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Elucidating the Factors Governing the Interannual Variability of Ozone Concentrations During Fall 2015-2019 in Sanya, China

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Abstract. While ozone pollution has been a major air pollution concern in metropolitans in China, the characteristics and governing factors of ozone concentrations in Sanya remains unclear. In this study, we first analyze the interannual variability of ozone based on observational data in Sanya, and identify it is in general characterized by a peak ozone season in fall and minimal ozone season in summer. Meanwhile, the substantial ozone enhancement in 2019 compared to the previous three to four years over Sanya clearly stands out. To elucidate the possible governing factors, we design a few numerical experiments based on regional air quality model, and find that the modulation of meteorology is key to steering the interannual variability of ozone in fall in Sanya. The spatial evolution further indicates that the transport from upwind regions like Pearl River Delta region is crucial in stimulating the ozone accumulation in Sanya. In addition, ship emissions play important roles in further enhancing their ozone concentration, ranging from 7% -10% during 2015-2019. The findings in this study imply that whereas an overall low ozone concentration in Sanya, ozone exceedance may still occur in particular under unfavorable meteorological conditions together with the concomitant transport from other regions facing ozone pollution. It stresses the importance of regional emission control, including anthropogenic emissions and ship emissions, on improving air quality in Sanya.

1. Introduction

Ozone concentrations in urban areas of China increase at a rate of 1-3 ppbv y^{-1} during 2013-2017[1], and major sources include anthropogenic emissions, ship emissions and biogenic emissions [2-4]. For example, in major urban agglomerations such as the Yangtze River Delta and the Pearl River Delta in China, anthropogenic sources contribute up to 39-73 ppbv when daily maximum 8-hour (MDA8) O_3 is greater than 100 ppbv in 2017[5]. In ports and major waterway areas, ship emissions can affect ozone production and associated free radical formation over the area, affecting air quality[6-8]. In addition, adverse meteorological conditions strongly affect the formation and accumulation of ozone [9, 10].

The 90th percentile ozone concentrations in Sanya show a generally increasing trend during the 13th Five-Year Plan period and peaked at 55.07 ppbv in 2019[11]. Despite of good air quality in general in Sanya, ozone exceedance still occurs sometimes. Therefore, this study aims to elucidate the effects of meteorological factors and ship emissions on ozone pollution in Sanya based on the WRF-CMAQ model, which is useful for ozone pollution control therein.

2. The model

2.1 Model configurations

In this study, the Weather Research and Forecasting Model (WRF v3.8.1) and the Community Multi-scale Air Quality Model (CMAQ v5.2) was used for the simulation. The simulation area is shown in Figure 1.

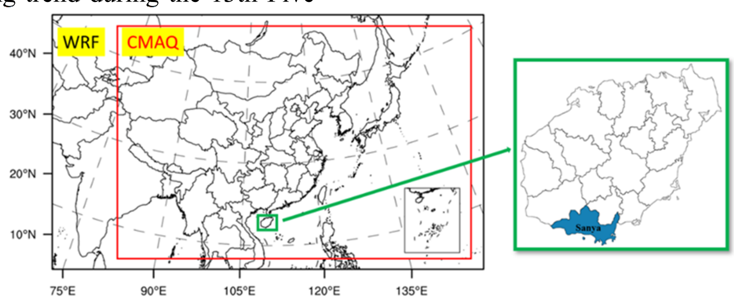

Figure 1. Simulation area and research area, black and red boxes indicate the simulation area for WRF and CMAQ respectively, and the green box refers to Hainan Island

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The initial and boundary conditions for the WRF model simulations were taken from the NCEP Climate Forecast System Reanalysis (CFSR) version 2[12]. The physical options used in this study are mostly the same as those used in our previous studies[13, 14]. The CMAQ v5.2 model was used to simulate ozone concentrations based on the same projection coordinate system as the WRF, using the Carbon Bond version 6 (CB6) gas-phase chemistry module[15] and the Aerosol Module Version 6 (AERO6) aerosol module[16], with initial and boundary conditions from the Model for Ozone And Related Chemical Tracers, version 4[17]. The simulation period is autumn 2015-2019.

2.2 Data sources

Anthropogenic emission inventories were obtained from MEICv1.3[18, 19], and the year of 2016 was applied. Biogenic emission inventories were obtained from the Model for Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN v2.1)[20]. Biomass burning emission inventories were obtained from the Global Fire Emissions Database, version 4 (GFED v4) [21]. Ship emission inventories were from the Global Ship Emissions Database (SEIM)[22, 23]. Meteorological observational data used for model evaluation were from the National Climatic Data Centre (NCDC, http://www.ncdc.noaa.gov/data-access/quick-links#dsi-3505). Hourly ozone monitoring data were available at the National Environmental Monitoring Center of China (CNEMC, http://www.pm25.in). 850 hPa wind vector data were obtained from the Copernicus Climate Data Store (Copernicus Climate Data Store, https://cds.climate.copernicus.eu/#!/home).

2.3 Simulation scenario setting

In order to quantify the contribution of meteorological factors and emissions to ozone in the city of Sanya, three scenarios were set up in this study. Base case is defined as the simulations with all emission sources. The scenario of Sce1 includes all emissions except ship emissions, so the differences between Base and Sce1 reflect the influence of ship emissions. Another scenario of Sce2 only includes anthropogenic emissions, so the interannual variability of ozone can be used to quantify the effect of meteorology.

2.4 Model evaluation

The WRF model simulation results were evaluated using Sanya meteorological observation data (Table 1). Overall, the model simulations well reproduce the meteorological parameters in Sanya, e.g., 2 m air temperature. Nevertheless, the mean bias and gross error of simulated 10 m wind speed and wind direction deviation is slightly larger than the benchmarks. The bias in wind speed is linked to the deviation of the simulated sea-land thermal difference in the coastal area, and large gross error in wind direction is likely attributable to the wind direction near 0 degree, e.g., wind directions close to 360 degree tend to yield large errors for observations with values of a few degrees although the directions are comparable to each other [24, 25].

Table 1. Evaluation of WRF simulation results

Variables		Sanya	Benchmarks[26]
Air temperature at 2m (℃)	Mean OBS	23.83	
	Mean SIM	23.07	
	Mean Bias	-0.76	≤±0.5
	Gross Error	1.56	≤2
Wind Speed at 10 m (m/s)	Mean OBS	5.68	
	Mean SIM	3.32	
	Mean Bias	-2.37	≤±0.5
	Gross Error	2.76	
Wind direction at 10 m (deg)	Mean OBS	95.72	
	Mean SIM	95.10	
	Mean Bias	-0.62	≤±10
	Gross Error	59.62	≤30

In addition to meteorology, the model performs well in terms of MDA8 O_3 in the autumn of 2015-2019 in Sanya (top to bottom panels in Figure 2), meeting the criteria of -30%<MFB<30% and MFE<50% recommended by Boylan et al[27]. It is noteworthy that for the periods with ozone peaks, the model tends to show underestimation.

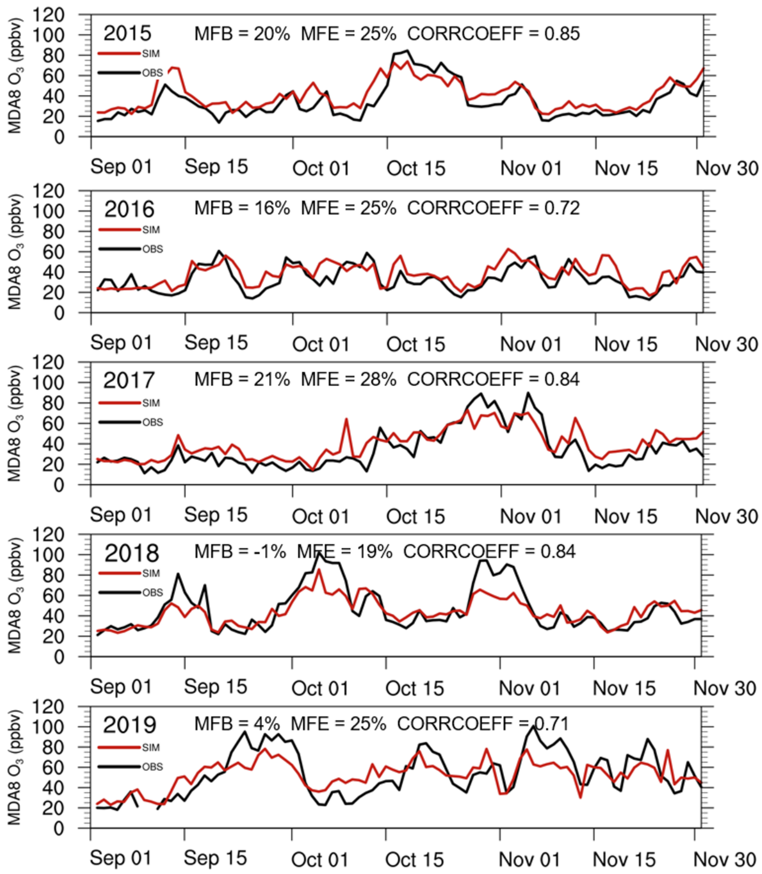

Figure 2. Simulation evaluation of MDA8 O_3 in fall of Sanya from 2015 to 2019

3. Results

3.1 Observation-based ozone pollution characteristics in Sanya

Monthly mean MDA8 O_3 from 2015-2019 is shown in Figure 3, with the 25th and 75th percentile in green shading and minimal (0 percentile) and maximal (100 percentile) in blue. It clearly delineates that the ozone peak season in Sanya is primarily in fall, whereas summer tends to be concomitant with the lowest seasonal ozone concentrations, likely attributable to the frequent rainfall therein. Therefore, the monthly MDA8 O_3 in Sanya is characterized by a V-shape, which normally increases rapidly from August to November, and then decreases gradually from December to July of the next year.

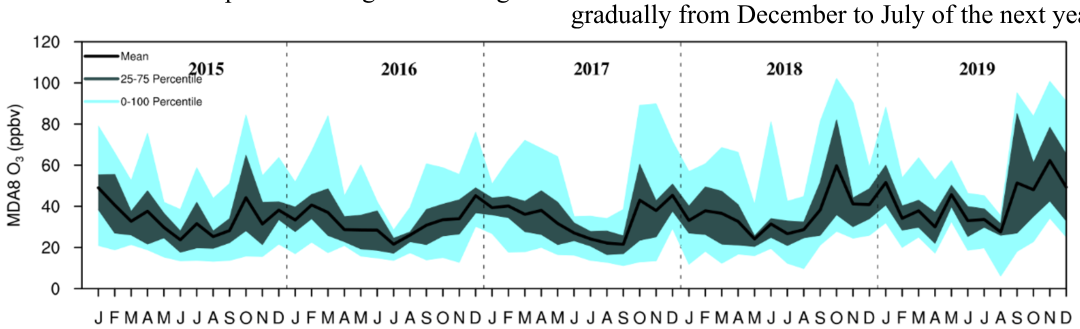

Figure 3. Observational monthly variation of MDA8 O_3 concentrations in Sanya from 2015 to 2019

To delve into the ozone evolution in seasonal perspective, the interannual variability of observed seasonal mean MDA8 O_3 during 2015-2019 is shown in Figure 4. There is a general increasing trend of annual MDA8 O_3. In particular, after 2016, the concentrations of MDA8 O_3 rise at an annual growth rate of 3.24 ppbv y^{-1}, and reach 42.07 ppbv in 2019. It is worth noting that MDA8 O_3 concentrations in the fall of 2019 in Sanya was substantially higher than that in other seasons during the five years, reaching 53.94 ppbv, with the specific reasons explored in the sections below.

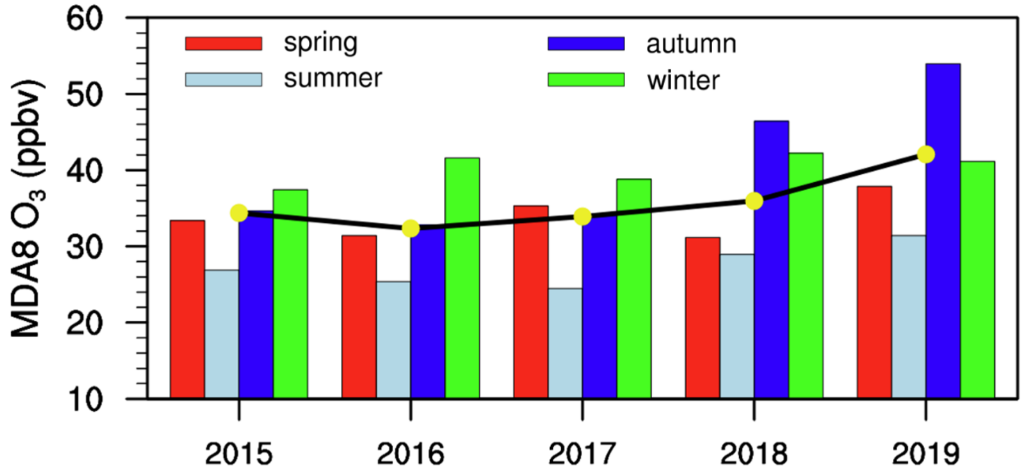

Figure 4. Interannual variations of seasonal (bars) and annual (orange dots) mean observational MDA8 O_3 in Sanya from 2015 to 2019

3.2 The impact of meteorology on interannual variability of ozone in Sanya

The correlations between hourly ozone and a few meteorological parameters are shown in Table 2. The statistically significant positive correlation is between downward surface solar radiation and ozone, indicating a stronger effect of downward surface solar radiation compared to near surface air temperature, which is consistent with our recent finding in elucidating the abnormally high ozone in fall 2019 over Pearl River Delta region[28]. The large negative correlation between ozone and relative humidity indicates the sink effect of water vapor. It is a bit wield that positive relationship between ozone and wind speed appears, which in general atmospheric stagnant conditions favors the ozone accumulation[29]. To understand the possible governance of wind vector on ozone in Sanya, a scatter plot is drawn among hourly wind direction, wind speed and ozone concentrations (Figure 5). The high ozone concentrations in Sanya mostly occur under the northeasterly wind conditions, indicating that strong transport from upwind areas such as Pearl River Delta region is likely an important contributor aggravating the ozone pollution in Sanya.

Table 2. Correlation coefficient between fall ozone concentration and meteorological factors in Sanya from 2015 to 2019

U_10m	V_10m	WS	RH	T_2m	RAD
-0.35*	-0.28*	0.29*	-0.60*	-0.09	0.31*

* means $p < 0.01$. U _ 10, V _ 10, WS, RH, T _ 2 and RAD represent 10 m wind speed U component, 10 m wind speed U component, wind speed, relative humidity, 2 m air temperature and downward surface solar radiation, respectively.

As was discussed in section 2.3, the interannual variability of ozone based on the scenario of Sce2, maintaining the same year of anthropogenic emissions only, reflects the modulation of meteorology. To this end, the mean MDA8 O_3 in fall in 2015 is shown in Figure 6a, with the differences between 2016-2019 and 2015 is displayed in Figure 6b-e. Compared to 2015, the mean MDA8 O_3 contributed by the meteorology is -2.38 ppbv, -0.42 ppbv, 6.21 ppbv, 14.85 ppbv, respectively from 2016 to 2019, indicating a substantially high ozone modulation from meteorology in 2019. Considering the discussion in our previous study[28], as well as the section above, it indicates the meteorological conditions, e.g., abnormally high downward surface solar radiation, induces the widespread ozone concentration increase in fall 2019 over the broad areas in eastern China including Sanya, and the high ozone in southeastern China particularly over Pearl River Delta region may play a large role in ozone enhancement of Sanya during the favorable wind conditions.

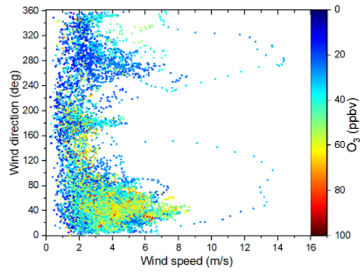

Figure 5. Scatter plot of simulated hourly ozone and wind speed and direction in Sanya autumn from 2015 to 2019

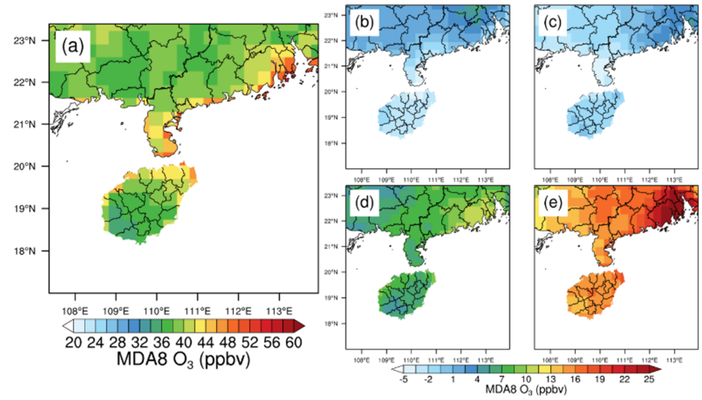

Figure 6. The effect of meteorology on MDA8 O_3 in fall under the same anthropogenic emissions. 2015 (a), 2016-2015 (b), 2017-2015 (c), 2018-2015 (d), 2019-2015 (e)

3.3 Impact of ship emissions on ozone in Sanya

Considering that Sanya is a coastal city, it is useful to quantify the contribution of ship emissions on ozone concentration in Sanya. The contribution of ship emissions to ozone can be obtained by subtracting Sce1 from base scenario (section 2.3). As shown in figure 7, the contribution of ship emissions to MDA8 O_3 over Sanya becomes larger in fall 2018 and 2019 in the absolute value compared to the previous three-year, whereas the overall fractional contributions are comparable (10% in 2018, 9% in 2019 vs. 9%, 8% and 7% during 2015-2017). It implies that ship emissions do play an important role affecting ozone concentration in Sanya, whereas the substantial ozone enhancement in 2019 is primarily triggered by meteorology as discussed above.

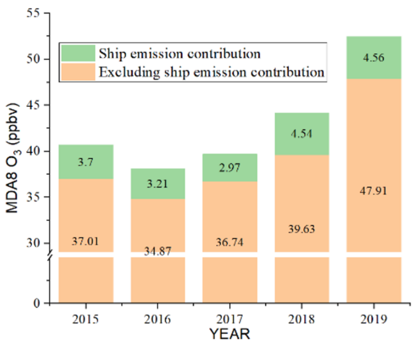

Figure 7. Contribution of different sources to ozone in Sanya, fall 2015-2019

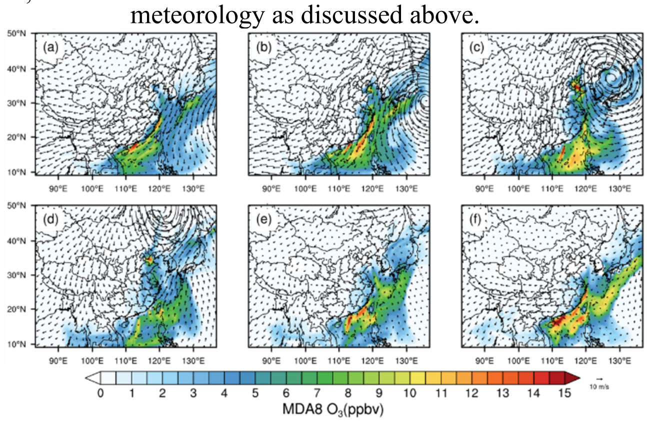

Figure 8. 850 hPa wind vector and MDA8 O_3 contributed by ship emissions (a-f) from October 2 to 7, 2018

In order to further view the spatial evolution of ship emission contributions to ozone, we select a period with relatively high ozone concentrations (October 2-7, 2018) and the MDA8 O_3 evolution contributed by ship emissions (Base minus Sce1) is displays in Figure 8. From October 2 to 7, 2018, due to the strong northeasterly wind and the sinking airflow outside the typhoon, ozone from ship emissions in the southern coastal area of China was transported southwestward, resulting in a few ppbv ozone enhancement in Sanya. Previous studies showed that high ozone pollution events in Hainan Island was mainly caused by the northeasterly wind concomitant with a cold high-pressure system in the northern flank, the warm high-pressure ridge or sinking airflow around the typhoon[30, 31]. The findings further emphasize the critical role of extreme weather events such as typhoon and the transport on ozone pollution in Sanya.

4. Conclusions

Based on observational ozone concentrations from 2015 to 2019, we find the seasonal ozone in Sanya tend to show peak in fall but low value in summer. The interannual variability indicates a generally increasing trend during 2015-2019, in particular after 2016, the concentrations of MDA8 O_3 show an annual growth rate of 3.24 ppbv y^{-1}. Numerical experiments based on WRF-CMAQ model show that meteorological conditions are the key to modulating the interannual variations of ozone in Sanya. It is noteworthy that the northeasterly wind, which can transport ozone from the upwind regions like Pearl River Delta region, favors the ozone accumulation in Sanya. In addition, ship emissions play important roles in enhancing the ozone concentration, e.g., more than 7 ppbv in typical cases. This study quantifies the factors affecting ozone concentrations in Sanya, potentially providing a useful guidance for the air quality improvement therein.

Acknowledgments

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