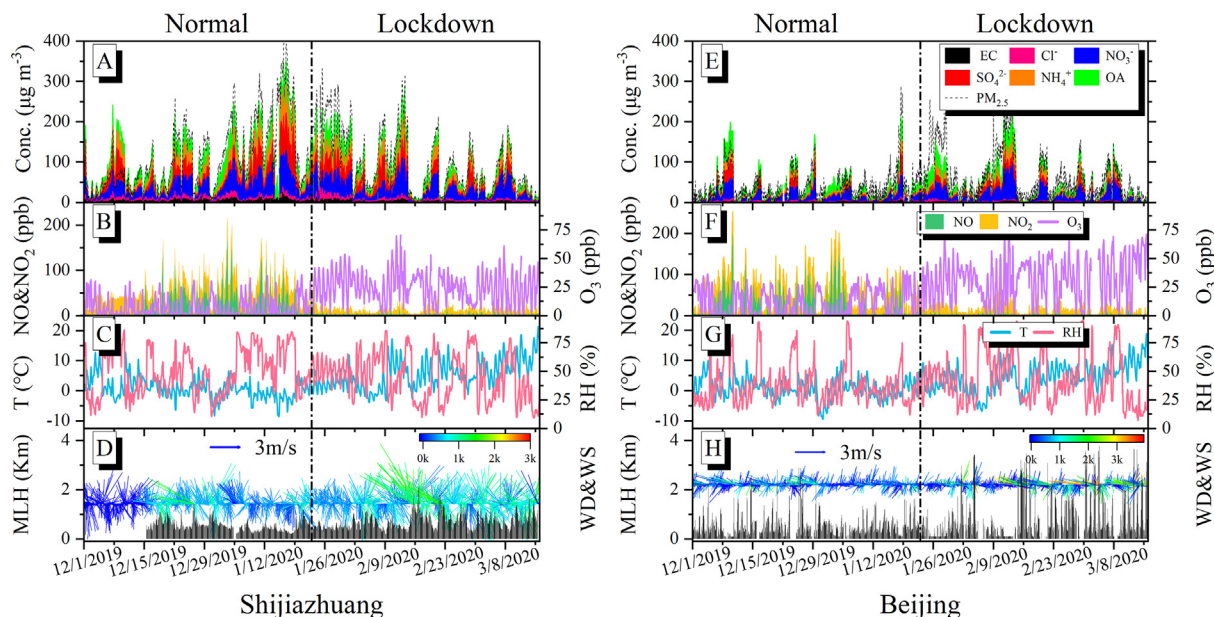


Fig. 1. Time series of components of $PM_{2.5}$, NO, NO_x, O₃, T, RH, WD, WS, and MLH in Shijiazhuang (A–D) and Beijing (E–H), the wind data was colored by MLH. The dash line divided the normal and the lockdown period.

normal period in Beijing, while a reduction was observed in Shijiazhuang. In addition, the relative contributions of local pollution and regional transport are varied. For the first pollution episode in Beijing, it is mainly due to the aggravation of local pollution, while the contribution of regional transport increased considerably in the second pollution episode (Zhao et al., 2020). It should be noted that the mean of MLH increased 29.3% (from 555.8 ± 233.3 to 718.9 ± 315.2 m) in Shijiazhuang and 55.4% (from 529.4 ± 419.1 to 822.4 ± 867.3 m) in Beijing during the lockdown period. In addition, the mean wind speed increased 30.8% (from 1.35 ± 0.73 to 1.77 ± 0.97 m s⁻¹) in Shijiazhuang and 10.6% (from 0.75 ± 0.55 to 0.82 ± 0.57 m s⁻¹) in Beijing. Thus, considering the improvement of dispersion ability, the concentrations of secondary pollutants ($PM_{2.5}$ and O₃) should decrease in both Shijiazhuang and Beijing during the lockdown period. The chemistry process should thus play important roles in both the elevated $PM_{2.5}$ and O₃ during the lockdown period.

NO_x is the precursors of nitrate, which is the dominator of $PM_{2.5}$ mass in the NCP region (Li et al., 2021b; Tao et al., 2017). This means that a decrease in NO_x concentration might be beneficial to $PM_{2.5}$ reduction. However, two large-scale $PM_{2.5}$ pollution episodes (Zhao et al., 2020) still occurred in the NCP region (Fig. 1A and E) at the beginning of the lockdown period although NO_x showed a significant reduction in the COVID-19 episode. This is consistent with previous results that emergency measures for severe haze prevention can only weakly reduce $PM_{2.5}$ concentration (Ma et al., 2020; Wang et al., 2020a). Previous works also suggested that the chemical transformation of gaseous pollutants to secondary inorganic aerosols may lead to the explosive growth of $PM_{2.5}$ (Chen et al., 2020a; Sun et al., 2019b; Wang et al., 2013; Zheng et al., 2015; Zhong et al., 2018). As shown in Fig. S4B, nitrate was the dominant contributor to inorganic salts during the whole campaign. This is consistent with previous observations during the COVID-19 lockdown in north China (Huang et al., 2021). The nitrogen oxidation ratio (NOR), which is calculated from the molar ratios of the particulate concentration and the sum of gaseous and particulate concentrations,

$$NOR = m(NO_3) / [m(NO_3) + m(NO_x)] \quad (7)$$

showed a significant increase of 109.6% (from 0.2 ± 0.2 to 0.5 ± 0.2) in Shijiazhuang and 40.2% (from 0.5 ± 0.2 to 0.7 ± 0.2) in Beijing (Fig. S5). This indicates the increase of atmospheric oxidation capacity during the lockdown period in both Shijiazhuang and Beijing. Chen et al.

(2020a) reported that the diurnal variation of NOR in Shanghai showed a prominent peak at midnight, which indicates the importance of nocturnal reactions to nitrate production.

NO_x is also one of the precursors of O₃. Huang et al. (2021) pointed out that the significant reduction of NO_x emissions from transportation led to the generation of O₃ and nocturnal NO₃ radicals during the lockdown period (Huang et al., 2021; Zhao et al., 2020). The increase in O₃ concentration (Fig. 1B and F), which is from the nonlinear photochemical reactions between NO_x and VOCs (Le et al., 2020; Nichol et al., 2020), also suggests an enhanced oxidation capacity. Ox (O₃ + NO₂) has been widely used to estimate the total atmospheric oxidation capacity (Chen et al., 2020a; Kley et al., 1994; Leighton, 2012; Notario et al., 2013). Meanwhile, carbon monoxide (CO) has a long lifetime against oxidation by OH radicals and thus can be used as a reasonable tracer of primary emissions to account for atmospheric dilution (De Gouw and Jimenez, 2009; DeCarlo et al., 2010). Thus, the ratios of other pollutant species to CO can partially eliminate the boundary layer effect (Yao et al., 2020). Fig. S4A shows the CO normalized atmospheric oxidant capacity (Ox/CO) during the whole campaign. Compared with the normal period, the atmospheric oxidation capacity during the lockdown period increased significantly in both sites. In addition, as shown in Fig. S4B, OA is always the most important contributor to $PM_{2.5}$ in both Shijiazhuang and Beijing. For example, the fraction of OA in $PM_{2.5}$ was 28.9% in Shijiazhuang and 34.6% in Beijing, respectively, during the lockdown period. In previous work, attention was mainly paid to the formation of secondary inorganic aerosol during the COVID-19 lockdown (Chen et al., 2020a; Lu et al., 2021), while less attention was paid to organic aerosol formation. In the following sections, we will focus on the evolution of OA component.

3.2. Increase of nocturnal SOC during lockdown periods

The mean concentration of OA showed a decrease of 17.6% (from 23.3 ± 12.9 to 19.2 ± 13.4 µg m⁻³) in Shijiazhuang, while a slight increase of 7.6% (from 13.4 ± 12.2 to 14.4 ± 12.9 µg m⁻³) in Beijing during the lockdown period. Interestingly, OA showed a less decreased amplitude in Shijiazhuang or a more increase amplitude in Beijing in the nighttime compared with the daytime (Fig. S6). This implies that there should have an additional OA source in the NCP region.

Fig. S7 shows the mean concentrations of OC, EC, POC, and SOC during different periods. The mean concentrations of OC, EC, and POC of

Shijiazhuang were 12.9 ± 8.9 , 3.6 ± 2.3 , and $7.7 \pm 5.3 \mu\text{g m}^{-3}$, respectively, during the lockdown period, which was 17.8, 32.7, and 30.6% lower than those during the normal period (15.7 ± 8.5 , 5.3 ± 3.1 , and $11.1 \pm 6.1 \mu\text{g m}^{-3}$). However, the SOC concentration increased by 13.6% (from 4.6 ± 4.3 to $5.2 \pm 6.3 \mu\text{g m}^{-3}$) in Shijiazhuang. In Beijing, the concentrations of OC, EC, and SOC increased significantly ($P < 0.05$) during the lockdown period. As mentioned above (Fig. 1), Beijing was more readily affected by the adverse meteorological conditions and regional transport of air mass than Shijiazhuang. The increase of the concentrations of OC and EC in Beijing might be related to the transport of the regional background although the anthropogenic emissions were reduced. The mean concentrations of SOC in both two sites showed significant increases contrasting with the reduction of POC during the lockdown period. This means that the secondary formation of OC was enhanced during the lockdown period while anthropogenic activities were suppressed.

Fig. 2 shows the diurnal patterns of OC, EC, POC, SOC, and SOC/OC during the normal (black dots and lines) and the lockdown periods (the red dots and lines). The area between the diurnal pattern lines means the difference of the pollutant between the lockdown period and the normal period. The yellow color indicates a positive difference, while the blue one means a negative difference. Both OC and EC showed a bimodal diurnal distribution. The peaks appeared at 09:00 and 02:00, respectively, in Shijiazhuang, while they occurred at 10:00 and 21:00 in Beijing. POC and EC are solely from primary emissions (De Gouw and Jimenez, 2009; Safai et al., 2014). In Shijiazhuang, the diurnal patterns of POC and EC changed little during the two periods. Thus, their differences remained stable (Fig. 2B and C). These results indicate that the emission intensity of EC and POC were reduced, while their emission patterns should remain during the lockdown period when compared with that during the normal period. However, OC and SOC showed different diurnal patterns during the normal and the lockdown period (Fig. 2A and D), especially in the nighttime. Either a larger yellow difference of SOC (Fig. 2D) or a lesser blue difference of OC (Fig. 2A) in nighttime indicates a strong nocturnal SOC source during the lockdown period. The diurnal patterns of SOC in Beijing (Fig. 2I) were similar to those in Shijiazhuang. In addition, the nighttime increase of the SOC during the lockdown period related to the normal period in Beijing (65.4%,

from 1.3 ± 1.1 to $2.1 \pm 2.6 \mu\text{g m}^{-3}$) was even more remarkable when compared with that in Shijiazhuang (23.2%, from 4.6 ± 4.4 to $5.7 \pm 6.6 \mu\text{g m}^{-3}$). Moreover, the SOC/OC in Beijing (Fig. 2J) showed a similar diurnal variation as SOC, i.e. a distinct nocturnal increase, especially in Beijing. The diurnal variation of SOC/OC showed a remarkable increase in both daytime and nighttime in Shijiazhuang (Fig. 2E), which resulting into a weak difference between the daytime and nighttime SOC/OC ratios. This obvious enhancement of the nocturnal SOC/OC might be related to the primary emissions, subsequently, strong photochemical formation of SOA during the normal period in Beijing (Kuang et al., 2020). Therefore, we speculate that the increase of nighttime SOC concentration should be related to nocturnal chemistry.

To verify the speculation mentioned above, PMF of OA measured using an ACSM in Beijing was performed. Four factors including more-oxidized aerosol (MOOA), less-oxidized organic aerosol (LOOA), cooking organic aerosol (COA), and fossil fuel aerosol (FFOA) were obtained. The time series and diurnal variations of the four factors are presented in Fig. S8 and S9. SOA (MOOA + LOOA) was significantly higher during the lockdown period than that in the normal period (Fig. S9), which is consistent with the change of SOC (Fig. 2H). Meanwhile, the diurnal pattern of SOA showed a more significant increase in the night like that of SOC (Fig. 2H) during the lockdown period when compared to the normal period. Furthermore, the diurnal variation of primary organic carbon (POA, FFOA + COA) derived from ACSM-PMF showed a reduction during the lockdown period. In addition, the SOA also showed a good correlation ($R^2 = 0.55$, Fig. S10) with SOC in polluted episodes (with $\text{PM}_{2.5}$ higher than $75 \mu\text{g m}^{-3}$) during the whole campaign. These results reveal that the conclusions based on SOC analysis are reasonable. In addition, it suggests the nocturnal chemistry is important in OA formation during the lockdown period.

3.3. Nocturnal atmospheric oxidation changes during COVID-19 pandemic

NO_3 radicals were the essential nocturnal oxidants in the atmosphere (Brown et al., 2003). In Beijing, the NO_3 radical concentrations were measured using a CI-API-TOF. The measured NO_3 radicals in Beijing showed a

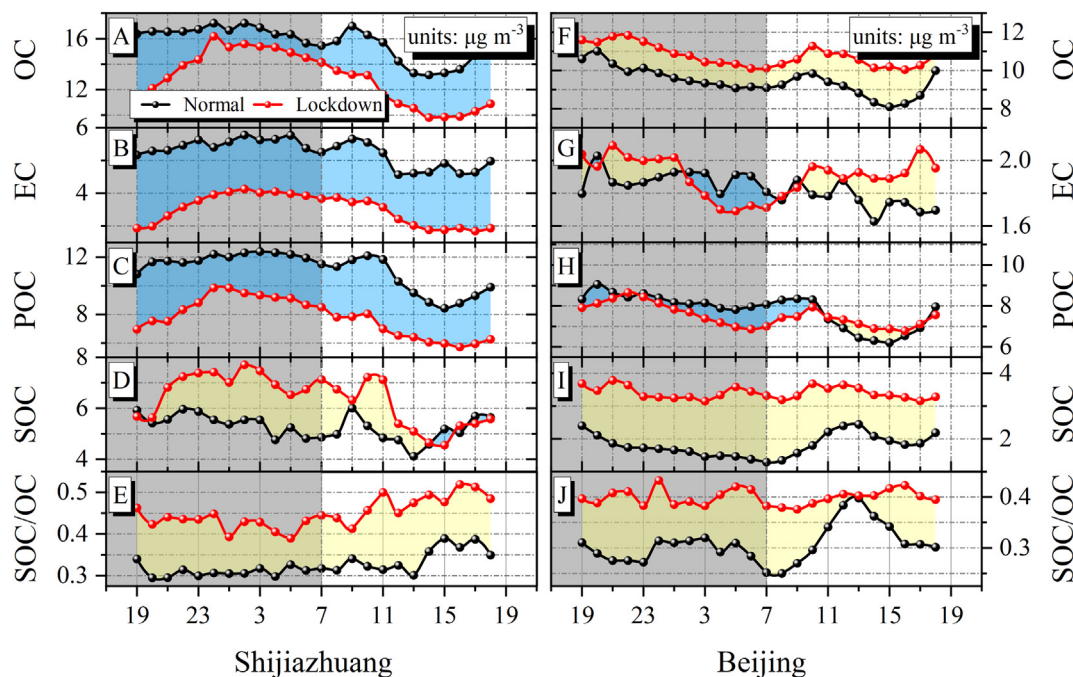


Fig. 2. The mean diurnal cycle of carbonaceous aerosol components: OC, EC, POC, SOC, and SOC/OC in Shijiazhuang (A–E) and Beijing (F–J) during the normal period (black line) and the lockdown period (red line). The unit of the species is $\mu\text{g m}^{-3}$. The shadow area indicates nighttime. The area between the two lines is filled with blue or yellow to distinguish the relative change of the difference between the normal period and the lockdown period. The yellow color indicates a positive difference, while the blue one means a negative difference.

significant increase from 0.6 ± 1.2 ppt in the normal period to 2.5 ± 5.1 ppt in the lockdown period (Fig. 3A). Meanwhile, the nocturnal NO_3 concentration was ~ 3 times higher than that in the daytime (Fig. 3B). This means the nighttime oxidation capacity also increased during the lockdown period in Beijing.

Unfortunately, NO_3 radical measurements were unavailable in the Shijiazhuang. As for the sources of NO_3 radicals in the atmosphere, there is an equilibrium between NO_3 and N_2O_5 , while N_2O_5 is formed from the reaction between NO_2 and O_3 . On the other hand, the reaction between NO and NO_3 is an important sink of NO_3 radicals. Thus, we use $\text{O}_3 \times \text{NO}_2/\text{NO}$ as a proxy of NO_3 concentration in the atmosphere (Brown et al., 2003). As shown in Fig. 3, although the absolute values are different between the measured NO_3 concentrations and the proxy of NO_3 radicals in Beijing, the estimated NO_3 radicals show a similar variation trend during different periods. At the same time, the diurnal patterns of the estimated NO_3 are also similar to that of the measured NO_3 (Fig. 3). Thus, we further estimated the NO_3 concentrations in Shijiazhuang (Fig. S11). It showed similar diurnal curves like Beijing. Obviously, the nocturnal NO_3 concentrations in both Beijing and Shijiazhuang significantly increased during the lockdown period when compared with the normal period. This can be explained by the reduction of NO and the increase of O_3 on a regional scale (Kang et al., 2021). In addition, the mean concentration of SOA increased as a function of NO_3 radical concentration when SOA concentrations were higher than $20 \mu\text{g m}^{-3}$, usually occurred in the lockdown period (Fig. S12). This phenomenon clearly illustrates that the atmospheric oxidation capacity was significantly increased in the lockdown period compared to the normal period. Therefore, we can ascribe the enhanced SOC formation in the lockdown period than the normal period to nocturnal chemistry related to NO_3 radicals although more evidence such as SOA tracers associated with nocturnal NO_3 radicals are required in the future.

3.4. Impact of COVID-19 on aerosol light absorption

Aerosol light absorption significantly influences on the earth's climate, tropospheric chemistry, and visibility (Ding et al., 2016; Sun et al., 2021a; Watson, 2002). BC and BrC, which play essential roles in the global radiative balance of the earth's atmosphere (Andreae and Gelencser, 2006; Bond, 2001), are the two fundamental optical carbon matters in $\text{PM}_{2.5}$. BC has been considered the most effective climatic forcing agent, with broadband absorption properties (Bond et al., 2013). BrC, as a kind of light-absorbing OC, shows a wavelength-dependent absorption that peaks in the ultraviolet (UV) spectral region and declines sharply in the visible spectral region (Andreae and Gelencser, 2006; Laskin et al., 2015). Both BC and BrC can be emitted directly from the combustion of fossil fuels and biomass burning, while BrC can also be formed in atmospheric chemical processes (e.g., multi-phase reactions between gas-phase, particles, and

in-cloud processes) (Andreae and Gelencser, 2006; Harrison et al., 2005; Laskin et al., 2015; Wang et al., 2019; Zhang et al., 2013b). The absorption of BrC at 370 nm ($b_{\text{abs, BrC}}(370 \text{ nm})$) was the most intensive in the seven wavelengths and regarded as the representative value of BrC absorption (Lin et al., 2021; Wang et al., 2019). In addition, the light absorption at 880 nm ($b_{\text{abs, BC}}(880 \text{ nm})$) was the most representative value of BC absorption due to BrC doesn't absorb at 880 nm (Drinovec et al., 2015; Lin et al., 2021). Hence, the light absorption at 370 and 880 nm was used to explore the optical properties of BrC and BC.

Fig. S13 shows the light absorption at the different wavelengths of BC and BrC during the normal and the lockdown periods. The mean $b_{\text{abs}}(880 \text{ nm})$ decreased from 31.2 ± 18.1 to $17.3 \pm 11.8 \text{ Mm}^{-1}$ (44.6%) and 21.0 ± 19.3 to $19.0 \pm 16.1 \text{ Mm}^{-1}$ (9.6%) in Shijiazhuang and Beijing, respectively, during the lockdown period. This illustrates the significant reduction of anthropogenic emissions during the lockdown period, especially from traffic exhaust. At the same time, the light absorption at 370 nm ($b_{\text{abs, total}}(370 \text{ nm})$) decreased by 42.4% (from 115.4 ± 64.9 to $66.5 \pm 50.4 \text{ Mm}^{-1}$) in Shijiazhuang during the lockdown period compared with that in the normal period because BC was the dominator of the light absorption substances even at 370 nm (Fig. S13). As shown in Fig. 2, although SOC increased significantly during the lockdown period in Shijiazhuang, both POC and EC significantly decreased. Thus, the significant decrease of light absorption in Shijiazhuang should be contributed to the decreases of POC and BC or EC due to anthropogenic emissions reductions. However, the $b_{\text{abs, total}}(370 \text{ nm})$ in Beijing showed a slight increase (3.9%, from 76.1 ± 63.4 to $79.2 \pm 67.3 \text{ Mm}^{-1}$) during the lockdown period. This can be explained by a slight decrease in POC and an obvious increase in SOC in Beijing (Fig. 2). The relative contribution of BrC to total absorption at 370 nm showed a slight increase of 2.5% (from 35.8% to 38.2%) in Shijiazhuang and 3.8% (from 36.8% to 40.6%) in Beijing, respectively, during the lockdown period (Fig. S13). This implies that secondary formation during regional transport of air mass might also contribute to the light adsorption of organic matters.

Fig. 4 shows the diurnal variations of the light absorption of BrC measured at 370 nm ($b_{\text{abs, BrC}}(370 \text{ nm})$). In Shijiazhuang, the $b_{\text{abs, BrC}}(370 \text{ nm})$ showed a similar diurnal pattern during the lockdown period like that during the normal period, while the absolute values during the lockdown period decreased significantly (Fig. 4A). The nighttime decrease of the $b_{\text{abs, BrC}}(370 \text{ nm})$ between these two periods was slightly smaller than the daytime counterpart in Shijiazhuang. In Beijing, besides a slight increase of the $b_{\text{abs, BrC}}(370 \text{ nm})$ during the lockdown period, the nighttime increase of the $b_{\text{abs, BrC}}(370 \text{ nm})$ between these two periods was obviously higher than that in the day (Fig. 4B). Fig. S14 shows the fraction of BrC in the total light adsorption at 370 nm. The diurnal pattern of the fraction of $b_{\text{abs, BrC}}(370 \text{ nm})$ showed a larger increase rate in nighttime during the lockdown period compared with that during the normal period in Shijiazhuang

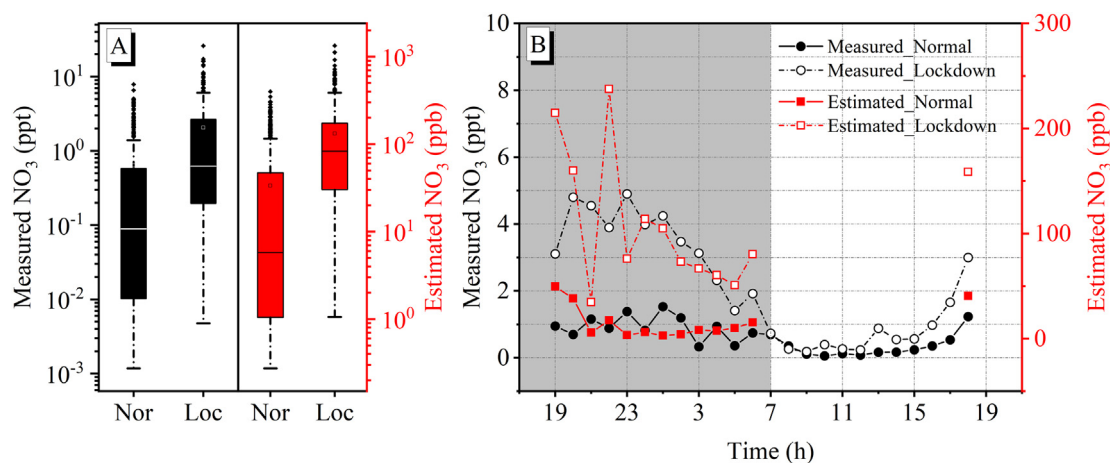


Fig. 3. (A) The concentration and (B) diurnal variations of measured and estimated NO_3 radicals of Beijing site.

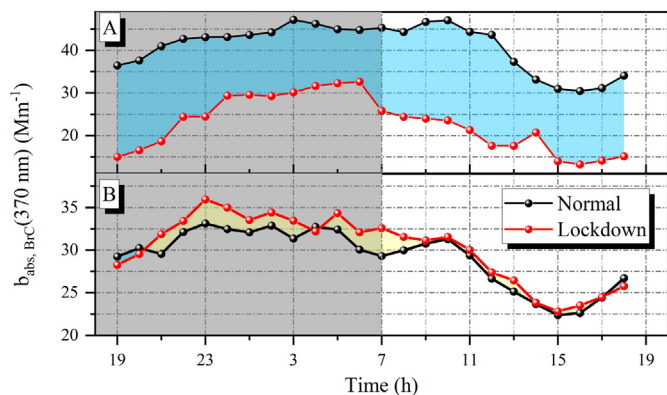


Fig. 4. The $b_{\text{abs, BrC}}(370 \text{ nm})$ diurnal variation of BrC of during the normal period (black line) and the lockdown period (red line) in (A) Shijiazhuang and (B) Beijing. The shadow area is used to indicate nighttime. The area between the two lines is filled with blue or yellow to distinguish the relative size of the different value between the normal period and the lockdown period. The yellow color indicates a positive difference, while the blue one means a negative difference.

(Fig. S14A). A similar phenomenon was observed at the Beijing site (Fig. S14B). These results imply that the nocturnal oxidation of VOCs or POC by NO_3 radicals might contribute to BrC generation. In addition, $b_{\text{abs, BrC}}(370 \text{ nm})$ showed a good correlation with SOC in polluted episodes ($\text{PM}_{2.5} > 75 \mu\text{g m}^{-3}$) during the lockdown period ($R^2 = 0.51$, Fig. S15), which was similar to the scatter plot of SOA vs. SOC (Fig. S10). Thus, it is reasonable to propose that BrC was an important contributor to SOC.

3.5. Backward trajectory and PSCF analysis of SOC

The potential sources are further analyzed for understanding the difference of SOC between the normal and the lockdown periods in the NCP region. The domain is from 70° E to 125° E and 30° N to 60° N , which was divided into 26,400 grid cells of $0.25^\circ \times 0.25^\circ$ latitude and longitude. The PSCF analysis for the hourly SOC data can provide information on regional transport and local emissions for carbonaceous aerosols. Fig. S16 shows the PSCF results of Shijiazhuang and Beijing during different periods. For Shijiazhuang, the areas with a high weighted potential source of SOC (with contribution function values >0.8) included Shijiazhuang and the regions between Hebei and Shanxi provinces (Fig. S16A and B), which agreed well with previous studies (Wang et al., 2018; Xie et al., 2019; Zong et al., 2018). For Beijing (Fig. S16C and D), the main potential

source areas of SOC were Beijing, the center of Inner Mongolia, the northern Shanxi province, and the central region of the Hebei province (Ji et al., 2018a; Zhang et al., 2013a). It has been pointed out that those potential areas were heavily polluted areas due to intensive pollutant emissions from industry, resident coal combustion and vehicle exhaust (Bie et al., 2021; Ji et al., 2018a; Xie et al., 2019; Zong et al., 2018).

To understand the variations of potential sources of SOC during lockdown periods, we subtracted the PSCF result during the normal period from that during the lockdown period (Fig. 5). Regions with a difference between 0.1 and -0.1 are denoted as colorless, which means that these regions contribute almost the same during normal and lockdown periods. The red color represents positive values, while the blue one represents negative. Larger absolute values represent a more significant difference of contribution in SOC. For Shijiazhuang, the southern areas of Shanxi province and the central regions of Inner Mongolia contribute more to the potential source of SOC in the normal period than that in the lockdown period, while Tianjin, Beijing, the eastern of Hebei province, the northern of Shandong province, and the southern of Liaoning province contribute more to SOC during the lockdown period. For Beijing, the contribution of central and western of Inner Mongolia (extend to the Mongolian regions) were decreased, while Tianjin, the southern of Hebei, the northern of Shanxi province, the northern of Shandong province, and the southern of Liaoning province contribute more during the lockdown period. Overall, the south-east regions of the NCP (including Beijing and Shijiazhuang) were the dominant contributors to SOC during the lockdown period, while a slight difference between these two locations was observable due to the different prevailing wind directions and the surrounded industry distribution.

4. Conclusion

The anthropogenic emissions in the NCP region were greatly reduced during the lockdown period caused by the COVID-19 pandemic, such as POC, NO , and NO_2 in Shijiazhuang (30.6, 87.9, and 41.8%) and in Beijing (4.0, 82.2, and 59.6%). However, the particle matter showed an unexpected increase during the lockdown period due to the adverse meteorological conditions and regional transport of air mass in Beijing. An increase in atmospheric oxidation capacity was observed during the lockdown period. The increased O_3 concentrations in Shijiazhuang (176.0%) and in Beijing (241.2%) were mainly due to the nonlinear O_3 production chemistry and the reduced titration of ozone by NO .

SOC significantly increased during the lockdown period in both Shijiazhuang (13.6%) and Beijing (53.0%) although the anthropogenic emissions drastically reduced. The diurnal pattern of SOC, SOA, and the absorption of BrC suggested an additional nighttime SOC source during the lockdown period in the NCP region. NO_3 radicals should play an important role in the formation of SOC in the nighttime because the nocturnal NO_3

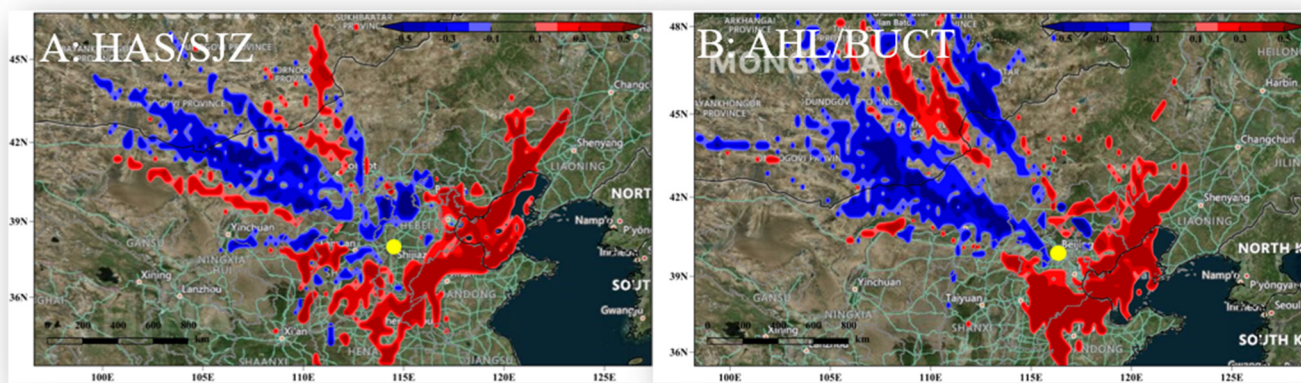


Fig. 5. Weighted potential source contribution function (WPSCF) difference value map for SOC arriving in the (A) Shijiazhuang and (B) Beijing region at the height of 100 m between the lockdown and normal periods.

concentrations increased significantly during the lockdown period when compared with that in the normal period. The southeast regions of NCP, northern regions of Shandong province, and the southern regions of Liaoning contributed more to the SOC during the lockdown period than the normal period. Our results mean that more attention should be paid to the nocturnal chemistry for further reduction PM concentration in China.

CRedit authorship contribution statement

Zemin Feng: Writing - original draft, Conducting experiments, Data curation. **Feixue Zheng:** Investigation, Data curation. **Yongchun Liu:** Conceptualization, Methodology, Data curation, Writing - review & editing. **Xiaolong Fan:** Investigation, Data curation. **Chao Yan:** Investigation, Data curation. **Yusheng Zhang:** Investigation, Data curation. **Kaspar R. Daellenbach:** Investigation, Data curation. **Federico Bianchi:** Investigation, Data curation. **Tuukka Petäjä:** Investigation, Data curation. **Markku Kulmala:** Investigation, Data curation. **Xiaolei Bao:** Investigation, Methodology, Data curation. All authors contributed to the paper with useful scientific discussions or comments.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.152191>.

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