Cause Analysis of PM_{2.5} pollution during the COVID-19 lockdown in Nanning, China

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Abstract

To analyze the cause of atmospheric PM_{2.5} pollution occurred during the COVID-19 lockdown in Nanning of Guangxi, China, Single Particulate Aerosol Mass Spectrometer, Aethalometer, Particulate Lidar, coupled with the monitoring of near-surface gaseous pollutants, meteorological conditions, remote fire spots sensing by satellite and Backward Trajectory Models were conducted during 18–24, Feb 2020. Three haze stages of pre-pollution period (PPP), pollution accumulation period (PAP) and pollution dissipation period (PDP) were identified. The dominant source of PM_{2.5} in PPP was biomass burning (BB) (40.4%), followed by secondary inorganics (28.1%) and motor vehicle exhaust (11.7%). The PAP was characterized by a large abundance of secondary inorganics, which contributed for 56.1% of the total PM_{2.5} concentration, followed by BB (17.4%). The absorption Ångström exponent (2.2) in PPP was higher than those of the other two periods. The analysis of fire spots monitored by remote satellite sensing indicated that open BB in regions around Nanning city could be one of the main factors matters. The planetary boundary layer-relative humidity-secondary particles matter-particulate matter positive feedback mechanism was employed to elucidate the atmospheric process in this study. This study highlights the importance of understanding the role of BB and meteorology in air pollution formation to call for policy for emission control strategies.

Introduction

Air pollution, particularly fine particulate matter (PM_{2.5}) have caused significant economic loss and adverse public health effects in countries such as China^{1–3}. In regards of severe air pollution issues and to protect public health, the State Council of China promulgated the most stringent Air Pollution Prevention and Control Action Plan (Action Plan) in 2013⁴, in which PM_{2.5} concentration reductions of 25%, 20%, and 15% in 2017 compared to the level in 2013 were mandated in Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD) and Pearl River Delta (PRD) areas, respectively. Tremendous efforts such as setting more restrict industrial and vehicle emission standards, closing down heavy pollution factories, upgrading industrial facilities for mitigations of various pollutants emissions. Evaluation of the effectiveness of these measures can provide crucial information for developing clean air strategies in China as well as in other developing countries facing similarly severe pollution issues⁵. Several studies have showed that severe air pollution events had been successfully mitigated by controlling anthropogenic emissions in China^{6–9}. For instance, a statistical model was developed and indicated that the implementation of stringent emission reduction measures alone could effectively lower PM_{2.5} levels by 20–24 µg.m⁻³ (27–33%) on average during the 2008 Beijing Olympic Games⁶. Meanwhile, some other studies also proved that meteorological factors play critical roles in the formation of atmospheric pollution incidents and should be taken into account to determine the actual influence of controlling procedures on pollution reduction^{9–12}.

Due to the coronavirus 2019 (COVID-19) pandemic occurring at the end of 2019¹³, the epidemic centre Wuhan city had announced the lockdown on January 23, 2020. All industrial production, transport system

and social activities had been reduced to the minimum extent throughout the whole country. Surprisingly, from 18 to 24, February, 2020, a regional PM_{2.5} pollution incident took place in Guangxi, China, slight atmospheric pollution was observed in the provincial capital Nanning city on 20 February. As during COVID-19 lockdown period, regular air pollutant emissions from industrial and domestic activities had decreased significantly due to reduced road motor vehicles volumes and postponed infrastructure constructions¹⁴⁻¹⁶, such a pollution incident was out of expectation and caused social concern. Li et al.¹⁴ indicated that even during the lockdown, with primary emissions reduction of 15%–61%, the daily average PM_{2.5} concentrations in YRD still ranged between 15 and 79 µg.m⁻³, which showed that background and residual pollutions were still high. Similarly, in spite of the extreme reductions in primary emissions in Guangxi during the COVID-19 lockdown, it could not fully tackle the current air pollution. Thus, it is meaningful to study the precise cause factors of atmospheric pollution formation.

In this study, the atmospheric pollution incident in Nanning was selected as a case investigation for pollution source analysis with multi-sets equipment installed in the atmospheric observatory station. Single Particulate Aerosol Mass Spectrometer, Aethalometer, particulate Lidar, surface meteorological and environmental data, satellite remote sensing data and modeled HYSPLIT4 trajectory were used to analyze the cause of PM_{2.5} pollution in Nanning during the COVID-19 control period, when a regional haze pollution episode was captured from 28th January to 3rd February in Guangxi.

Results And Discussion

Overall regional pollution situation of in Guangxi. From 18 to 24 February, 2020, a regional air pollution event with a cumulative pollution period of 1.1 days occurred in Guangxi, and the key pollutant was PM_{2.5} (see Supplementary Fig. S1). During this period, this pollution event first started on 19 February, and slight pollution (AQI >100, PM_{2.5} > 75µg.m⁻³) was identified in four cities, namely Laibin, Chongzuo, Liuzhou and Hechi; among of them, Chongzuo represented the highest AQI value (AQI=123). However, the most severe pollution was observed in Hechi city on 20th Feb (AQI> 200, PM_{2.5}>150 µg.m⁻³), then the levels of pollutants began to decline (with slightly rise up on 23rd of February) and finally ended on February 24.

To evaluate the regional overall pollution status of Guangxi during this period, the comparison of average concentrations of atmospheric PM_{2.5} in Guangxi during 19-23 Feb 2020 with those of all other provinces was illustrated in Supplementary Fig. S2. It showed that the average PM_{2.5} concentration was 61 µg.m⁻³, which was the third highest for nationwide in these days (the average PM_{2.5} concentrations in Hechi city even reached up to 79 µg.m⁻³). In contrast, the average atmospheric PM_{2.5} concentrations in surrounding provinces, e.g., Guangdong, Yunnan, Guizhou and Hunan were only 31, 35, 44 and 46 µg.m⁻³ respectively, which were significantly lower than that of Guangxi. This indicated that the pollutants could potentially originate from local sources rather than the transmissions from the neighboring provinces. Interestingly, although the influence of industrial and domestic activities had been reduced to a minimum extent within

Guangxi or neighboring provinces during the COVID-19 lockdown, the regional pollution event in Guangxi did take places. Accordingly, we carried out the cause analysis of this PM_{2.5} pollution event based on the data from different kinds of equipment installed in Scientific Research Academy of Guangxi Environmental Protection.

The data representativeness in the sampling site. In order to evaluate the representativeness of the data measured in the sampling site, the homochronous data released by the national real-time municipal air quality platform ([http:// 106. 37. 208. 233: 20035](http://106.37.208.233:20035)) of China National Environmental Monitoring Centre (CNEMC) were employed for the comparison (see Supplementary Table S1). It showed that the daily average values of PM_{2.5}, PM₁₀, SO₂, NO₂, O₃ and CO were relatively consistent within these two monitoring systems. The correlation analysis of hourly data of PM_{2.5} and CO showed in Supplementary Fig. S3. The correlation coefficient for the two pollutants were both above 0.93, which indicated an excellent representativeness of data was recorded in the station for the ambient air quality of SRAGEP in reference to CNEMC.

Overall air pollution situation in Nanning City.

The time series of mass concentration of atmospheric pollutants along with key meteorological conditions in Nanning city from 18 to 24, Feb 2020 were illustrated in Figure 1. According to the PM_{2.5} concentration variations, the whole progression of pollution could be divided into three stages, i.e. pre-pollution period (PPP), pollution accumulation period (PAP) and pollution dissipation period (PDP). The corresponding average PM_{2.5} concentrations for PPP, PAP and PDP were 32, 73, 35 µg.m⁻³, respectively, PM_{2.5} concentration of PAP was twice as high as those of the other two stages. Similarly, the average concentrations of CO in PAP (1.00 mg/m³) was also higher than those of PPP and PDP (0.57 and 0.90 mg.m⁻³, respectively). Interestingly, while PAP and PDP possessed the average relative humidity (RH) 71% and 70%, respectively, the average of RH in PPP was only 36%. The main reason could be the lower wind speed (WS) in PAP (0.71 m/s) than those of PPP (0.87 m.s⁻¹) and PDP (1.19 m.s⁻¹). Additionally, the average planetary boundary layer heights (PBLH) were 1253 m, 787 m and 987 m in PPP, PAP and PDP, respectively. Moderate atmospheric pollution was identified in Nanning city on 20 Feb with a PBLH ranged from 297-934 m on that day. Generally, PBLH is higher during the daytime, especially at noon, however, a minimum value of PBLH (297m) was observed at 11 a.m. on 20 February, which indicated that the potential adverse meteorological conditions for the horizontal and vertical diffusions of pollutants could result in the high PM_{2.5} concentration on that day. However, during PDP, particularly on 24 Feb, the atmospheric diffusion conditions were in favor due to the low air pressure near ground and airflow currents towards the south direction, thus the calm weather maintained for several days was mitigated through the enhanced airflow conditions, as a result, the air quality began to be improved.

Single particle source analysis. By analyzing the composition of particulate matters, the sources of PM_{2.5} in municipal area could be categorized into: catering (kitchens), dust, biomass burning (BB), vehicle exhaust, coal, industrial process (non-combustion process), secondary inorganic source and others

(Figure 2). It showed that the dominant source of $PM_{2.5}$ in the stage of PPP was BB (accounts for 40.4%), followed by secondary inorganics (accounts for 28.1%) and motor vehicle exhaust (11.7%). In contrast, secondary inorganics dominated in PAP (56.1%) and PDP (30.2%), and it seemed that there was an obvious increasing of secondary transformed particulates during the PAP. The second largest contribution sources of $PM_{2.5}$ in PAP and PDP was BB, which accounts for the proportions of 17.4% and 26.1%, respectively. By analyzing the fire spots map of satellite remote sensing monitoring, large numbers of fire spots were identified in Nanning city and regions around, however, there was no significant pollution generated attributed to relatively better meteorological diffusion conditions on February 18-19. As the atmospheric horizontal and vertical diffusion conditions were worsened afterwards, pollutants began to accumulate and produce more particulates of secondary transformation, which induced more severe $PM_{2.5}$ pollution on February 20.

As mentioned above, BB had been confirmed to be the largest contribution to the pollution source during the PPP and the second largest contributor in the other two stages. Fire spots maps were constructed based on data of satellite remote sensing (Figure 3), the straw incineration fire spots were identified from February 18 to 24. The total count of straw fire spots in Guangxi was 421, and 52 of them were in Nanning city, while 70, 45 and 44 fire spots were distributed in three cities border on Nanning (Laibin, Chongzuo and Guigang, respectively). The fire spots of these four cities were the most intensified among 14 cities of Guangxi during the period of observation, which implied that the pollution in Nanning was attributed to the regional BB. As the three largest sugar production cities of Guangxi, Nanning (No.1), Laibin (No.2) and Congzuo (No.3) possess large areas of sugarcane farming that produces large amount of waste sugarcane leaves. The open-air incineration of sugarcane leaves induces significant difficulties in regional air pollution reduction particularly the urban atmospheric pollution control. It has been reported that the incineration of agricultural straw posed significant effects on air quality, public health and climate in China¹⁷⁻¹⁹. In northern China, pollution due to the straw incinerations usually take place in autumn²⁰, for instance, the open BB contributed 52.7% of atmospheric $PM_{2.5}$ in Northeast region of China during November 1-4, 2015. However, there is little research on how BB affects air quality in Southern China. Guangxi has the highest pollutants emission from BB among 31 provinces in China²¹. During the new-corona epidemic control period, pollution from other sources such as industry, automobile vehicles were significantly decreased, however, the activities of straw incinerations in vast rural regions kept going on as per traditional farming seasons. The observation of NO_2 concentration increased at about 12:00 and reached the peak at about 18:00 which was consistent with the practice of farmer usually burning the straw in afternoons.

Statistics of aerosol light absorption coefficient and AAE. Mean values of aerosol light absorption coefficients σ_{abs} at seven wavelengths and AAE during different stages are summarized in Table 1. During the COVID-19 strict control period, the light absorptions of both ultraviolet ($\sigma_{abs,370}$) and infrared ($\sigma_{abs,880}$) wavelength in Nanning were significantly lower than reported data that in Xiamen²² and in Lhasa²³.

Stages	$\sigma_{\text{abs},370}$ (Mm^{-1})	$\sigma_{\text{abs},470}$ (Mm^{-1})	$\sigma_{\text{abs},520}$ (Mm^{-1})	$\sigma_{\text{abs},590}$ (Mm^{-1})	$\sigma_{\text{abs},660}$ (Mm^{-1})	$\sigma_{\text{abs},880}$ (Mm^{-1})	$\sigma_{\text{abs},950}$ (Mm^{-1})	AAE
PPP	23.5	12.5	9.6	7.6	6.3	3.5	3.0	2.2
PAP	15.3	8.9	7.0	5.7	4.7	2.7	2.3	2.0
PDP	3.8	2.5	2.0	1.7	1.4	0.8	0.7	1.7

Table 1. Aerosol light absorption properties.

It should be noted that the value of AAE could be used as an indicator of aerosol from burning processes of fossil fuel or biomass²⁴, as the major content of fossil fuel burning is black carbon (BC), thus, the AAE of aerosol caused by fossil fuel burning is close to 1^{25,26}. In contrast, aerosol from BB contain rich amount of brown carbon (BrC), so it could generate even larger AAE than that from fossil fuel²⁴. In this case, AAE value of aerosol from BB was usually larger than 2.0²⁷. In this study, AAEs of PPP and PAP were 2.2 and 2.0 respectively, which implied that aerosol absorptions in Nanning city could be influenced by the burning of both biomass and fossil fuel. Additionally, considering the significant reduction of fossil fuel consumption during the special control period of new-corona epidemics, the BB could be the dominator factor resulting in the higher AAE in PPP and PAP, which was also supported by the large number of fire spots detected by satellites (commercial data) (Figure 3).

Impact analysis of secondary pollution. The formation of secondary particulate matter might also significantly contribute to this pollution event, this conclusion was based on the following two reasons. The first is the synergistic effects of NO_2 , NH_3 in the condition of high relative humidity on promoting the liquid-phase oxidation of SO_2 , which led to the obvious increase of sulfate particulates in PAP. Figure 4 shows the relationships between sulfate particle number concentration with levels of SO_2 , NO_2 , NH_3 and RH, and it seems that the positive correlations between concentration of sulfate particulates and NO_2 , NH_3 , RH were identified during the PAP with relatively better linearity. However, in the stage of NPAP (PPP and PDP), there was no significant linearity correlation even between SO_2 with sulfate particulate identified. During the whole observation period, the concentration of SO_2 remained at a relatively stable level maybe due to the reduction of industrial emission and absence of obvious SO_2 source around the observation site in the central district of Nanning city, and there was no significantly increase of SO_2 concentration even during the stage of PAP when the height of boundary layer decreased. Interestingly, the concentrations of sulfate particulates showed the positive correlation relationships with NO_2 , NH_3 and RH during PAP. It may be caused by the coexistence of NO_2 and NH_3 could promote the conversion of SO_2 to sulfate particulates under the condition of relatively high humidity, which has been reported in the previous studies through both experimental simulation and on-field observations and NO_2 would enhance the liquid phase oxidation of SO_2 in the cloud process or in the existence of NH_3 under high RH conditions. Additionally, Wang et al.²⁸ adopted the method of Density Function Theory (DFT) to

investigate how H_2O and NH_3 promoted the oxidation reaction of SO_2 and NO_2 , and found that NH_3 played an important role in stabilizing the complex products. Chen et al.²⁹ employed the glassware reactors and demonstrated that NH_3 could increase the dissolution of SO_2 and hence enhanced the liquid-phase oxidation of SO_2 .

Secondly, the increase of secondary particulates could be explained by the positive feedback mechanism of pollution boundary layer (PBL)-relative humidity (RH)- secondary particles matter (SPM)-particulate matter (PM), which has been proposed in the previous studies, and the relative humidity (RH) and the PBLH are essential factors which may affect the formation of atmospheric $\text{PM}_{2.5}$ ^{30,31}. The positive feedback mechanism of PBL-RH-SPM-PM suggests that PM level and RH would be low while PBLH is high at the beginning stage of a haze event. Since the atmospheric diffusion condition could be adversely changed by weather situation, PM would begin to accumulate, then the radiation effects due to the increase of PM could cause PBLH decreasing, and further induce the increase of PM and RH. On the other hand, the moisture absorbed by PM would increase and enhance the radiation effect which could further decrease PBLH. Thus, RH would keep increasing and accordingly enhance the formation of SPM. So, through the comprehensive analysis of extinction coefficient, PBLH, PM and RH which were observed by lidar, it is concluded that PBL-RH-SPM-PM positive feedback mechanism is an ideal model to elucidate the occurrence and development of the pollution.

Figure 5(a) and (b) show the time series of the vertical distribution of the extinction coefficient and depolarization ratio of the lidar at 532 nm from February 18-24, 2020, respectively. They demonstrate that the near-ground extinction coefficients for Nanning were relatively small in PPP and gradually increased; its depolarization ratio was relatively small with little variation for the whole observation period. The PBLH gradually reduced from about 2 km to 1 km which was adverse for atmospheric convection and resulted in the atmospheric diffusion condition worse, thus the pollutants began to accumulate as concentrations of $\text{PM}_{2.5}$ and secondary inorganics aerosol (SIA) levels slowly increased (Figure 5(c) and (d)). Specially, at 12:00 on February 20 in PAP, the wind speed was small and PLH was relatively low (minimum values was 293 m only), the atmospheric diffusion condition turned even worse and accordingly caused the cumulative accumulations of pollutants. At about 0:00 20 Feb, the extinction coefficients near-ground suddenly increased, meanwhile, the concentration of $\text{PM}_{2.5}$ and SIA level also rapidly increased and reached peak value (179 ug.m^{-3} for $\text{PM}_{2.5}$). From 00:00, Feb. 20th to 12:00, Feb. 21st, the boundary layer was slightly uplifted, and the wind speed got faster, then the vertical diffusion conditions improved and the level of pollutant consequently declined. Afterwards, the boundary layer dropped and wind speed decreased from 12:00 to 22:00 on Feb. 21st, and $\text{PM}_{2.5}$ and SIA level continued to rise to reach the peak. From 0:00 to 21:00 on Feb. 22nd, the diffusion capability was relatively good, $\text{PM}_{2.5}$ and SIA decreased gradually, however, at 22:00, near-ground distinction coefficient suddenly increased, $\text{PM}_{2.5}$ and SIA rose up and reached a peak value again at 0:00 23rd. Then $\text{PM}_{2.5}$ and SIA gradually decreased with atmospheric diffusion, and at 0:00 23rd, extinction coefficient rose up again, $\text{PM}_{2.5}$ and SIA climbed up slowly afterwards. Finally, in the period of PDP, boundary layer was uplifted,

wind speed got stronger, the overall atmospheric diffusion condition turned better with relatively small amount of pollutant emissions near ground, thus the levels of PM_{2.5} and SIA stepwisely declined and maintained at relatively low levels.

It could be concluded that near-ground PM_{2.5} accumulated as PBLH significantly decreased, other factors including the increase of moisture level, higher RH and local straw burning also resulted in the slight pollution. Additionally, the extinction coefficient would increase with the increase of PM_{2.5}; PBLH would be further lowered down, and with the increase of RH, more moisture would be absorbed by PM_{2.5}, which could enhance the gas-particulate formation to generate more SIA. In all, the high RH, low PBLH, poor horizontal and vertical atmospheric diffusion capacities combined with the effects of local straw burning, made the pollution gradually turn worse. Thus, on February 24, PBLH was obviously uplifted, and wind speed got faster, then the diffusion factors turned better, and then the air quality improved.

Analysis for the pollutions from regional transportation. It is well known that pollutants generated by BB could be transported to long distances^{32,33}. As there were many fire spots identified in the regions around Nanning City, the pollutions from regional transportation should also be considered. The HYSPLIT model proposed by NOAA (<https://www.arl.noaa.gov/>) was adopted to analyze the airflow back trajectory of pollutants during the sampling period. Airflows at the heights of 500 m, 1000 m and 2000 m were selected to calculate the backward trajectory figure (Figure 6(a), (b), (c), (d)). From February 18 to 21, the air masses of 500 m and 1000 m were mainly influenced by the air flows from Guangdong Province and Beibu Gulf located in the southeast direction, while air masses at 2000 m were mainly influenced by the airflow from Vietnam. Based on the satellite remote sensing monitoring maps (Figure 6.), it was found that lots of fire spots were in Vietnam, Laos, Thailand and Cambodia during the period of pollution event occurred in Nanning. The backward trajectory calculations indicated that the long distance transportation of biomass incineration pollutants from these countries would have some influence on the regional atmosphere in Nanning. Recently, Yue et al.³⁴ showed that CO level near the ground decreased 17% compared with the same period of the previous year, while the concentration of CO in troposphere increased by 2.5%. These also supported the previous conclusion that long distance transportation of biomass incinerations from foreign countries could affect the atmosphere in the south of China. In this study, the increase of CO level during PAP in Nanning was identified, and it was also believed due to the influence of BB both in regional cities and in Southeast Asia. Interestingly, on February 22 (Figure 6(e)), it had been showed that the origins of air masses at 500m, 1000m and 1500m were from localities within Nanning. Based on the analysis of meteorological land weather conditions, it was found that the pollutants accumulated as a wind convergence zone was over Nanning that day. However, on February 23 (Figure 6(f)), the airflow was from cities including Laibin and Guigang which had the relatively intensive fire spots; on February 24 (Figure 6(g)), the airflows of three heights were mainly from neighboring Guangdong province and Beibu Gulf, and as the meteorological diffusion conditions had been improved, thus the pollution began to gradually dissipate. Overall, based on the analysis of fire spots contribution maps and backward trajectory, it had concluded that the air masses had passed the

regions of intensive fire spots, and this PM_{2.5} pollution event in Nanning was generated not only from local BB but also from transportation from surrounding countries in Southeast Asia.

Conclusion

In this work, we comprehensively investigated the sources and causes of PM_{2.5} pollution that occurred in Nanning of Guangxi during the COVID-19 lockdown in February 2020. Three haze stages were categorized as PPP, PAP and PDP, with average hourly PM_{2.5} concentration 32, 73 and 35 $\mu\text{g}\cdot\text{m}^{-3}$, respectively. The dominant source of PM_{2.5} in PPP was BB (40.4%), followed by secondary inorganics (28.1%) and motor vehicle exhaust (11.7%). The PAP was characterized by a large abundance of secondary inorganics, which contributed for 56.1% of the total PM_{2.5} concentration, followed by BB (17.4%). The significant increase of secondary inorganics could be due to the high concentrations of NO₂ and NH₃, which enhanced the liquid-phase oxidation of SO₂ under relatively high humidity. Interestingly, absorption Ångström exponent (2.2) in PPP was higher than the other two periods, and based on the analysis of fire spots monitored by remote satellite sensing, aerosol absorptions in Nanning city could be influenced by the burning of both biomass and fossil fuel. Additionally, pollutants produced by BB accumulated and caused haze event in PAP, which was partially due to the unfavorable meteorological conditions such as increased humidity and decrease of PBLH. The PBL-RH-SPM-PM positive feedback mechanism was employed to elucidate the atmospheric process in this study, which indicated that it is an ideal model to explain the occurrence and development of the pollution. Finally, this study highlights the importance of understanding the role of BB and meteorology to call for policy for emission control strategies.

Material And Method

Description of sampling site. Located in southwestern China, Nanning is the provincial capital of Guangxi (Figure 7). Ambient measurements including Single Particulate Aerosol Mass Spectrometer (SPAMS), Aethalometer (AE-31), particulate Lidar and solar radiometer were conducted at the atmospheric observation station set in the Scientific Research Academy of Guangxi Environmental Protection (SRAGEP 22.8067°N, 108.3335°E), Nanning, China, which was a height of about 30m above the ground and surrounded by heavy traffic, residential area and restaurants. Additionally, there are eight national air-quality monitoring stations in Nanning, and Figure 7 represents the locations of our sampling site and the eight national stations. It should be noted that the sampling site was located in the Nanning center region, so it was representative of Nanning urban area.

Measurements of pollutants. PM_{2.5} was measured with combined methods of aerosol light scattering and beta ray attenuation (Synchronized Hybrid Ambient Real-time Particulate Monitor, 5030i, Thermo Scientific), and the PM₁₀ measurement was performed by beta attenuation method (Continuous Particulate Monitor, FH 62 C14, Thermo Scientific). The concentrations of CO, NO and NO₂, SO₂ were monitored by the method of infrared radiation absorption (CO Analyzer, 48i, Thermo Fisher Scientific),

Chemiluminescence (NO-NO₂-NO_x Analyzer, 42i-HL, Thermo Scientific) and pulsed UV fluorescence method (SO₂ analyzer, 43i, Thermo Scientific), respectively. All above data were real-time recorded by 24 hours a day with continuously sampling.

Single Particulate Aerosol Mass Spectrometer (SPAMS). SPAMS has been employed to monitor both the diameter and chemical compositions of single particulate aerosol^{35,36} and the applications of SPAMS have been widely described in many published literature³⁷⁻³⁹. Briefly, ambient particles were introduced into the SPAMS with the sampling flow rate of 75 mL.min⁻¹. Aerodynamic lens were used to achieve the focus of accelerated single aerosol particulates, and the aerodynamic diameters of particles of 0.13–3 μm could be measured by scattering signals of two continuous Nd: YAG laser beams. A laser (wavelength of 266 nm with the energy of 0.5~0.6 mJ and energy density of 108 W.cm⁻²) was simultaneously triggered to ionize the measured particle, and the generated cations and anions were analyzed by dual polarity time-of-flight mass spectra. Generally, the range of particle diameters could be analyzed at 0~2.5 μm with the rate of 20 particles.s⁻¹ and bombarded rate of >20%. The MS resolution was better than 500 FWHM, and the measurable range of chemical composition (molecular weight) is 1~500u.

Aethalometer. Particles collected on quartz fiber filter (QFFs) were analyzed by Aethalometer (AE-31 Magee Scientific Inc., USA) with 7 measurement wavelengths (370, 470, 520, 590, 660, 880, 950 nm). The attenuation (ATN) of light beam transmitted through the sample was measured and the b_{ATN} of particulate time zone was calculated based on the change rate of ATN (ΔATN) by the equations below⁴⁰: **See formulas 1 and 2 in the supplementary files.**

Where I_0 and I are the light intensity before and after the light beam passing through the filter membrane (W·cm⁻²), respectively. A is the area of the spot on quartz belt, cm². Δt is the reading period (5 min used in this study). Q is the air flow rate, L.min⁻¹ (4.9 L.min⁻¹ in this study). When particles accumulated in one spot on the filter belt reached the largest attenuation value -125 (λ at 370 nm), the belt would be shifted to the next spot and the analysis was initialized, thus the testing processes were automatically completed in cycled periods. Light absorption coefficient of aerosols (σ_{abs}) was calculated based on the measured light attenuation at seven wavelengths as shown in the published literature^{22,41,42}. Determination of absorption Ångström exponent (AAE) was calculated based on the previous studies^{22,43}.

Particulate Lidar. The application of particulate lidar is based on the return signal due to elastical backscatter by atmospheric particles, and the processes could be expressed by equation (3) as:

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Particulate Lidar. The application of particulate lidar is based on the return signal due to elastical backscatter by atmospheric particles, and the processes could be expressed by equation (3) as: **see formula 3 in the supplementary files.**

Where $P(R)$ is the energy received by the lidar from the backscattering signals of atmosphere at altitude R ; E_0 is the emission energy of laser; η_L is the overall efficiency of the optical and detected parts of lidar system; $O(R)$ is the laser overlap factor; $\beta(R)$ and $\alpha(R)$ represent the backscatter coefficient and extinction coefficient of atmosphere, respectively. Generally, different approximating methods were proposed to calculate the actual $\beta(R)$ and $\alpha(R)$ in the literature, and “Fernald method” has been widely adopted among all of them^{44,45}. Lidar system used in this study (AGHJ-I-LIDAR, Wuxi CAS Photonics Co., Ltd.) was composed of laser emission unit, optical receiving unit and signal acquisition unit. The laser emission unit is mainly composed of laser and emission telescope. The detective wavelength was 355 nm and 532 nm, and the single pulse energy was about 20 mJ. The optical receiving system consisted of a Cassegrain telescope, a narrow-band filter and a photo detector. The laser emitting unit emitted 355 nm and 532 nm detective pulses to the target area, and the pulses were scattered by atmospheric aerosols, particles or clouds in the transmission path. Then the backscattered light was received by the receiving telescope, and the backscattered signal light successively passed through small aperture, collimator and spectroscopy and divided into two signals with the wavelengths of 355 nm and 532 nm which were received and finally converted to the corresponding electrical signals by photomultiplier after polarizing prism.

Meteorological factors. Meteorological factors included precipitation, temperature (T), relative humidity (RH), air pressure (P), wind speed (WS), wind direction (WD), visibility (V) and total solar radiation were monitored by solar radiometer (Beijing Huatron Technology Development Co. Ltd.) with temporal resolution of 1 min. Hourly average of each factor was adopted in this study.

Declarations

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Author Contributions: Zhaoyu Mo: Formal analysis, Writing-Original Draft, Data Curation. Jiongli Huang: Methodology, Writing-Original Draft. Zhiming Chen: Formal analysis, Data Curation. Bin Zhou: Conceptualization, Methodology, Supervision. Kaixian Zhu: Writing-Original Draft. Huilin Liu: Investigation. Yijun Mu: Investigation. Dabiao Zhang: Investigation. Shanshan Wang: Writing-Review&Editing.

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Table

Stages	$\sigma_{\text{abs},370}$	$\sigma_{\text{abs},470}$	$\sigma_{\text{abs},520}$	$\sigma_{\text{abs},590}$	$\sigma_{\text{abs},660}$	$\sigma_{\text{abs},880}$	$\sigma_{\text{abs},950}$	AAE
	(Mm ⁻¹)	(Mm ⁻¹)	(Mm ⁻¹)	(Mm ⁻¹)	(Mm ⁻¹)	(Mm ⁻¹)	(Mm ⁻¹)	
PPP	23.5	12.5	9.6	7.6	6.3	3.5	3.0	2.2
PAP	15.3	8.9	7.0	5.7	4.7	2.7	2.3	2.0
PDP	3.8	2.5	2.0	1.7	1.4	0.8	0.7	1.7

Table 1. Aerosol light absorption properties.

Figures

Figure 1

Time series of (a) meteorological conditions (WS, WD and PBLH); (b) the main pollutants (PM₁₀, PM_{2.5} and CO); (c) the main gaseous pollutants (O₃, SO₂, and NO₂); and (d) meteorological conditions (temperature and RH) during the study period at the SRAGEP site.

Figure 2

Temporal change for the percentage of single particle source resolution

Figure 3

Distribution of fire spots in Guangxi from February 18 to 24, 2020. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 4

Sulfate particle number concentration versus (a) concentration of SO_2 ; (b) concentration of NO_2 ; (c) concentration of NH_3 and (d) RH during the pollution accumulation period (PAP) and non-pollution accumulation period (NPAP, i.e. Pre-pollution and pollution dissipation period) from February 18 to 24, 2020.

Figure 5

(a) and (b) are the time series of the vertical distribution of the extinction coefficient and depolarization ratio of the lidar at 532 nm from February 18 to 24, 2020, respectively, and (c) is the time sequence diagram of PM_{2.5} mass concentration and secondary inorganic particle number concentration in the same period. (d) is a diagram of RH and PBLH from February 18 to 24, 2020.

Figure 6

24hrs backward trajectory figure from 18th to 24th February, 2020 in Nanning (Fire Spots Map of Southern China and Southeast Asia was cited from NASA data, <https://firms.modaps.eosdis.nasa.gov/>). Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 7

Schematic diagram for the locations of observation stations. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

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