

Response of Surface Ozone to Atmospheric Aerosol Absorption is More Sensitive than to Scattering

Pengfei Tian¹, Xin Song¹, Tao Du¹, Zeren Yu¹, Jinsen Shi², Yi Chang³, and Lei Zhang¹

¹Lanzhou University

²College of Atmospheric Sciences

³Gansu Province Environmental Monitoring Center

December 14, 2022

Abstract

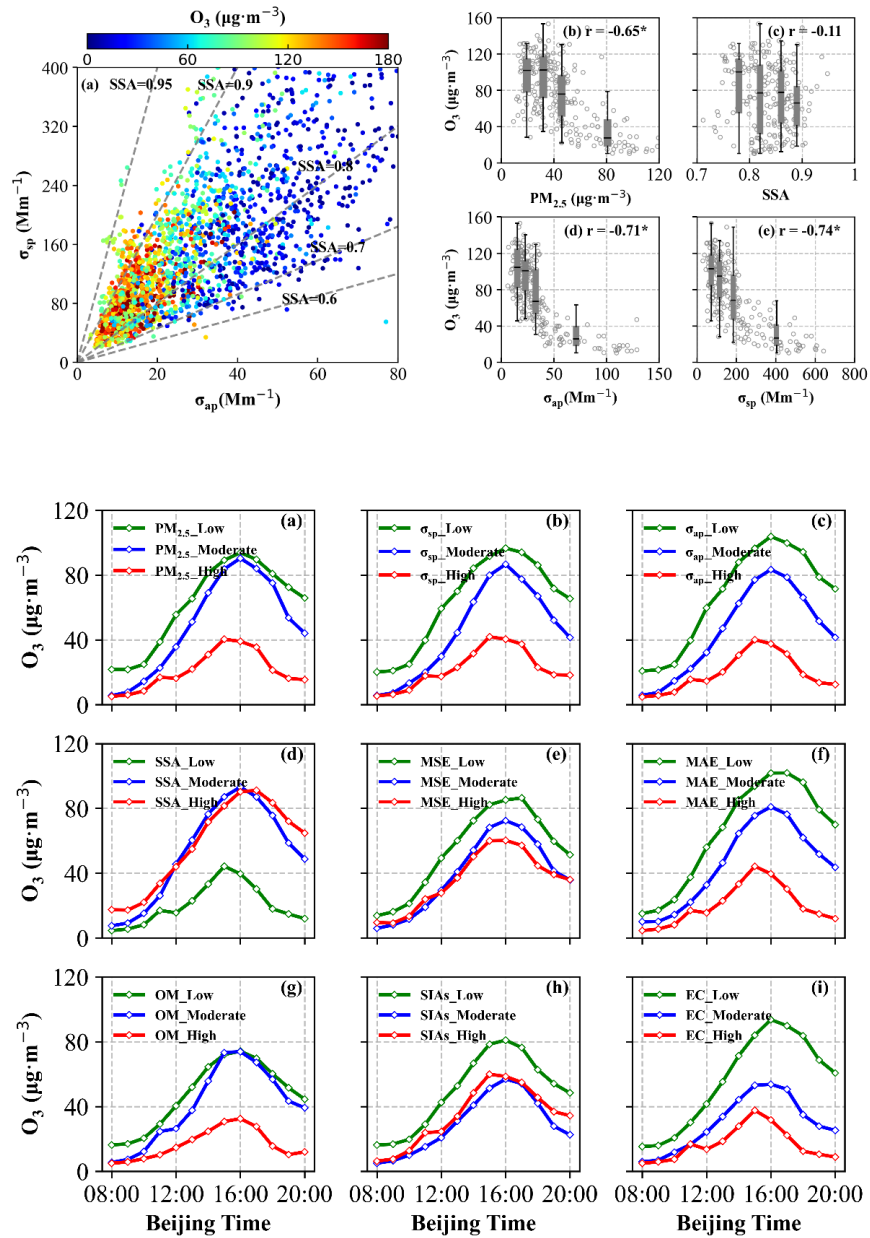
Interactions between atmospheric aerosols and ozone have a significant impact on air pollution and the climate. However, the relative importance of the response of surface ozone to aerosol scattering and absorption has been poorly quantified from in situ observations. Results derived from a one-year in situ observational study conducted in a semi-arid region showed that the response of ozone to aerosol absorption was more sensitive than to scattering. Specifically, the change in surface ozone from low to high absorption coefficients was approximately five times that from low to high scattering coefficients. The mass scattering and absorption efficiencies, rather than the single-scattering albedo, which are commonly applied in numerical simulations, were able to clearly distinguish surface ozone. The positive correlation between aerosol and ozone in summer showed the promotion of secondary aerosols by ozone. This study provides robust observational evidence of the response of surface ozone to aerosol scattering and absorption.

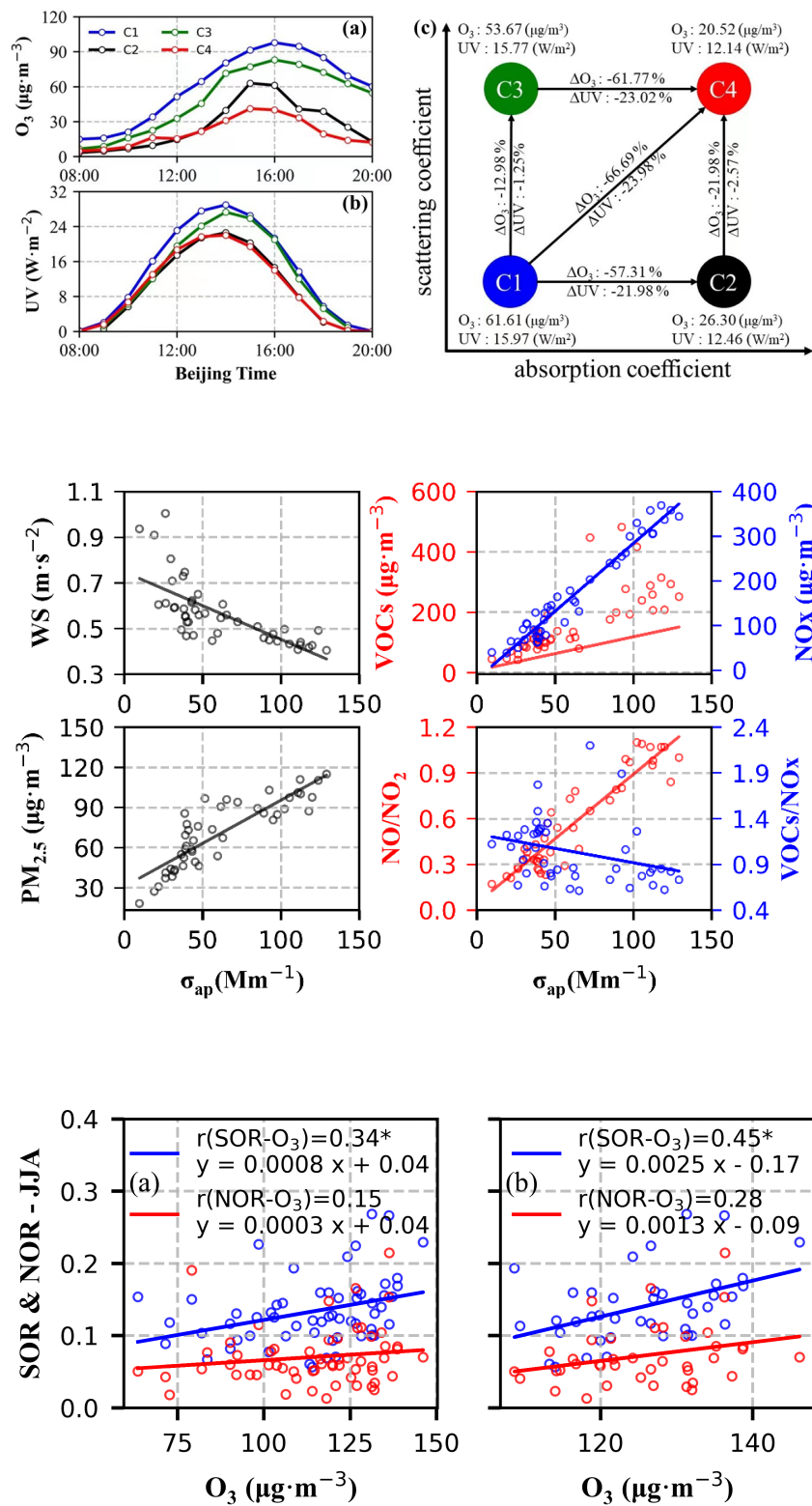
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4 **Xin Song¹, Pengfei Tian^{1*}, Tao Du², Zeren Yu¹, Jinsen Shi^{1,3}, Yi Chang⁴, Lei**
5 **Zhang^{1,3}**

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7 ¹Key Laboratory for Semi-Arid Climate Change of the Ministry of Education, College
8 of Atmospheric Sciences, Lanzhou University, Lanzhou 730000, China

9 ²Institute of Plateau Meteorology, China Meteorological Administration, Chengdu
10 610000, China

11 ³Collaborative Innovation Center for Western Ecological Safety, Lanzhou University,
12 Lanzhou 730000, China

13 ⁴Gansu Province Environmental Monitoring Center, Lanzhou 730020, China

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15 Correspondence to: P. Tian (tianpf@lzu.edu.cn)

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Abstract: Interactions between atmospheric aerosols and ozone have a significant impact on air pollution and the climate. However, the relative importance of the response of surface ozone to aerosol scattering and absorption has been poorly quantified from *in situ* observations. Results derived from a one-year *in situ* observational study conducted in a semi-arid region showed that the response of ozone to aerosol absorption was more sensitive than to scattering. Specifically, the change in surface ozone from low to high absorption coefficients was approximately five times that from low to high scattering coefficients. The mass scattering and absorption efficiencies, rather than the single-scattering albedo, which are commonly applied in numerical simulations, were able to clearly distinguish surface ozone. The positive correlation between aerosol and ozone in summer showed the promotion of secondary aerosols by ozone. This study provides robust observational evidence of the response of surface ozone to aerosol scattering and absorption.

Plain Language Summary: The concentration of surface ozone has gradually increased over the last few decades. The increasing trend in ozone was partially due to a reduction in atmospheric aerosols. The response of ozone to aerosol scattering and absorption remains unclear. The lack of robust observational evidence was the main reason for this finding. Here, we used one-year observational data to study the interactions between aerosols and surface ozone. The change in ozone concentration from low to high aerosol absorption was approximately five times that from low to high aerosol scattering. This robust evidence indicates that the response of surface ozone to aerosol absorption is more sensitive than that to scattering. The commonly applied parameter of single-scattering albedo in numerical simulations cannot distinguish the surface ozone. The positive correlation between aerosol loading and ozone in summer was caused by the oxidation effects of ozone on secondary aerosols.

1. Introduction

Tropospheric ozone is a highly reactive gas and strong oxidant that has important implications for human health, atmospheric oxidation, and global climate change (Steinfeld et al., 1998; Ainsworth et al., 2012; Lu et al., 2020). Surface ozone has been increasing in the last few decades and in recent years has increased more rapidly, resulting in serious ozone pollution regionally and globally (Velasco and Retama, 2017; Gaudel et al., 2020; Xue et al., 2020; Wang et al., 2022). Studies have shown that the recent increase in surface ozone concentration is related to the reduction in particulate pollutants, that is, atmospheric aerosols (Anger et al., 2016; Li et al., 2019). Aerosols have a substantial influence on surface ozone via atmospheric chemical and physical processes (Dickerson et al., 1997; Martin et al., 2003; Gao et al., 2018). As a strong oxidant, ozone modulates atmospheric oxidation, thus affecting the generation of secondary aerosols (Jenkin et al., 2000; Wang et al., 2016; Wu et al., 2020). Therefore, studying the interactions between aerosols and ozone is crucial for collaborative management of pollution characterized by particulate matter and ozone.

There has been an increasing interest in studying the influence of atmospheric aerosols on tropospheric ozone and several research advances have been made over the last few decades (Meng et al., 1997; Pere et al., 2015; Zhu et al., 2021; Wang et al., 2022). Many studies have revealed that aerosols affect ozone by changing the photolysis frequency (Jacobson et al., 1998; Benas et al., 2013; Wang et al., 2019), some studies have quantified the impacts of absorbing aerosols on surface ozone (Li et al., 2005; Mok et al., 2016; An et al., 2021), and recent studies have compared the

differences in the impacts of scattering and absorbing aerosols on ozone (Gao et al., 2022; Shi et al., 2022). Aerosols also absorb oxidative radicals, inhibit the formation of ozone in the troposphere (Jacob et al., 2000; Lou et al., 2014; Li et al., 2018), and influence ozone by altering atmospheric dynamics, that is, affect the evolution of the atmospheric boundary layer and the vertical exchange (Xing et al., 2017; Gao et al., 2020; Yang et al., 2021). Studies have shown that high concentrations of ozone promote the generation of secondary aerosols by enhancing atmospheric oxidation, thereby increasing the risk of haze events (Fu et al., 2020; Huang et al., 2021).

The interactions between aerosols and ozone have mainly been studied using model simulations (Tie et al., 2005; Pere et al., 2015; Ma et al., 2021). However, model simulations may suffer from uncertainties owing to deficiencies in emission inventories, insufficient consideration of physical and chemical processes, and inaccurate meteorological field simulations (Feng et al., 2016; Wang et al., 2019). Observations are derived in real-time from the atmosphere; thus, a reasonable analysis of the *in situ* observation data may yield results that could not have been drawn in model simulations. Several studies have analyzed the relationships between *in situ* measured PM_{2.5} and ozone (e.g., Wang et al., 2020; Chu et al., 2020; Dai et al., 2021). Some studies have attempted to investigate the influence of aerosol optical properties on ozone using observational data (Gharibzadehet al., 2021; Shao et al., 2022). A recent study compared the influence of scattering and absorbing aerosols on ozone (Shi et al., 2022). However, scattering or absorbing aerosols can also scatter and absorb light. So, the study of aerosol effects on ozone from the perspective of scattering and absorbing

aerosols does not fully distinguish between the effects of aerosol scattering and absorption on ozone. The relative importance of aerosol scattering and absorption by ozone has not yet been quantified. Thus, further studies need to address these gaps especially using *in situ* observations, to quantify the relative importance of aerosol scattering and absorption.

Several studies have used model simulations to discuss the influence of aerosol optical properties on ozone by changing the values of single-scattering albedo (SSA) and aerosol optical depth (AOD) (Castro et al., 2001; Qu et al., 2018). For example, decreasing the SSA under a constant AOD decreases the scattering and increases the absorption. However, scattering and absorption always exhibit the same increasing/decreasing trends in the atmosphere. Therefore, whether SSA is a good parameter for studying the effect of aerosol optical properties on ozone using observational data requires further investigation.

Furthermore, studies have shown that enhancement of atmospheric oxidation caused by ozone promotes the production of secondary aerosols (Khoder, 2002; Ding et al., 2013; Chen et al., 2019). According to previous studies, sulfate, nitrate, ammonium, and organic matter were the major chemical species of PM_{2.5} in Lanzhou (Wang et al., 2021). Therefore, strong oxidation might lead to the co-occurrence of ozone and PM_{2.5}. The interactions between aerosols and ozone can be understood more comprehensively using observation data.

To address these issues, a one-year field campaign on aerosol optical properties and ozone was conducted in a semi-arid region of Lanzhou, Northwest China. The *in*

situ observation data were then used to study aerosol effects on ozone from the perspective of aerosol scattering and absorption, identify representative parameters to clearly distinguish ozone using *in situ* data, and discuss the promoting effect of ozone on secondary aerosols in summer. *In-situ* observation data were also used to illustrate the influence of aerosols on ozone by altering atmospheric dynamics.

2. Data and methods

Located in northwest China, Lanzhou is a transitional region between the Qinghai and Tibet Plateau and the Loess Plateau. It has exhibits typical semi-arid climate, and most areas are hills and basins covered by loess. The Lanzhou landform is long, narrow, and has the characteristics of a basin city. However, the special terrain conditions in the valley basin area are not conducive to pollutant dilution and diffusion. Lanzhou is dominated by calm winds throughout the year, and the temperature inversion phenomenon is considerable; thus, air pollution is usually more severe. With a population of 4.38 million in 2021, Lanzhou used to be one of the world's most polluted cities due to heavy industrial pollution such as petrochemical and smelting, as well as domestic and traffic emissions (Tang et al., 1989; Wang et al., 2009).

The data in this study were obtained from the Lanzhou Atmospheric Components Monitoring Superstation (LACMS; 36.05°N, 103.87°E) in Lanzhou. LACMS is a comprehensive atmospheric environment monitoring station jointly established by the Department of Ecology and Environment of Gansu Province and Lanzhou University for studying fine particulate matter and ozone pollution mechanisms. Built at Lanzhou University, the monitoring station uses 27 advanced environmental monitoring instruments, comprising an ambient air quality monitoring system, photochemical

pollution observation system, ground-based remote sensing observation system, and remote sensing data services. The LACMS is located on the roof of a 10-story building on the campus of Lanzhou University in Chengguan District, Lanzhou City (approximately 20 m above the ground). It is far from the main road and industrial sources and avoids the influence of local meteorological and local pollution sources. The observation results are representative of the cities. Detailed descriptions of the instruments at the LACMS can be found in other studies (Du et al., 2020; Chang et al., 2022).

Sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) are criteria for measuring the degree of secondary conversion of SO₂ and NO₂ to sulfate and nitrate, respectively. SOR and NOR were calculated using the following formulas:

$$\text{SOR} = [\text{SO}_4^{2-}] / ([\text{SO}_4^{2-}] + [\text{SO}_2]) \quad (1)$$

$$\text{NOR} = [\text{NO}_3^-] / ([\text{NO}_3^-] + [\text{NO}_2]) \quad (2)$$

where [SO₄²⁻] and [NO₃⁻] are the molar concentrations of SO₄²⁻ and NO₃⁻ in PM_{2.5} (μmol·m⁻³) respectively, and [SO₂] and [NO₂] are the molar concentrations of SO₂ and NO₂ in gas phase (μmol·m⁻³) respectively. SOR and NOR > 0.1 indicate the secondary formation of sulfate and nitrate (Ohta and Okita, 1990).

Organic matter (OM) was determined from OC multiplied by a mass conversion factor of 1.6 (Turpin and Lim, 2001; Xing et al., 2013; Chow et al., 2015), the following formula:

$$\text{OM} = 1.6 \times \text{OC} \quad (3)$$

The SSA was calculated using aerosol scattering coefficients (σ_{sp}) and absorption coefficients (σ_{ap}) at 520 nm. The mass scattering efficiency (MSE) and mass absorption efficiency (MAE) were calculated using the ratios of σ_{ap} and σ_{sp} to PM_{2.5}. The calculation formulae are as follows:

$$SSA = \sigma_{sp}/\sigma_{ext} = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap}) \quad (4)$$

$$MAE = \sigma_{ap}/PM_{2.5} \quad (5)$$

$$MSE = \sigma_{sp}/PM_{2.5} \quad (6)$$

The observation period in this study was from December 1, 2019, to November 31, 2020, and the LACMS site data were missing from February 11, 2020, to March 2, 2020. The data during daytime (08:00–20:00) under sunny days were selected for this study. The seasonal solar radiation values were divided into three equal parts, and sunny days were selected according to the seasonal solar radiation values. If the solar radiation of the day was greater than 1/3 of the seasonal solar radiation value, the day was considered sunny.

3. Results and discussion

3.1 Relationships between aerosol optical parameters and surface ozone

Lanzhou is a semi-arid basin with relatively high surface ozone and particulate pollution (Du et al., 2020). The average ozone and $PM_{2.5}$ concentrations during the field campaign were 81.1 and $44.9 \mu g \cdot m^{-3}$, respectively. Seasonally, winter was the season with the heaviest particulate pollution, while summer was the most polluted season for surface ozone (Fig. S1). Specifically, the seasonal average ozone ($PM_{2.5}$) was 43.4 (68.9), 98.8 (40.6), 114.6 (24.7), and 53.3 (50.2) $\mu g \cdot m^{-3}$ in winter, spring, summer, and autumn, respectively. Recent studies have revealed that atmospheric aerosols in Lanzhou efficiently absorb solar radiation (Guan et al., 2021) and have a potential impact on surface ozone (Chang et al., 2022).

The relationships between aerosol optical parameters and surface ozone are

illustrated in Fig. 1. The most stark feature was that high values of ozone corresponded to low scattering and absorption, and low ozone concentrations fell in areas with high scattering and absorption (Fig. 1a). This reflected the effect of aerosol loading, which was also demonstrated by the relationship between ozone and $PM_{2.5}$ (Fig. 1b). Significant negative correlations were found between ozone and $PM_{2.5}$, ozone and scattering coefficient, and ozone and absorption coefficient ($p < 0.05$, Fig. 1d and 1e). Despite a slightly higher (Pearson) correlation coefficient for ozone and the scattering coefficient, the relationship between ozone and the absorption coefficient seemed to be more robust, as seen from the distribution of the scatter plots in Fig. 1d and 1e. SSA is also a parameter used to represent aerosol effects on ozone (Liu et al., 2019; Ma et al., 2021; Shao et al., 2022), which is clearly reflected in Fig. 1a when the scattering and absorption coefficients are relatively high.

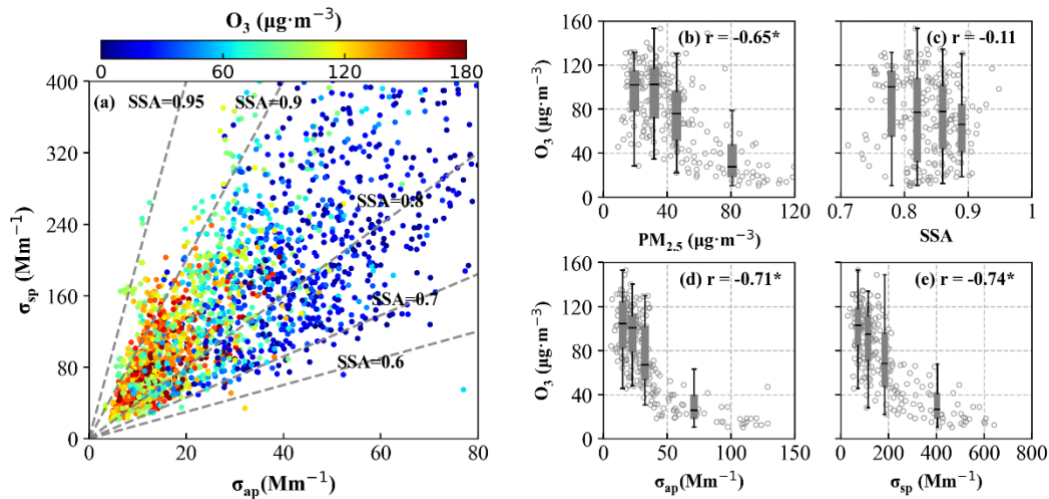


Figure 1. (a) Hourly surface ozone as a function of aerosol scattering and absorption coefficients; (b, c, d, e) scatter plots of daily ozone and aerosol parameters of $PM_{2.5}$, single-scattering albedo (SSA), and absorption and scattering coefficients (* indicates that the correlation between the two passed the significance test of 0.05).

A negative correlation was found for ozone and scattering, as well as for ozone and absorption, for the total samples and for each season. However, distinct seasonal variations were found in the relationships between ozone and aerosol parameters of $\text{PM}_{2.5}$ and SSA (Fig. S2). Ozone and $\text{PM}_{2.5}$ exhibited significant negative correlations in winter and autumn, weak negative correlations in spring, and weak positive correlations in summer. The negative correlation between ozone and $\text{PM}_{2.5}$ indicated the dominant influence of aerosols on ozone via photolysis effects, whereas a positive correlation mainly showed the promotion of secondary aerosols by ozone (Jia et al., 2017). Ozone and SSA exhibited a strong positive correlation in winter but showed little correlation in the other seasons, which also suggests the significant influence of aerosols on ozone in winter.

3.2 Responses of surface ozone to aerosol scattering and absorption

The responses of surface ozone to aerosol parameters were first investigated by studying the daytime evolution of ozone and UV radiation under low, moderate, and high levels of aerosol parameters of $\text{PM}_{2.5}$, scattering and absorption coefficients, and SSA (Fig. S3 and S4). The responses of surface ozone to aerosol scattering and absorption will be analyzed mainly using data from winter when aerosol loading is the highest. The responses of ozone to aerosol loading were evident in the evolution of daytime ozone under different levels of $\text{PM}_{2.5}$, as well as scattering and absorption coefficients (Fig. 2). The differences between ozone under low and high absorption coefficients were larger than those under low and high scattering coefficients, which means that ozone is more sensitive to aerosol absorption than to scattering. This was

more evident in the daytime evolution of ozone under different levels of MAE and MSE.

The difference between ozone under low and high MAE conditions at 16:00 was approximately three times that under low and high MSE conditions at 16:00.

Furthermore, little difference was found between ozone under moderate and high SSA levels, but ozone under low SSA levels was much lower than that under the other two levels.

Thus, it seems that SSA might not be the best parameter for studying the responses of surface ozone to atmospheric aerosols, whereas MSE and MAE seem to be good characterizing parameters.

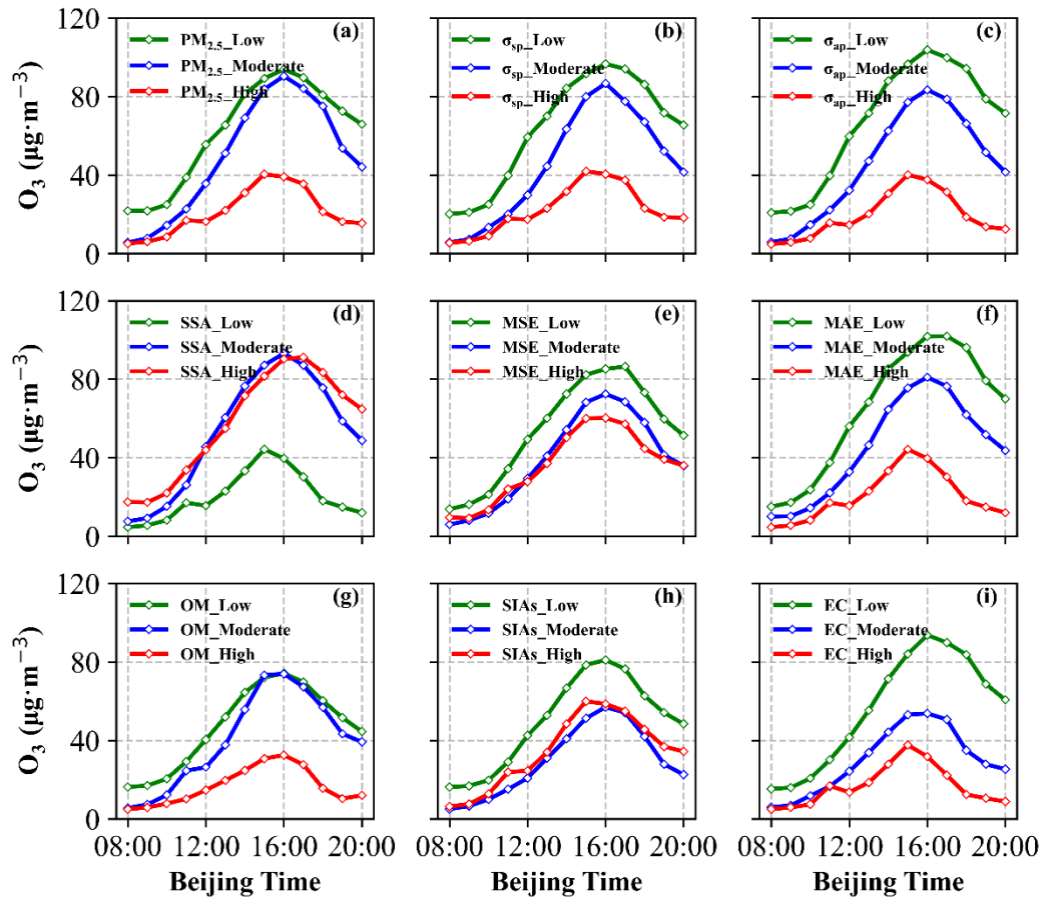


Figure 2. Daytime evolution of ozone under low, moderate, and high levels of aerosol parameters of (a) $PM_{2.5}$; (b) scattering coefficient; (c) absorption coefficient; (d) single-scattering albedo (SSA); (e) mass scattering efficiency (MSE); (f) mass absorption

efficiency (MAE); main chemical species in PM_{2.5} of (g) organic matter (OM); (h) secondary inorganic aerosols (SIAs); and (i) elemental carbon (EC) in winter.

The changes in secondary inorganic aerosols (SIAs) and OM were consistent with those in PM_{2.5}, and they all reduced the ozone concentration by affecting the photolysis frequency. Therefore, SIAs, OM, and PM_{2.5} were negatively correlated with ozone. There was a significant negative correlation between EC and ozone in winter (Fig. S5). The reason might be that the solar radiation intensity is weak in winter, and EC can make the atmosphere more stable so that NO can accumulate. Weak photochemical reactions and enhanced ozone consumption by NO reduced the surface ozone; thus, the negative correlation between the EC and ozone was more pronounced.

The responses of surface ozone to aerosol scattering and absorption were further investigated by comparing the daytime evolutions of ozone with the scattering and absorption coefficients above and below their averages (Fig. 3). For example, C2 had the same scattering coefficient as C1 but a higher absorption coefficient than C1. Therefore, the ozone difference between C2 and C1 is caused by a change in aerosol absorption with low scattering. Similarly, the ozone differences between C4 and C3, C3 and C1, and C4 and C2 were caused by a change in aerosol absorption with high scattering, scattering with low absorption, and scattering with high absorption, respectively. The ozone difference between C4 and C1 was attributed to changes in both scattering and absorption. The ozone differences between C4 and C3 (relative difference of -61.77%) and C2 and C1 (-57.31%) were significantly higher than those between C4 and C2 (-21.98%) and C3 and C1 (-12.98%), which clearly indicated that

the response of ozone to aerosol absorption was more sensitive than to aerosol scattering. The difference in ozone concentrations between C1 and C2 (C3 and C4) was 4.45 (5.78) times of that between C1 and C3 (C2 and C4), indicating that the variation in ozone due to aerosol absorption was \sim five times that due to aerosol scattering.

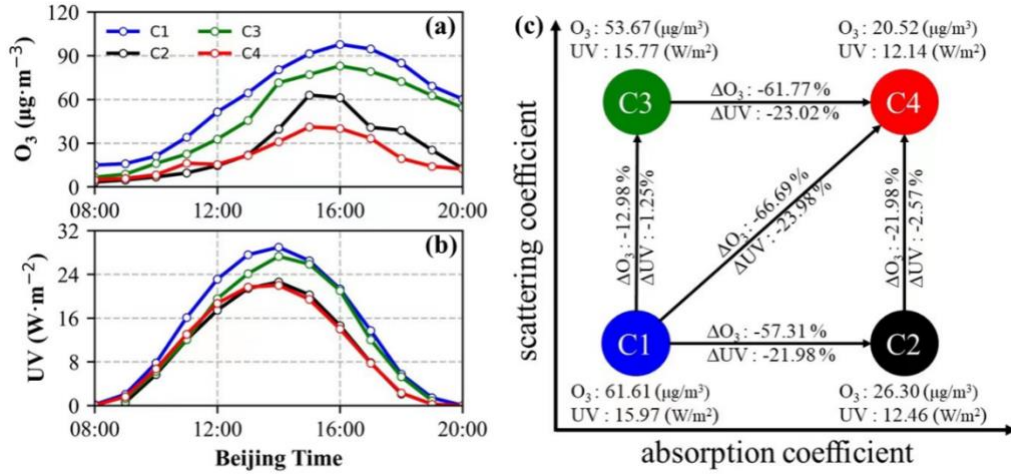


Figure 3. Daytime evolutions of ozone and UV radiation with scattering and absorption coefficients above and below their averages and their comparisons (c). C1, C2, C3, and C4 represent the conditions of $\sigma_{\text{ap}} < \bar{\sigma}_{\text{ap}}$ and $\sigma_{\text{sp}} < \bar{\sigma}_{\text{sp}}$, $\sigma_{\text{ap}} > \bar{\sigma}_{\text{ap}}$ and $\sigma_{\text{sp}} < \bar{\sigma}_{\text{sp}}$, $\sigma_{\text{ap}} < \bar{\sigma}_{\text{ap}}$ and $\sigma_{\text{sp}} > \bar{\sigma}_{\text{sp}}$, and $\sigma_{\text{ap}} > \bar{\sigma}_{\text{ap}}$ and $\sigma_{\text{sp}} > \bar{\sigma}_{\text{sp}}$, respectively.

In addition to the photolysis impacts of aerosols on ozone, aerosols also influence surface ozone concentration via atmospheric dynamic effects: (1) changing the concentrations and configurations of precursors, and (2) affecting the vertical exchange of ozone (Hansen et al., 1997; Gao et al., 2018; Yang et al., 2021). Previous studies have analyzed atmospheric dynamic effects using numerical models such as WRF-Chem (Gao et al., 2020; Qu et al., 2020; An et al., 2021). With the aim of addressing this issue based on observation data, wintertime data were also used to show the dynamic atmospheric effects of aerosols on ozone.

The decreasing trend of wind speed with increasing absorption coefficient indicates weakened boundary layer evolution by aerosol absorption (Fig. 4). On one hand, the weakened boundary layer inhibited ozone exchange in the vertical distribution and resulted in lower surface ozone. However, the weakened boundary evolution led to the accumulation of ozone precursors of NO_x and VOCs. However, NO_x increased faster than VOCs with increasing absorption, leading to a decreasing trend in the VOCs-to-NO_x ratio. Ozone responds nonlinearly to the precursors, and Lanzhou is in the VOCs-limited region in the Ekma diagram (Li et al., 2022). Thus, the decreasing trend in the ratio of VOCs to NO_x led to lower surface ozone. In addition, the ratio of NO to NO₂ can reflect the relative importance of the consumption and generation of ozone because NO undergoes a titration reaction with ozone, while NO₂ is the precursor of ozone (Han et al., 2011; De Souza et al., 2017). Thus, the rapid increase in NO to NO₂ ratio indicates an increase in ozone consumption with increasing aerosol absorption.

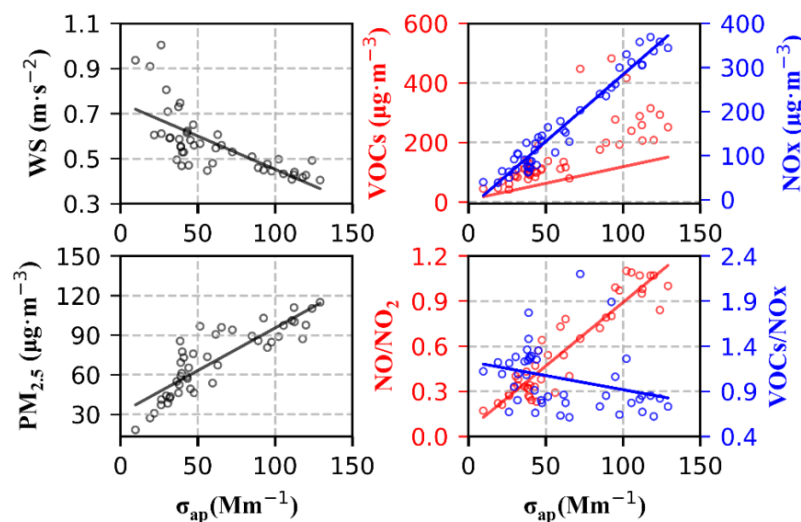


Figure 4. Wind speed, PM_{2.5}, and ozone precursors as a function of absorption coefficient in winter.

3.3 Impacts of ozone on secondary aerosol formation

The impact of ozone on secondary aerosol formation was revealed based on summertime observation data (Fig. 5). Both NOR and SOR exhibited positive correlations with ozone for the total summer samples, indicating the promotion of sulfate and nitrate by ozone (Fig. S6, Fig. S7). The correlations were stronger for samples with ozone concentrations over $109 \mu\text{g}\cdot\text{m}^{-3}$ (Fig. 5), which further illustrates the promotion of sulfate and nitrate by ozone. This was probably the reason for the positive correlation between summertime ozone and $\text{PM}_{2.5}$. The correlation between ozone and SOR was significant and stronger than that between ozone and NOR, suggesting that the impact of ozone on sulfate was stronger than that on nitrate. The weaker effect of ozone on nitrate may be due to the evaporation of ammonium nitrate with increasing temperature (Wen et al., 2015).

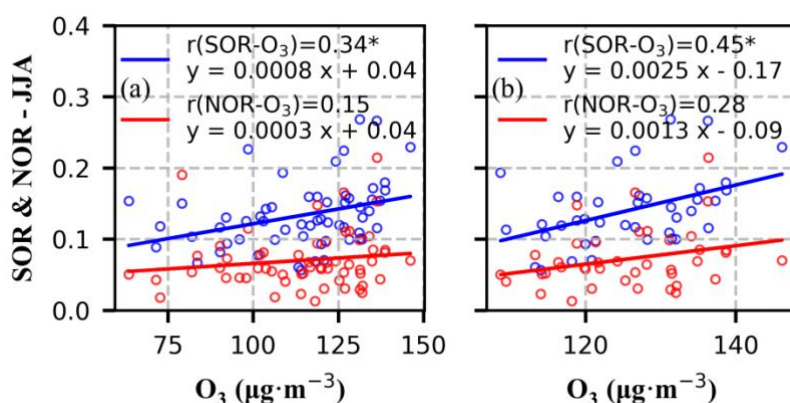


Figure 5. Sulfur oxidation ratio (SOR) and nitrogen oxidation ratio (NOR) as a function of ozone in summer. (a) represented the total summer sample; (b) represented the samples with ozone concentration over $109 \mu\text{g}\cdot\text{m}^{-3}$ (* indicates that the correlation between the two passed the significance test of 0.05).

4. Conclusions

In this study, the relationships between aerosol optical parameters and surface ozone, responses of surface ozone to aerosol scattering and absorption, and effects of ozone on secondary aerosols were investigated using long-term observational data. We found that both aerosol scattering and absorption can lead to a decrease in surface ozone; however, the response of ozone to aerosol absorption is more sensitive than aerosol scattering. The change in surface ozone from low to high absorption coefficients was approximately five times that of ozone from low to high scattering coefficients. The correlation between SSA and ozone and the difference in ozone concentration under different SSA levels indicate that SSA might not be the best parameter for studying the responses of surface ozone to atmospheric aerosols, whereas MAE and MSE seem to be good characterizing parameters.

Observation data were simultaneously used to verify that aerosol absorption can change the surface ozone concentration by influencing atmospheric dynamics. Aerosol absorption weakened the evolution of the atmospheric boundary layer and resulted in lower surface ozone by inhibiting ozone exchange in the vertical direction, accumulating ozone consumers, such as NO and PM_{2.5}, and decreasing the ratio of VOCs to NO_x. Furthermore, the impact of ozone on secondary aerosol formation was revealed based on summertime observational data. The positive correlation between aerosol loading and surface ozone in summer showed the oxidation effects of ozone on secondary aerosols when aerosol loading was light.

Acknowledgments: This research was supported by the National Natural Science

Foundation of China (42175093 and 41905017). The authors thank the staff of the Lanzhou Atmospheric Components Monitoring Superstation (LACMS) for the data used in this study.

Open Research

The LACMS data are available online at <https://www.zenodo.org/record/7425931#.Y5aye1FBxPa>. Figures were made with Matplotlib version 3.2.1, available under the Matplotlib license at [Matplotlib — Visualization with Python](#).

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Figure 1.

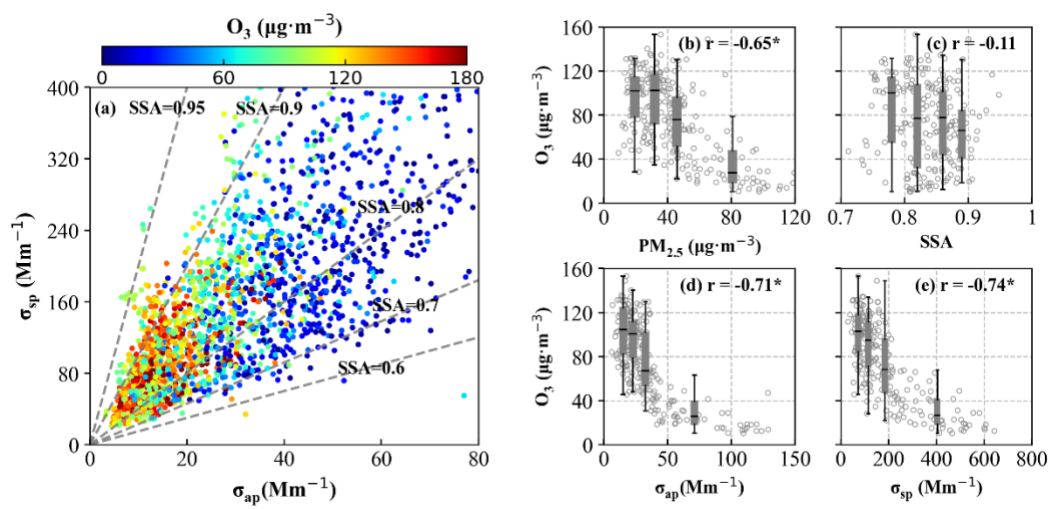


Figure 2.

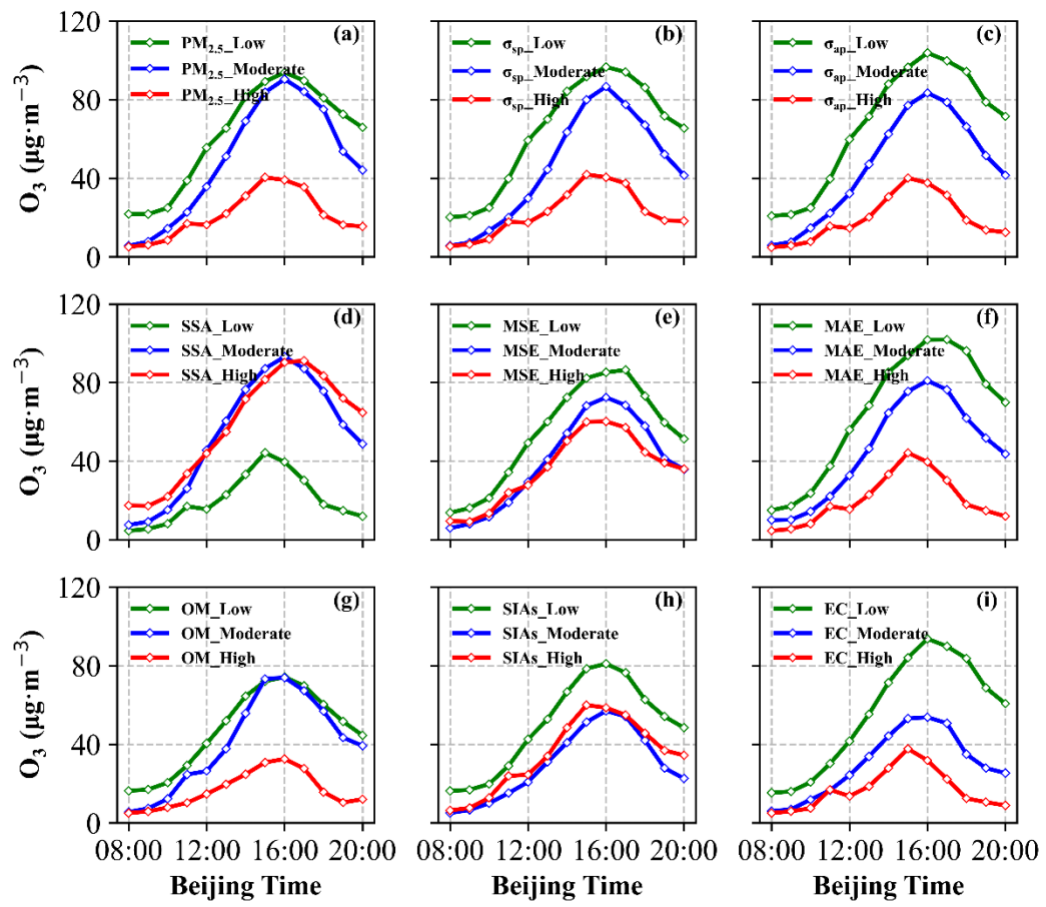


Figure 3.

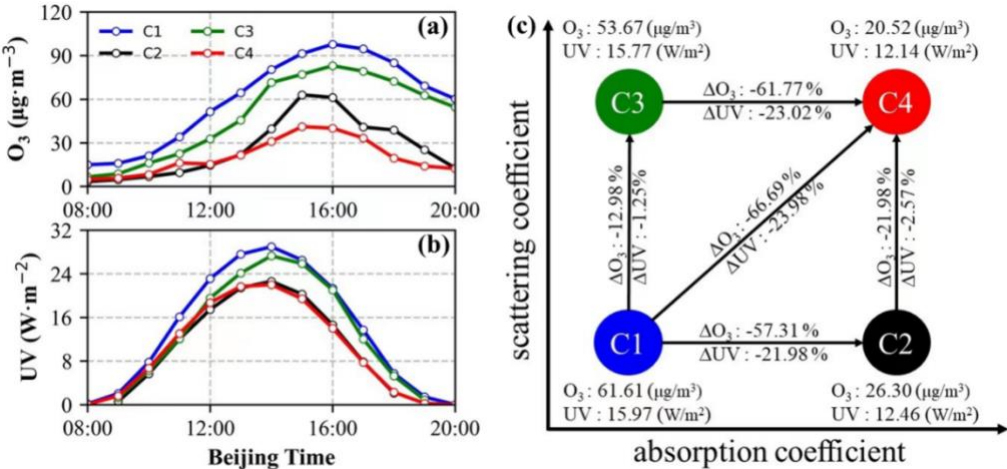
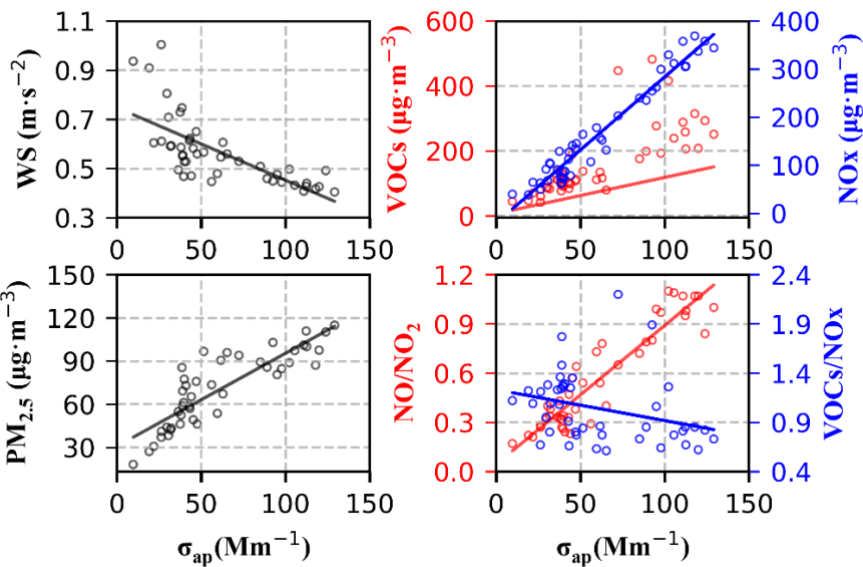


Figure 4.



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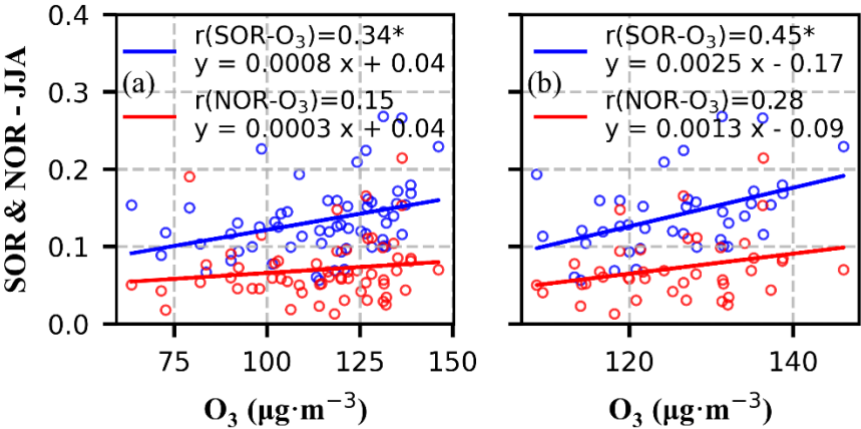
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661 **Figure 5.**



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