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Changes in physical and chemical properties of urban atmospheric aerosols and ozone during the COVID-19 lockdown in a semi-arid region

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HIGHLIGHTS

- \(\text{O}_3\) increased by \(>100\%\) at urban sites in Lanzhou during the COVID-19 lockdown.
- Photochemical production rate of secondary inorganic aerosols (SIAs) increased.
- Increase in sulfur oxidation ratio accounted for 48.7\% of the total sulfate.
- Increased VOCs to NOx ratio and decreased aerosol enhanced \(\text{O}_3\) production.
- Light extinction of organic matter relatively increased while that of SIAs decreased.

ARTICLE INFO

**Keywords:**
Atmospheric aerosols
Ozone
Single scattering albedo
Collaborative management

**ABSTRACT**

The synergistic response of urban atmospheric aerosols and ozone (\(\text{O}_3\)) to reduction of anthropogenic emissions is complicated and still needs further study. Thus, the changes in physical and chemical properties of urban atmospheric aerosols and \(\text{O}_3\) during the Coronavirus Disease 2019 (COVID-19) lockdown were investigated at three urban sites and one rural site in Lanzhou with semi-arid climate. Fine particulate matter (PM\(_{2.5}\)) decreased at four sites by \(~20\%\) while \(\text{O}_3\) increased by \(>100\%\) at two urban sites during the COVID-19 lockdown. Both primary emissions and secondary formation of PM\(_{2.5}\) decreased during the lockdown. Significant increase in both sulfur and nitrogen oxidation ratios was found in the afternoon, which accounted for 48.7\% of the total sulfate and 40.4\% of the total nitrate, respectively. The positive matrix factorization source apportionment revealed increased contribution of secondary formation and decreased contribution of vehicle emissions. Aerosol scattering and absorption decreased by 33.6\% and 45.3\%, resulting in an increase in visibility by 30\% and single scattering albedo (SSA) at 520 nm slightly increased by 0.02. The enhanced \(\text{O}_3\) production was explained by increased volatile organic compounds to nitrogen oxides ratio, decreased aerosol, as well as increased SSA. The primary emissions of secondary aerosol precursors significantly decreased while Ox (i.e., NO\(_2\) and \(\text{O}_3\)) exhibited little change. Consequently, Ox to CO ratio, PM\(_{2.5}\) to elemental carbon (EC) ratio, secondary inorganic aerosols to EC ratio, and secondary organic carbon to EC ratio increased, confirming enhanced secondary aerosol production efficiency during the lockdown. Positive feedback among \(\text{O}_3\) concentration, secondary aerosol formation, and SSA was revealed to further promote \(\text{O}_3\) production and secondary aerosol formation. These results provide scientific guidance for collaborative management of \(\text{O}_3\) and particulate matter pollution for cities with semi-arid climate.

1. Introduction

Atmospheric aerosols and ozone (\(\text{O}_3\)) significantly impact on air quality, climate change and public health (IPCC, 2013; Tian et al., 2017; Li et al., 2019b; Ma et al., 2022). To mitigate air pollution and protect public health, China has introduced the strictest air pollution control.
measures since 2013, and air quality has improved significantly (Li et al., 2019a; Wang et al., 2020a; Ma et al., 2020, 2021b; Zhang et al., 2020; Zhao et al., 2021). However, China still experiences severe air pollution under unfavorable meteorological conditions and via regional transport (Li et al., 2019c; Huang et al., 2021). The improvement in visibility is also not significant in many cities because of the increased fraction of secondary particulate matter (PM) species such as nitrate (Ma et al., 2019; Liu et al., 2020). The Chinese government has adopted several stringent short-term emission control measures, which has been successful in some important national events such as the 2008 Beijing Olympics, the 2014 Asia-Pacific Economic Cooperation (APEC) Summit, the 2015 China Victory Day Parade, and the 2016 G20 Summit (Wang et al., 2016; Sun et al., 2016; Li et al., 2017). Overall, such emission control measures are valuable in studying the impacts of emission controls on air quality and help propose reasonable and effective emission reduction measures.

Due to the spread of the Coronavirus Disease (COVID-19) at the end of 2019, the Chinese government implemented strict restrictions including reducing public transport, closing markets and small industries, and banning public gatherings. These prevention measures effectively mitigated the spread of the COVID-19 epidemic and significantly reduced anthropogenic emissions in China, which provided a unique opportunity to assess the impact of primary emission reductions on atmospheric aerosols and O3. The average concentrations of fine particulate matter (PM2.5 particles less than 2.5 μm in diameter), inhalable particulate matter (PM10 particles less than 10 μm in diameter), sulfur dioxide (SO2), nitrogen dioxide (NO2), and carbon monoxide (CO) over mainland China decreased by 14%, 15%, 12%, 16%, and 12%, respectively, while O3 concentration increased by 9% in 2020 compared to 2019 (Chen et al., 2020b). The decreased PM and gaseous pollutants were mainly caused by reduced emissions from transport and industry and varied with regions (Wang et al., 2020b). For example, nitrate decreased by ~60% and was the main contributor to the decline in PM2.5 in Shanghai (Chen et al., 2020a), the proportion of nitrate in PM2.5 increased while that of sulfate decreased in Tangshan (Li et al., 2021), and the decrease in submicron particles (PM1; particles less than 1 μm in diameter) was also recorded in Xi’an (Tian et al., 2021). Furthermore, studies revealed that enhanced atmospheric oxidizing capacity increased secondary aerosol formation, which partially offset the reduction of primary emissions during the lockdown (Huang et al., 2020; Li et al., 2021; Liu et al., 2021b). However, due to different meteorological changes and different levels of nitrogen oxides (NOx; both nitric oxide and nitrogen dioxide) and volatile organic compounds (VOCs) reductions, O3 decreased in the south but increased in most other regions during the lockdown (Li et al., 2021b). Specifically, O3 increased by more than 100% in Wuhan (Yao et al., 2021), Tangshan (Li et al., 2021), Chongqing (Chen et al., 2020c), and some other cities. Previous studies clarified the changes in PM and O3 concentrations, PM chemical species, and revealed the mechanisms of the enhanced secondary aerosol formation during the lockdown. However, the changes in aerosol physical properties were poorly studied, especially the possible relationships among aerosol physical properties, chemical species, and O3 formation during the lockdown.

Lanzhou is a typical valley city in the semi-arid region of northwest China, where coal is the main fuel used for residential heating in winter (Tan et al., 2017). Previous studies suggest that secondary inorganic aerosols (SIAs) in Lanzhou in winter were mainly from photochemical reactions (Du et al., 2020) and motor vehicle emissions were a major contributor to PM pollution in Lanzhou (Wang et al., 2021). A recent study revealed that PM2.5 reduced by 50% during the lockdown, mainly contributed by the reduction in secondary species (Xu et al., 2020). This reduction in secondary formation was opposite to the increased secondary formation of atmospheric aerosols in other studies (Huang et al., 2021a; Liu et al., 2021a; Tian et al., 2021). However, results in Xu et al. (2020) were derived from PM1 while results of the other studies were from PM2.5. Thus, further study is required to reveal the formation mechanisms of PM2.5 as well as O3 in Lanzhou during the lockdown. Further study on the consequently changes in optical properties of atmospheric aerosols during the lockdown was also required to better investigate the improvement in atmospheric visibility.

The present study investigated the changes in physical and chemical properties of urban atmospheric aerosols and O3 during the COVID-19 lockdown at three urban sites and one rural site in Lanzhou. The sites, data, and methods were introduced in Section 2. Section 3 presented results and discussion on temporal evolution of major pollutants before and during the lockdown, change in chemical species and sources during the lockdown, change in optical properties of atmospheric aerosols during the lockdown, enhanced O3 and production rate of secondary aerosols, and comparisons between relative PM1 and PM2.5. The results were finally concluded in Section 4.

2. Data and methodology

2.1. Sites and data

The used data were collected at the Lanzhou Atmospheric Components Monitoring Superstation (LACMS; 36.05° N, 103.87° E, 1520 m a.s.l.), which is settled on the campus of Lanzhou University at downtown area of Lanzhou City. Multiple online instruments were deployed to continuously measure the PM concentrations with different sizes, aerosol optical properties, chemical compositions, gases, atmospheric visibility, and meteorological parameters. Detailed introduction of the LACMS site, instruments, and data are in Table S1 and available elsewhere (Du et al., 2020; Guan et al., 2021; Wang et al., 2021). PM2.5 chemical composition data at the LACMS site were missing from February 10, 2020 to March 3, 2020. Data from three monitoring sites of the Ministry of Ecology and Environment (MEE) network were also used. The population of Lanzhou is concentrated in the Lanzhou Basin, so Lan Lian (LL) and Sheng Wu Suo (SWS) in the basin were chosen for urban representation and Yu Zhong (YZ) was selected as a rural site (Fig. 1).

2.2. A brief introduction to parameters

Organic matter (OM) was determined from organic carbon (OC) multiplied by a mass conversion factor of 1.6 (Turpin and Lim, 2001; Xing et al., 2013; Chow et al., 2015). The sulfur and nitrogen oxidation ratios (SOR and NOR) are generally used to infer the degree of atmospheric secondary transformation of SO2 and NO2 to sulfate and nitrate, which were calculated by the following equations (Ohta and Okita, 1990):

\[ \text{SOR} = \frac{[\text{SO}_4^{2-}]}{([\text{SO}_4^{2-}] + [\text{SO}_3^-])} \]  
\[ \text{NOR} = \frac{[\text{NO}_3^-]}{([\text{NO}_3^-] + [\text{NO}_2^-])} \]

where [SO4^{2-}] and [NO_3^-] are the molar concentrations (μmol·m⁻³) in
PM$_{2.5}$ and [SO$_2$] and [NO$_2$] are the molar concentrations (μmol·m$^{-3}$) in gas phase. SOR and NOR >0.1 indicate secondary generation of sulfate and nitrate.

Unless otherwise specified, $\sigma_{sp}$ is the aerosol scattering coefficient at 520 nm and $\sigma_{ap}$ is the aerosol absorption coefficient at 520 nm. The single scattering albedo (SSA) was calculated from $\sigma_{sp}$ and $\sigma_{ap}$ at 520 nm:

$$\text{SSA} = \frac{\sigma_{sp}}{\sigma_{sp} + \sigma_{ap}}$$

(3)

The scattering Ångström exponent (SAE) is a parameter related to aerosol size that was calculated as follows (Guan et al., 2021):

$$\text{SAE} = -\frac{\log(\sigma_{sp,\lambda_1}) - \log(\sigma_{sp,\lambda_2})}{\log(\lambda_1) - \log(\lambda_2)}$$

(4)

where $\lambda_1 = 520$ and $\lambda_2 = 450$ nm. SAE is relatively low for large particles and high for small particles. The relationship between $\sigma_{ap}$ and wavelength is expressed as (Liu et al., 2018):

$$\sigma_{ap}(\lambda) = k\lambda^{-\text{AAE}}$$

(5)

where $k$ is a constant, $\sigma_{ap}(\lambda)$ is the absorption coefficient at wavelength $\lambda$, and AAE is absorption Ångström exponent. Thus, AAE was calculated via power-law fitting based on the absorption coefficients at 370, 470, 520, 590, 660, 880, and 950 nm wavelengths. AAE is an indicator of the dominant light absorber in aerosols.

2.3. A brief introduction to models

The revised Interagency Monitoring of Protected Visual Environments (IMPROVE) algorithm (Pitchford et al., 2007) was used to evaluate the effect of chemical species on aerosol light extinction. The algorithm details are available in a recent study (Guan et al., 2021) and in the Supplementary materials.

The source apportionment was conducted using the Positive Matrix Factorization (PMF) 5.0 model, technical details are available in a previous study (Wang et al., 2021) and in the Supplementary materials. The tropospheric ultraviolet radiation (TUV) model, which was jointly developed by Madronich and Flocke (1998) to calculate the tropospheric ultraviolet radiation and some of the visible radiation, was also used. The model considers wavelengths of 121–735 nm, which allows it to calculate the ultraviolet irradiance, actinic flux, and photolysis rate of matter (i.e., NO$_2$). The radiation was calculated by solving the radiative transmission equation using the 8-stream scheme (Liu et al., 2019).

Fig. 2. Time series and statistics of major air pollutants (PM$_{2.5}$, PM$_{10}$, CO, SO$_2$, NO$_2$, O$_3$) before and during the COVID-19 lockdown in Lanzhou. Light blue and yellow shades represent P1 and P2, respectively. YZ is a rural site while SWS, LL, and LACMS are urban sites.
3. Results and discussion

3.1. Temporal evolution of major pollutants before and during the lockdown

A total of 40 days from January 26, 2020 to March 5, 2020 (the same as in Fan et al., 2021) was defined as the COVID-19 lockdown period (P2; Fig. 2). Correspondingly, the period from December 20, 2019 to January 23, 2020 (P1; also 40 days) was chosen for comparison reasons before the COVID-19 lockdown. The period of 24–25 January 2020 (i.e., New Year’s Eve and Chinese New Year) was skipped to avoid emissions from large amount of celebration fireworks.

The trends of major pollutants at the four sites were the same from P1 to P2 except PM10 that was mainly from natural sources (Fig. 2; Table S1). PM2.5 decreased by 15.3%, 25.2%, 23.7%, and 24.0% at YZ, LL, SWS, and LACMS, respectively. PM10 decreased by 9.9%, 13.7%, and 16.1% at LL, SWS, and LACMS, respectively, while increased by 9.7% at the site of YZ. Gaseous pollutants of CO, NO2, and SO2 all decreased at four sites. Although values may change, the reductions in SO2 were the smallest among the three gaseous pollutants at each site. Higher reduction in NO2 than SO2 indicated higher reduction of vehicular emissions and weak reduction of industrial emissions. NOx reduced by 53.6% at LACMS (Table S1), where vehicular emissions were estimated to be the largest contributor (36.0%) to PM2.5 in winter (Wang et al., 2021). Significant reductions in gaseous pollutants and PM2.5 indicated large reductions of anthropogenic emissions during the COVID-19 lockdown in Lanzhou. Lower reduction in PM2.5 at the rural site of YZ than three urban sites further indicated the weakening of anthropocentric activities during the lockdown. Opposite to the decrease trend in gaseous pollutants and PM2.5, a very significant increase was found for O3 at the four sites, especially at the urban sites. Specifically, O3 increased by 36.9%, 110.5%, 78.5%, and 103.7% at the site of YZ, LL, SWS, and LACMS, respectively. O3 were even more than doubled at LL and LACMS. The decrease in O3 at the rural site was much lower than that at urban sites while the decrease in PM2.5 was the least at the rural site. Large decrease in PM2.5 at urban sites was generally explained by dramatically weakened human activities. The reason for the significant increase in O3 is studied in Section 3.4. The reduction in PM2.5 (~20%) was comparable to observation results in North China Plain (Li et al., 2021; ~18%) and lower than those in Shanghai, Wuhan, Chongqing, and Hangzhou (Chen et al., 2020a; Chen et al., 2020b; Yao et al., 2021; Yuan et al., 2021; 30–50%). The increase in O3 of more than 100% was also found in other cities such as Wuhan, Tangshan, Chongqing (Qiao et al., 2021; Chen et al., 2020c; Li et al., 2021).

The influence of the COVID-19 lockdown on major pollutants were also revealed by the changed diurnal variations in these pollutants (Figs. S1–S4). The concentration of PM2.5 decreased at the four sites at every hour of the day. The relative diurnal variation of PM2.5 at the rural site of YZ during P2 was like that during P1 while noticeably changed at the three urban sites. Specifically, the P1 period was characterized by a double peaked diurnal variation of PM2.5, which was enhanced by morning and evening human activities. These peaks weakened during P2 due to decreased human activities and diurnal variation seemed to be more controlled by diurnal variation of diffusion conditions including the atmospheric boundary layer (Zhang et al., 2021). Significant change was found for NO2 during the lockdown period. NO2 continuously accumulated during daytime and began to decrease after the evening rush hours during P1. Opposite to the daytime accumulation during P1, NO2 exhibited a trough during P2 in the afternoon when diffusion condition was the best of a day. Small change was found for the diurnal variations of SO2 and CO. The peak values of O3 significantly increased during the lockdown at the four sites while trough values of O3 kept the same at three urban sites.

3.2. Change in chemical species and sources during the lockdown

Variation in chemical species of PM2.5 during the lockdown was investigated to reveal the chemical mechanisms of fine PM pollution (Fig. 3). OM and SIAs were the major species both before and during the lockdown. Fine soils (FS), chloride and elemental carbon (EC) were minor PM2.5 species. All PM2.5 species decreased during the lockdown. The contribution of SIAs slightly decreased from 42.5% in P1 to 39.0% in P2 while OM increased from 36.8% in P1 to 40.1% in P2. The decrease in nitrate contribution (3.1%) was higher than that in sulfate contribution (1.0%), which was consistent to the higher reduction in NOx than SO2. The contribution of EC, which was considered as a reference for primary fine PM emission, decreased from 4.2% in P1 to 2.5% in P2. Thus, both primary emission and secondary formation of fine PM decreased during the lockdown. The increase in K’ was attributed to celebrating fireworks during the Spring Festival Holidays, which was also recorded by a previous study (Liu et al., 2021a).

The ratios between specific species were studied to better understand chemical mechanisms of fine PM pollution during the lockdown (Fig. 3). The ratio of PM2.5 to EC increased by 79% from P1 to P2, which indicated relatively significant increase in secondary fine PM during the lockdown. The ratio of sulfate to nitrate increased from 0.88 to 1.16, indicating enhanced emissions from stationary sources (industrial emissions) and reduced emissions from mobile sources (i.e., vehicular emissions). The ratio of nitrate to EC increased despite the great reduction in NOx. The increase in the ratio of sulfate to EC was higher than that of nitrate to EC. As a result, the ratio of SIAs to EC increased from 16.3 in P1 to 20.2 in P2. Both the ratios of OC to EC and secondary organic carbon (SOC) to EC significantly increased during the lockdown. The ratio of SOC to OC slightly increased from P1 to P2, leading to higher ratio of SOC to EC than OC to EC. Significant increase in both ratios of SIAs to EC and SOC to EC indicated relatively enhancement of secondary formation of fine PM during the lockdown. On this premise, increased OM contribution but decreased SIAs contribution to PM2.5 indicated higher enhancement of SOC than SIAs.

Relationships between OC and EC were further studied to investigate the behavior of carbonaceous species during the lockdown (Fig. S5). Carbonaceous species are mainly originated from primary emissions of coal combustion, vehicles, and biomass burning and secondary formation in the atmosphere. Since cold season residential heating in Lanzhou was powered by coal and biomass burning was little, the change in emission source of carbonaceous species was mainly in vehicles. The increase in the ratio of OC to EC was mainly contributed by the increase in SOC rather than primary organic carbon. Specifically, OC to EC ratio of 7.3 in P1 while that of 10.9 in P2.

The diurnal variations of SOR, NOR, and related gaseous and particulate species were investigated to better understand the formation of SIAs during the lockdown (Fig. 4). Extremely low NOR of 0.11 and very low SOR of 0.19 were found during P1 partially due to semi-arid climate environment (Du et al., 2020). Significant increase in both NOR and SOR was found in the afternoon (i.e., 12:00 to 20:00). Peak values of NOR (16:00) and SOR (17:00) increased by 66.1% and 36.7%, respectively. This significant increase of NOR and SOR in the afternoon indicated enhanced photochemical production rate of SIAs during the lockdown. Furthermore, nitrate peaked while NO2 exhibited a trough in the afternoon, which clearly indicated reduced primary emissions of gaseous pollutants while enhanced secondary production rate of fine PM during the lockdown. SOR and NOR exhibited little change during nighttime and in the morning, indicating little change in aqueous phase formation rate of SIAs. Specifically, chemical species during the lockdown but NOR and SOR before the lockdown were used to estimate SIAs induced by the increased production rate during the lockdown (green lines in Fig. 4). The production of sulfate by increased SOR accounted for 48.7% of the total sulfate while that of nitrate by increased NOR accounted for 40.4% of the total nitrate (Fig. 4 and Table S2). Although NOR and SOR were low before and during the lockdown, the increase in SOR and NOR
during the lockdown was evident in the afternoon. This phenomenon is different from results conducted in Hangzhou, where SOR and NOR obviously increased both during daytime and at night (Liu et al., 2021a). Thus, the photochemical conversion of SIAs during the afternoon increased while aqueous phase production during the nighttime exhibited little change during the lockdown.

The change in fine PM sources during the lockdown was confirmed using PMF source apportionment (Fig. 3). The vehicular emissions were the greatest fine PM source both in P1 and P2. The secondary formation, coal combustion, and mental smelting industries were also major sources of PM$_{2.5}$. Decrease in vehicular emissions and increase in secondary formation of fine PM were clearly revealed by the PMF source apportionment. Specifically, the contribution of vehicular emissions to PM$_{2.5}$ decreased from 42.0% in P1 to 37.0% in P2 while that of...
secondary formation increased from 26.0% to 30.0%. Decrease in smelting industrial emissions was caused by production reduction while increase in power plant emissions was explained by enhanced residential electricity. The dust event in P2 was also identified by the PMF source apportionment.

3.3. Change in optical properties of atmospheric aerosols during the lockdown

Aerosol scattering and absorption decreased by 33.6% and 45.3%, respectively during the lockdown (Fig. 5). Consequently, visibility increased by 30% and SSA at 520 nm slightly increased by 0.02 (from 0.86 to 0.88). Higher reduction in absorption than scattering was attributed to reduction in primary emissions of both scattering and absorbing species while enhanced secondary aerosols were mainly scattering species. Vehicular emitted EC was the main absorbing species at LACMS during winter (Wang et al., 2021). Thus, the reduction in absorption was mainly caused by reduced vehicular emissions during the lockdown. The relatively increased SSA might contribute to higher absorbing species while enhanced secondary aerosols were mainly increased by 30% and SSA at 520 nm slightly increased by 0.02 (from 3.3. Change in optical properties of atmospheric aerosols during the lockdown

such changes in aerosol optical properties during the lockdown in Lanzhou are different from results in Wuhan, where higher reduction of aerosol scattering than absorption led to slight decrease in SSA (from 0.92 to 0.91) (Yao et al., 2021). Although the decrease in aerosol scattering and absorption coefficients was comparable (~30–40%) in these two cities, the visibility improvement in Wuhan (106.7%) was much higher than in Lanzhou (17.8%). An important reason was the very large reduction of ammonium nitrate in Wuhan while slight decrease in Lanzhou.

3.4. Enhanced O₃ and production rate of secondary aerosols

Urban ground atmospheric O₃ is mainly produced by photochemical reactions with NOx and VOCs as precursors. The emissions of precursors, solar radiation intensity, and physical processes such as horizontal and vertical transportation mainly determine the concentration of ground O₃ (Sun et al., 2019; Ni et al., 2018; Liu et al., 2019). Production of ground O₃ is nonlinearly governed by NOx and VOCs (Heus et al., 2003; Qu et al., 2018). Change in NOx to VOCs might contribute to higher O₃ production during the lockdown. PM in the atmosphere indirectly affects the ground O₃ by (1) reduces the solar radiation that reaches the ground and thus reduces photolysis frequency (Dickerson et al., 1997; Jacobson, 1998); and (2) absorbs free radicals that related to O₃ photochemistry and reduces O₃ production. Thus, PM reduction during the lockdown might also lead to higher O₃ production. In addition to PM loading, light scattering and absorbing properties of atmospheric aerosols may change the ultraviolet radiation intensity and affect O₃ production. Increased SSA during the lockdown indicated relatively more scattering and less absorbing of solar radiation by aerosols, which may also contribute to higher O₃ production.

The increase of O₃ during the lockdown exhibited obvious diurnal variation with very large inearcrease in the afternoon, which exceeded 60 μg m⁻³ at 17:00 to 18:00 (Fig. 7). The increase of O₃ was relatively low during nighttime and little increase in O₃ was found in the morning (i.e., 07:00 and 08:00); then continuously increased until 18:00 and
began to decrease until the next morning. Little increase in minimum diurnal value while very large increase in maximum value led to significant increase of 103.7% in average O$_3$ at LACMS during the lockdown. The downtown area of Lanzhou was revealed to be VOCs limited region (Li et al., 2020) and average VOCs to NOx ratio increased from 0.9 in P1 to 1.5 during the lockdown, which was a major reason for the increase in O$_3$. The maximum increases in O$_3$ and VOCs to NOx ratio were at 18:00, which confirmed the contribution of the change in VOCs to NOx ratio to O$_3$ increase. However, the increase in VOCs to NOx ratio was only relatively high from 17:00 to 20:00, which did not support the large O$_3$ increase from 12:00 to 16:00 and suggested other reasons for O$_3$ increase in addition to the increase in VOCs to NOx ratio.

The increase in ultraviolet radiation might also be another major reason for the O$_3$ increase during the lockdown (Fig. 7). The increase in ultraviolet radiation began to increase in morning and kept at relatively high values from 11:00 to 17:00. A combination of diurnal variation of the increases in VOCs to NOx and ultraviolet radiation reasonably explained the diurnal variation of the O$_3$ increase. Specifically, small O$_3$ increase in the morning (08:00 to 11:00) was caused by small increase in both ultraviolet radiation and VOCs to NOx ratio, the O$_3$ increase in the afternoon was mainly contributed by increase in ultraviolet radiation (12:00 to 17:00), the O$_3$ increase in the evening was mainly contributed by increase in VOCs to NOx ratio (18:00 to 20:00), and the small nighttime O$_3$ increase was governed by small increase in VOCs to NOx ratio (21:00 to the next morning). The increase in ultraviolet radiation was higher than that caused by only annual cycle of the solar ultraviolet radiation (Fig. 7). The extra increase in ultraviolet radiation (Fig. S6) might be explained by decreased PM in the atmosphere, which was confirmed by the maximum increase in ultraviolet radiation and maximum decrease in PM$_{2.5}$ at 14:00. To sum up, both natural increase in ultraviolet radiation caused by annual solar cycle and anthropogenic increase in ultraviolet radiation caused by decreased PM contributed to O$_3$ increase during the COVID-19 lockdown in Lanzhou.

The reduction in aerosol scattering and absorption and increase in O$_3$

![Fig. 6. Aerosol optical parameters and visibility before and during the COVID-19 lockdown at LACMS.](image1)

![Fig. 7. Change in PM$_{2.5}$, O$_3$, the ratio of VOCs to NOx, and ultraviolet radiation during the COVID-19 lockdown in Lanzhou. Change in ultraviolet radiation by natural annual cycle, red line in Fig. 7c, was calculated using the TUV model.](image2)
during the lockdown are clearly shown in Fig. 8. Relatively low SSA in P1 indicated strong aerosol absorption in Lanzhou (Tian et al., 2018a; Guan et al., 2021, 2022). Generally, lower aerosol scattering/absorption coefficient corresponded to higher O$_3$ while higher SSA related to higher O$_3$ both in P1 and P2. Data samples of the same SSA value in P2 corresponded to higher O$_3$ than in P1, which was mainly caused by increased VOCs to NOx ratio and enhanced ultraviolet radiation. Nevertheless, data samples of SSA $<0.80$ decreased while those of SSA $>0.90$ increased from P1 to P2, leading to higher average SSA in P2 than P1. Thus, relatively weaker absorption and stronger scattering of solar radiation was more favorable for O$_3$ production. Furthermore, O$_3$ increased faster with increasing SSA in P2 than P1, indicating that the influence of aerosol optical properties on O$_3$ was enhanced during the lockdown. Relationships among O$_3$, SSA, and secondary organic/inorganic aerosols will be further discussed below.

The significantly increased O$_3$ might lead to higher oxidation ability of the atmosphere and result in higher production of secondary organic and inorganic aerosols. Since the emission had greatly decreased during the lockdown, this issue was investigated based on PM species relative to EC and gaseous species relative to CO (Fig. 9). SIAs decreased throughout the day while SOC decreased during daytime but increased during nighttime during the lockdown. Total oxidants (Ox, i.e., O$_3$ and NO$_2$) exhibited little change during the lockdown due to increased O$_3$ while decreased NO$_2$. Consequently, relative Ox (i.e., NO$_2$ and O$_3$ relative to CO) largely increased during the lockdown, which may promote the secondary production of aerosols. Both relative SOC and SIAs increased during the lockdown and the increase of relative SOC was higher than that of relative SIAs. In contrast to one peak of relative SIAs in the afternoon, relative SOC exhibited two comparative peaks with one in the afternoon and one during nighttime. The afternoon peaks in relative SIAs and SOC was attributed to increased secondary formation caused by increased relative Ox and ultraviolet radiation.

The above results suggested a positive-feedback mechanism among O$_3$ concentration, secondary organic/inorganic aerosol formation, and aerosol SSA. The increase in O$_3$ concentration increased the oxidation capacity of the atmosphere and thus enhanced the formation of secondary organic/inorganic aerosols, which has been suggested by previous studies based on numerical modelling and observation data (e.g., Chan et al., 2017; Wang et al., 2016; Huang et al., 2020; Kang et al., 2021; Le et al., 2020; Li et al., 2021). The secondary formed inorganic aerosols and most of the organic aerosols were efficient in scattering and inefficient in absorbing the solar radiation, leading to higher SSA of the total atmospheric aerosols (Deng et al., 2016; Wang et al., 2021). In turn, aerosols with higher SSA relatively absorbed less while scattered more solar radiation, which promoted the formation of O$_3$ (Liu et al., 2019; Ma et al., 2021a). The above three mechanisms have been studied separately in literatures. Interestingly, combining them results in a positive-feedback mechanism: increase in O$_3$ enhanced secondary aerosol formation, secondary aerosols with high scattering efficiency led to higher aerosol SSA, aerosols with higher SSA promoted the formation of O$_3$ in turn. This positive-feedback mechanism among O$_3$ concentration, secondary organic/inorganic aerosol formation, and aerosol SSA might further increase O$_3$ and secondary aerosol formation to some extent.

### 3.5. Comparisons between relative PM$_1$ and PM$_{2.5}$

Comparisons between PM$_1$ and PM$_{2.5}$ were conducted since previous studies revealed contrast trends of secondary formation between PM$_1$ in Lanzhou and PM$_{2.5}$ in other cities (Xu et al., 2020). Both PM$_1$ and PM$_{2.5}$ exhibited significant peaks at 13:00 and weak peaks at 21:00 before the lockdown (Fig. 10). PM$_1$ and PM$_{2.5}$ decreased by 31.6% and 24.0%, respectively, during the lockdown. The diurnal variation of PM$_1$ was like that of PM$_{2.5}$ before and during the lockdown. The PM$_1$ to PM$_{2.5}$ ratio rapidly increased from 08:00 to 13:00 and kept at a peak value of ~0.70 until 17:00, then rapidly decreased to 0.63 at 20:00 before the lockdown. Thus, the afternoon peak was mainly attributed to photochemical production of secondary aerosols. The PM$_1$ to PM$_{2.5}$ ratio during the nighttime gradually decreased to its minimum value of 0.59 in the morning of the next day, which was originated from the gradual increase in PM size caused by aqueous phase conversion and hygroscopic growth. The PM$_1$ to PM$_{2.5}$ ratio during the lockdown was smaller than that before the lockdown. Opposite to the afternoon peak in the afternoon before the lockdown, the PM$_1$ to PM$_{2.5}$ ratio exhibited a trough during the lockdown in the afternoon. The PM$_1$ to PM$_{2.5}$ ratio reached its minimum value at 14:00 and rapidly increased to relatively higher values. The PM$_1$ to PM$_{2.5}$ ratio during the lockdown gradually increased after 01:00 and became close to that before the lockdown in the next morning. The reduction of PM$_1$ to PM$_{2.5}$ ratio, especially during the daytime, provided a reasonable explanation for the contrast trends of secondary formation between PM$_1$ and PM$_{2.5}$. This was further illustrated by the absolute reductions in PM$_1$ and PM$_{2.5}$, i.e., almost all (96.0%) the reduction in PM$_{2.5}$ was contributed by the reduction in PM$_1$. The overall reduction in PM$_1$ was reasonably attributed to the weakened primary emissions from anthropogenic activities (Xu et al., 2020).

### 4. Summary and conclusions

The Coronavirus Disease 2019 (COVID-19) lockdown provided a unique opportunity to investigate the synergistic response of urban atmospheric aerosols and ozone (O$_3$) to reduction of anthropogenic emissions, which is complicated and needs further study. The present study investigated the changes in physical and chemical properties of urban atmospheric aerosols and O$_3$ during COVID-19 lockdown in Lanzhou with semi-arid climate. A total of 40 days from January 26, 2020 to March 5, 2020 was defined as the COVID-19 lockdown period.

![Fig. 8. Relationships between aerosol optical properties (i.e., scattering coefficient, absorption coefficient, and SSA) and O$_3$ before and during the COVID-19 lockdown in Lanzhou.](image-url)
Fig. 9. Diurnal variation of SIAs, SOC, and Ox before and during the COVID-19 lockdown in Lanzhou.

Fig. 10. Diurnal variation of PM$_{1}$, PM$_{2.5}$, PM$_{1}$ to PM$_{2.5}$ ratio, and change in PM$_{1}$ and PM$_{2.5}$ before and during the COVID-19 lockdown in Lanzhou.
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(P2) and the period from December 20, 2019 to January 23, 2020 (P1) was chosen for comparison.

Fine particulate matter (PM$_{2.5}$) decreased at four sites from 15.3% to 25.2% while O$_3$ increased by >100% at two urban sites during the COVID-19 lockdown. All PM$_{2.5}$ species decreased during the lockdown. The contribution of secondary inorganic aerosols (SIAs) slightly decreased from 42.5% in P1 to 39.0% in P2 while organic matter (OM) increased from 36.8% in P1 to 40.1% in P2. The large increase in ratios of secondary inorganic and organic aerosols to elemental carbon (EC) indicated relatively enhancement of secondary aerosol formation than primary emission during the lockdown. Positive Matrix Factorization (PMF) source apportionment also revealed that the contribution of vehicular emissions to PM$_{2.5}$ decreased from 42.0% in P1 to 37.0% in P2 while that of secondary formation increased from 26.0% to 30.0%. The enhancement of secondary organic aerosols was higher than that of SIAs. Significant increase in both sulfur and nitrogen oxidation ratios was found in the afternoon, which accounted for 48.7% of the total sulfate and 40.4% of the total nitrate, respectively. The PMF source apportionment revealed increased contribution of secondary formation and decreased contribution of vehicle emissions.

Aerosol scattering and absorption decreased by 33.6% and 45.3%, resulting in an increase in visibility by 30% and single scattering albedo (SSA) at 520 nm slightly increased by 0.02. Higher reduction in absorption than scattering was attributed to reduction in primary emissions of both scattering and absorbing species while enhanced secondary aerosols were mainly scattering species. The contributions of SIAs and EC to light extinction decreased while that of organic matter (OM) increased during the lockdown. Reduction in SIAs and EC contributions was mainly attributed to reduced emissions while increased contribution of OM was the result of relatively increased secondary organic carbon (SOC).

The enhanced O$_3$ production was explained by increased volatile organic compounds (VOCs) to NOx ratio, decreased aerosol, as well as increased SSA. Specifically, the average VOCs to NOx ratio increased from 0.9 before to 1.5 during the lockdown, which was a major reason for the increase in O$_3$ since the maximum increases in both O$_3$ and VOCs to NOx ratio were at 18:00. The extra increase in ultraviolet radiation addition to that caused by only annual cycle was be explained by decreased aerosol in the atmosphere, which was confirmed by the maximum increase in ultraviolet radiation and maximum decrease in PM$_{2.5}$ at 14:00. Both natural increase in ultraviolet radiation caused by annual solar cycle and anthropogenic increase caused by decreased aerosol contributed to O$_3$ increase. Data samples of SSA <0.80 decreased while those of SSA >0.90 increased during the lockdown, which resulted in higher O$_3$ production.

The primary emissions of secondary aerosol precursors (i.e., SO$_2$, NOx, and VOCs) significantly decreased while Ox (i.e., NO$_2$ and O$_3$) exhibited little change. Consequently, Ox/CO, PM$_{2.5}$/EC, SIAs/EC, and SOC/EC increased, confirming enhanced secondary aerosol production efficiency during the lockdown. Positive feedback among O$_3$, secondary aerosols, and SSA was revealed to further promote O$_3$ production and secondary aerosol formation during the lockdown.

The present study revealed the synergistic response of urban atmospheric aerosols and O$_3$ to reduction of anthropogenic emissions in a basin city of Lanzhou with semi-arid climate. The results provide scientific guidance for collaborative management of O$_3$ and particulate matter pollution. New insights of the positive feedback among O$_3$, formation of secondary aerosols, and SSA was also proposed. Change in aerosol optical properties may also affect the atmospheric stability (Tian et al., 2018b) and thus impact aerosol and O$_3$ concentrations, which requires further studies.

CRediT authorship contribution statement

Yi Chang: Formal analysis, Data curation, Methodology, Software, Visualization, Writing – original draft, Writing – review & editing. Tao Du: Data curation, Formal analysis, Writing – review & editing. Xin Song: Data curation, Formal analysis, Writing – review & editing. Wenfang Wang: Formal analysis, Visualization, Writing – review & editing. Pengfei Tian: Conceptualization, Formal analysis, Funding acquisition, Project administration, Validation, Writing – original draft, Writing – review & editing. Xu Guan: Data curation, Software, Visualization. Naiyue Zhang: Writing – review & editing. Min Wang: Writing – review & editing. Yumin Guo: Writing – review & editing. Jinsen Shi: Data curation. Lei Zhang: Resources.

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.

Data availability

LACMS data is available online: http://climate.lzu.edu.cn/English/Data_Sharing/LACMS_data.htm.

Acknowledgement and data availability

This research was supported by the National Natural Science Foundation of China (42175093 and 41905017). The authors thank the staffs from the Lanzhou Atmospheric Components Monitoring Superstation (LACMS) and the Ministry of Ecology and Environment (MEE) network for the data used in this study. The MEE network data is available at http://www.pm25.in/ and LACMS data is available online at the Semi-Arid Climate and Environment Observatory of Lanzhou University (SACOL), http://climate.lzu.edu.cn/English/Data_Sharing/LACMS_data.htm.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2022.119270.

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