Characteristics and Meteorological Conditions of Ozone Pollution in Shantou City

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Abstract Based on routine monitoring data of air quality and meteorological data in Shantou City during 2015 – 2019, temporal-spatial variation characteristics of O_3 pollution and its correlation with meteorological conditions in Shantou City were explored. The research results showed that O_3 pollution days in Shantou City showed an increasing trend year by year, and O_3 pollution had far-distance transportation and the development trend from offshore Nan'ao Island to urban district. In spring and autumn, there was serious O_3 pollution, and it was the most prominent in October. Its diurnal variation in O_3 pollution days was mainly wide-peak type in the afternoon, showing as that O_3 concentration declined slowly after the noon. In O_3 pollution days, higher O_3 concentration was easy to appear at night, causing that O_3 peak in the second day was uplifted, and there was continuous O_3 pollution. Combining backward trajectory analysis chart, it was found that Shantou was mainly affected by coastal transport of northerly polluted air mass, and it was transported into Shantou City from the east to the northeast. O_3 from long-distance transmission superimposed with locally generated O_3, which commonly pushed up the level of O_3 concentration. The weather of O_3 pollution in Shantou City had the characteristics of high temperature and low humidity. There was 25 – 30 ℃ of temperature interval and 46% – 60% of relative humidity interval, and it was accompanied by grade-2 easterly wind.

Key words Shantou City; Ozone; Pollution characteristics; Meteorological conditions

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Ozone (O_3) is a product of photochemical pollution in the troposphere, a key factor of atmospheric oxidation capacity, and an important greenhouse gas[1]. Seen from formation mechanism of O_3, O_3 near the ground is secondary pollutant of some precursors (NO_x and VOCs) via a series of photochemical reactions under the action of solar radiation[2], and is an important pollutant of affecting urban ambient air quality, which has important influence on atmospheric radiation and ecological environment[3]. In recent years, atmospheric photochemical pollution taking O_3 as characteristic pollutant has become an increasingly prominent atmospheric environmental problem in urban areas of China with economic globalization and rapid development of market economy[4]. Since the promulgation and implementation of the *Ambient Air Quality Standard* (GB 3095 – 2012), O_3 monitoring has been successively carried out in cities above prefecture level, especially it was fully implemented in Beijing – Tianjin – Hebei, the Yangtze River Delta and the Pearl River Delta since 2013. The results showed that O_3 pollution problem in the cities of China was prominent and concentrated. In 2015, 75.9% of prefecture-level cities with O_3 exceeding the standard in China were in the three regions[1]. The proportion of pollution days taking O_3 as primary pollutant gradually rose since 2016. In 2017 and 2018, primary pollutant of all polluted weather was O_3, and pollution concentration showed a rising trend. O_3 has replaced fine particulate matter ($PM_{2.5}$) and inhalable particulate matter (PM_{10}) as the primary factors restricting air quality comprehensively reaching the standard in Shantou City.

Shantou is a major coastal port city and an important hub of comprehensive transportation system. At present, there is less research on O_3 pollution in Shantou City, and the study on O_3 pollution characteristics in Shantou City has very important significance. In this paper, O_3 concentration monitored by each national monitoring point of Shantou City in the past 5 years (2015 – 2019) was used to analyze variation characteristics of O_3 concentration in different years, seasons, months and time, and the influence of meteorological factors on O_3 concentration was explored. The research aimed to provide theoretical support for understanding O_3 pollution characteristics in Shantou and making corresponding prevention and control measures.

1 Materials and methods

1.1 Data source Routine monitoring data of O_3 and air quality and meteorological data at each station of Shantou City from 2015 to 2019 were provided by Shantou Environmental Protection Monitoring Station. In this paper, O_3 concentration at six national ambient air quality monitoring sites of Shantou City was averaged as that in the urban area, and they were Jinping District, Longhu District, Chenghai District, Haojiang District, Chaoyang District, and Chaonan District. Nan'aozi Station was taken as suburban rep-

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resentative for comparative analysis with urban district, and the site was that of Guangdong evaluation website.

1.2 Analysis methods According to the *Ambient Air Quality Standards* (GB 3095 – 2012), the *Technical Regulation on Ambient Air Quality Index (on trial)* (HJ 633 – 2012), and the *Technical Regulation for Ambient Air Quality Assessment* (on trial) (HJ 663 – 2013), validity and evaluation method of data were judged. O_3 concentration of the 90^{th} percentile of the maximum 8 h (O_3-8 h-90 per) and exceeding days at each monitoring site of Shantou City during 2015 – 2019 were counted, and the condition and characteristics of O_3 pollution in Shantou City were analyzed. Using trajectory model of backward air mass (HYSPLIT4), air mass transport process in typical pollution period was simulated. Clustering was conducted according to air mass transport process. In different classification trajectories, quantitative analysis of pollutant source area and quantity was conducted. Combining local actual data and experience, source reliability of air pollutants was analyzed[5].

2 Results and analyses

2.1 Characteristics of O_3 pollution

2.1.1 General situation. Via statistical analysis, it was found that annual excellent days in Shantou City during 2015 – 2018 were between 337 and 357 d, and the standard-reaching rate was above 92%. The annual pollution days were 9 – 28 d, all of which were light pollution. Among them, air quality was better in 2016, and there were only 9-d light pollution in Shantou City in whole year, with 97.50% of standard-reaching rate. But the pollution days gradually increased after 2016, and the standard-reaching rate declined obviously. To 2018, the standard-reaching rate declined by 5.20% than that in 2016. Under the unified standard situation, the standard-reaching rate in 2019 was still further declining, and the pollution days increased by 9 d than that in 2018. It was clear that there was severe pollution situation.

O_3, $PM_{2.5}$, PM_{10} and NO_2 were main pollutants affecting air quality in Shantou City. The proportion of main pollutants during 2015 – 2019 was shown in Fig. 1. During 2015 – 2016, exceeding days of O_3 and $PM_{2.5}$ were equal in Shantou City. During 2017 – 2019, air quality in Shantou City not reaching the standard was all caused by O_3 pollution. O_3 pollution has become main environmental problem faced by Shantou City, which was increasingly serious.

2.1.2 Annual variation characteristics. Annual variation characteristics of O_3 in Shantou urban district and Nan'aozi Station during 2015 – 2019 were shown in Fig. 2. Annual average concentration of O_3 in Shantou City during 2015 – 2018 did not exceed grade-two evaluation standard of ambient air quality (160 μg/m^3). O_3-8 h-90 per and exceeding days in Shantou urban district and Nan'aozi Station during 2015 – 2016 showed a declining trend, and it overall showed a rising trend after 2016. O_3-8 h-90 per and exceeding days in Nan'aozi Station were the most serious in 2015, and were far higher than those in Shantou urban district. During 2017 – 2019, O_3-8 h-90 per in Shantou urban district was all higher than that in Nan'aozi Station, and pollution situation was more serious in Shantou urban district in 2019, illustrating that O_3 pollution had the development trend from suburb to city.

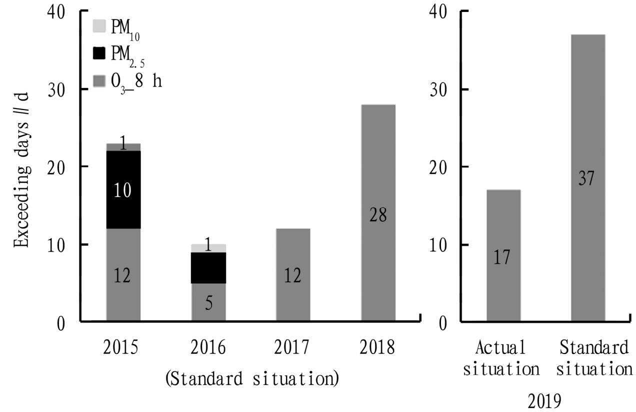


Fig. 1 The proportion of primary pollutant in Shantou City during 2015 – 2019

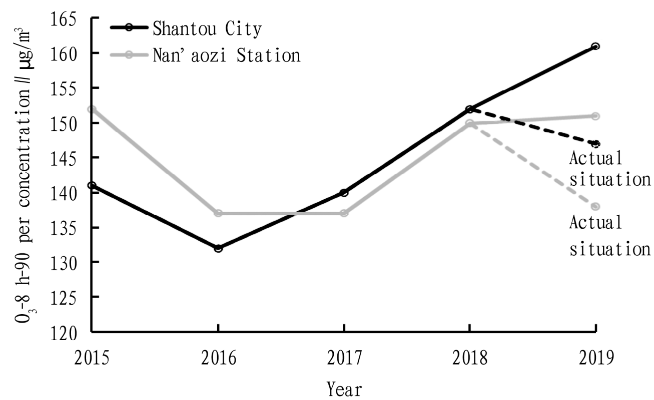
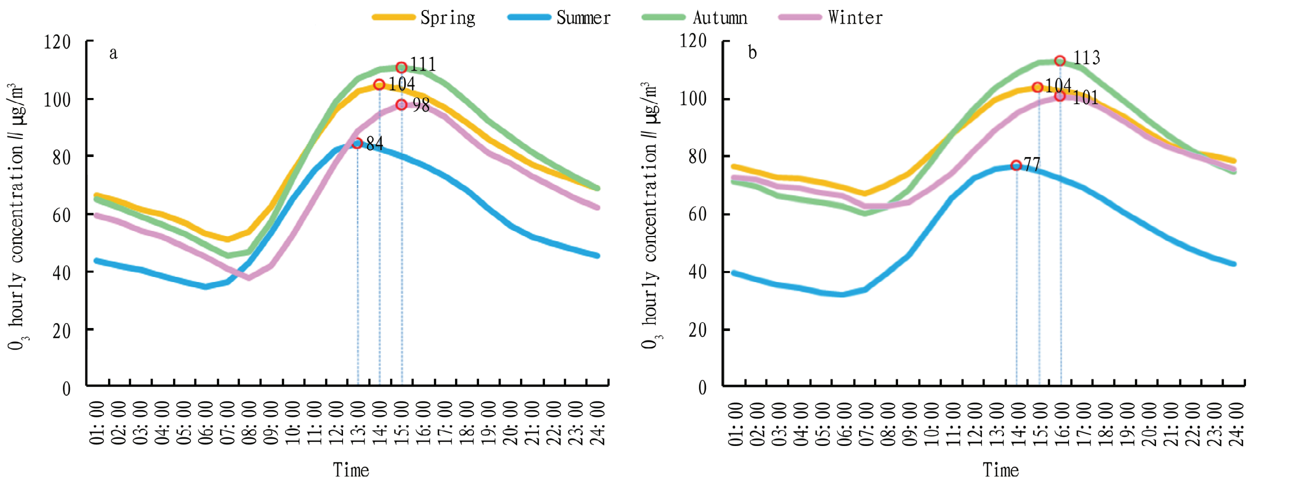


Fig. 2 Annual variation trend of O_3 during 2015 – 2019

2.1.3 Seasonal variation characteristics. Hourly concentration variation of O_3 in Shantou City showed an obvious seasonal difference characteristics. As shown in Fig. 3, it all showed as single-peak type in four seasons. Seen from average concentration of peak, it showed as summer < winter < spring < autumn whether in urban or suburban areas. Average concentration of peak in Shantou urban district in summer was 84 μg/m^3, which was obviously higher than that in Nan'aozi Station. In winter, spring and autumn, average concentration of peak in Shantou urban district was respectively 98, 104 and 111 μg/m^3, which was basically equal to or slightly lower than that in Nan'aozi Station, illustrating that O_3 pollution was serious in spring and autumn. Due to more rainfall in summer, the wet removal of pollutants was obvious. Moreover, southerly wind prevailed, and it was mainly affected by ocean background wind, and overall concentration level of the brought marine clean air mass pollutants was also lower. Compared with suburban district, more O_3 precursors (NO_X and VOCs) were discharged in urban district. Therefore, O_3 generation intensity at daytime could be higher than that in suburban district, making that O_3 peak was higher in Shantou urban district in summer. In winter, spring, and autumn, the northerly or easterly wind was dominant in Shantou City, and it was easy to be affected by air mass from north inland and coast. Therefore, it might bring O_3 from the north or the sea by long-distance transmission. Superimposing O_3 generated by local photochemical reaction, O_3 concentration in Shantou City was risen.

By the influence of temperature and sunlight, occurrence time of O_3 peak in Shantou City in each season was different somewhat. Occurrence time of peak was as below: 13:00 in summer, 14:00 in spring, and 15:00 in autumn and winter. Occurrence time of peak in Nan'aozi Station in each season delayed by 1 h than that in Shantou urban district.



Note: a. Shantou urban district; b. Nan'aozi Station.

Fig. 3 Seasonal variation characteristics of O_3 hourly mean

2.1.4 Monthly variation characteristics. Seen from Fig. 4, monthly average concentration distribution of O_3-8 h-90 per in Shantou urban district had significantly monthly variation characteristics, showing "M shape". There were double peaks in April and October, and the peak was the highest in October, while the lowest concentration appeared in June. Monthly variation characteristics of Shantou was consistent with Zhuhai[6]. Monthly average concentration of O_3 in south cities of the Pearl River Delta (Jiangmen, Shenzhen, Zhongshan and Zhuhai) was the lowest in June, and the variation characteristics may be related to that South China Sea monsoon in southwest direction was prevail in the Pearl River Delta in summer[6]. It was worth noting that the O_3 concentration was 165 μg/m^3 in June of 2018, which was the highest in recent years, and it exceeded by 5 μg/m^3 than limit of two-level concentration standard. It was clear that although overall concentration of O_3 in summer was lower, prevention and control of O_3 pollution should not be ignored under the sunny, hot, calm, stable and unfavorable-diffusion weather conditions by the influence of downdraft around typhoon. Additionally, exceeding days of O_3 also showed a similar variation characteristics. There was no O_3 exceeding in January, February, November and December, and exceeding days in April and October was the most, and O_3 pollution was the most prominent in October.

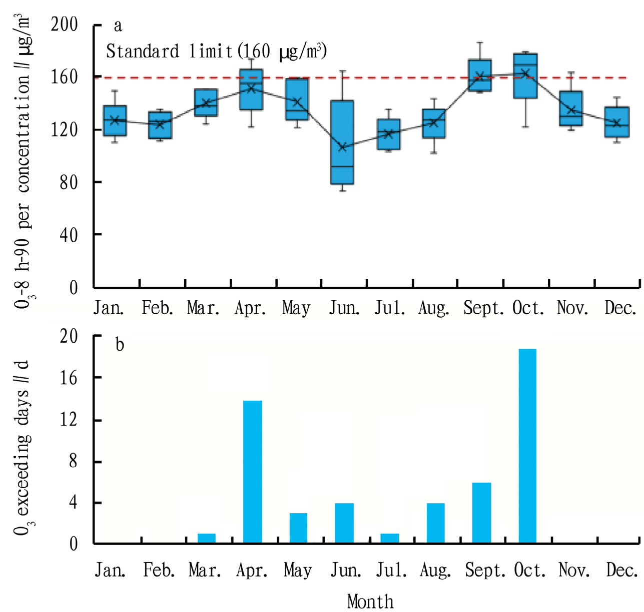


Fig. 4 Monthly variation trends of O_3-8 h-90 per concentration (a) and exceeding days (b) in urban district

2.1.5 Diurnal variation characteristics. Average diurnal variation of O_3 in Shantou urban district and Nan'aozi Station was shown in Fig. 5. The results showed that diurnal variation of O_3 hourly concentration in Shantou City overall showed the characteristics of "single-peak type". As night came, the light faded, and O_3 hourly concentration gradually declined, and it declined to the lowest at 08:00. As the solar radiation increased after sunrise, the photochemical reaction rate increased, and O_3 hourly concentration gradually rose, and it reached the peak at 14:00. After that, photochemical reaction weakened with the sunlight gradually weakened, and O_3 generation rate declined, as well as O_3 hourly concentration. The diurnal variation characteristics of O_3 in Shantou City were consistent with those in 9 cities of the Pearl River Delta[6], 17 prefecture-level cities of Henan Province[7], Beijing[8], and Taiyuan[9]. In general, there is no direct emission source of O_3 near the surface, which is a secondary air pollutant[10]. It is mainly from formation of photochemical reaction and transport of stratospheric O_3 to troposphere, and the latter contributes less than the former[11]. Therefore, the diurnal photochemical reaction of O_3 and precursors is the basis of its concentration variation. The diurnal variation rule is commonly formed by photochemical reaction intensity of O_3 precursors and atmospheric physical process under meteorological condition. Hourly mean of O_3 in Shantou urban district (101 μg/m^3) was slightly higher than that in Nan'aozi Station (98 μg/m^3). Before photochemical reaction started, O_3 in Shantou urban district and Nan'aozi Station both exceeded 40 μg/m^3, and the contribution rate to peak concentration reached 40.8%, which was larger. Seen from hourly extreme high value, they all exceeded limit of grade-two standard concentration (200 μg/m^3) in Shantou urban district and Nan'aozi Station at daytime and night. Occurrence frequency and concentration of extremely high value were higher in Nan'aozi Station, and they were not synchronous with the occurrence time of urban extreme value in urban district, and there should be long-distance transportation.

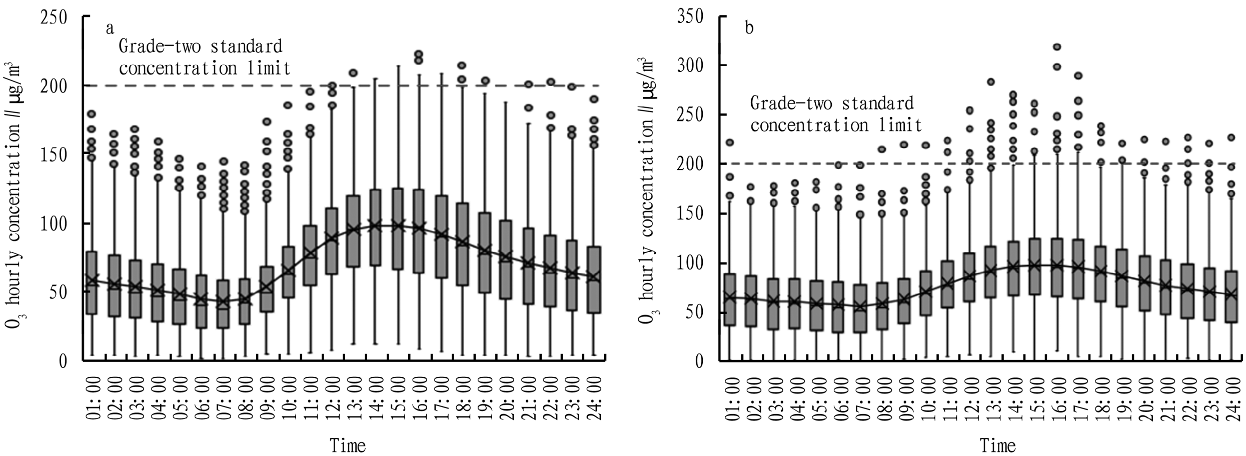


Fig. 5 Diurnal variation of O_3 in Shantou urban district (a) and Nan'aozi Station (b)

2.1.6 Diurnal variation characteristics of pollution days. According to variation characteristics of O_3 concentration, diurnal variation of O_3 in pollution days in Shantou City was divided into below types: O_3 single-peak type, O_3 double-peak type and O_3 no obvious main peak type. O_3 single-peak type was also divided into below types: the peak type and wide peak type, as well as the special initial high concentration type according to duration of the peak; noon type (peak appeared during 12:00 – 14:00), afternoon type (peak appeared during 15:00 – 17:00), and evening type (peak appeared during 18:00 – 20:00) according to occurrence time of peak.

By the influence of meteorological conditions and precursors' concentration, diurnal variation characteristics of O_3 were various, and different diurnal variation characteristics also appeared in one pollution process lasting for several days. Fig. 6 was main peak type of pollution days in Shantou: diurnal variation of afternoon wide peak type. The peak of O_3 concentration appeared once during 15:00 – 17:00, and duration of peak concentration was longer, and O_3 concentration declined slowly.

By the influence of transmission and residue, the high concentration at night was an important feature of the diurnal variation of O_3 in pollution days in Shantou City, and representative curve was shown in Fig. 7. The data of O_3 pollution days over the years showed that the days of O_3 concentration still maintaining more than 100 μg/m^3 after 22:00 accounted for 90.4% of O_3 pollution days in statistical years, while the days of O_3 concentration still maintaining more than 150 μg/m^3 also reached 44.2%. NO_x ($NO + NO_2$) has titration effect on O_3, and NO_x in lower concentration is often accompanied by higher concentration of O_3[12]. When titration effect of NO_x is not enough to consume excessive O_3 remained at night, O_3 in high concentration could last until the next morning. Seen from diurnal variation characteristics of O_3 average hourly value in Shantou City in recent years, the peak of O_3 concentration in Shantou City was greatly affected by O_3 initial concentration before photochemical reaction activity (07:00), and its average contribution rate to peak concentration was 40.8%. The days of O_3 diurnal variation characteristics of initial high concentration type accounted for 21.2% of O_3 pollution days. It was clear that long-term prevention and control, regional joint prevention and control, and emergency prevention and control measures were indispensable in Shantou City.

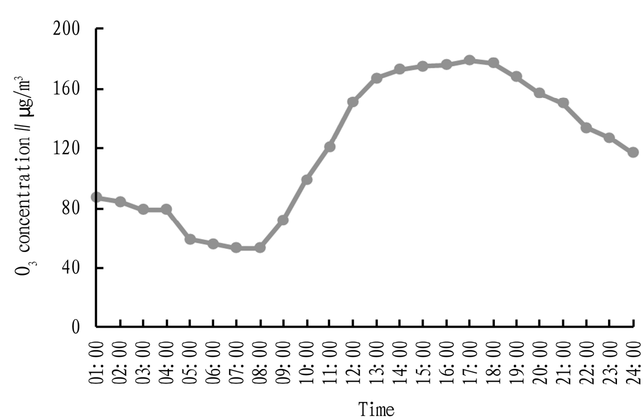
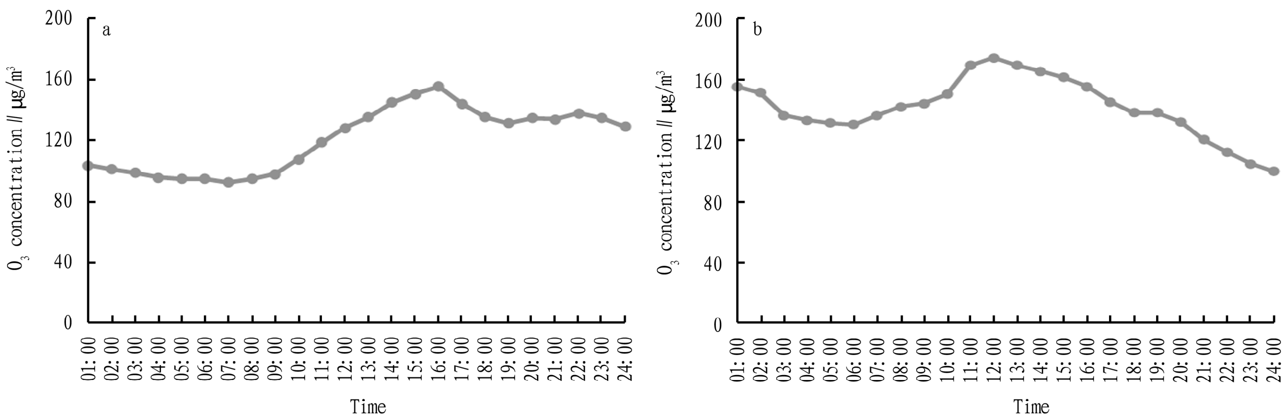


Fig. 6 Diurnal variation of O_3 afternoon wide peak type on October 27, 2017

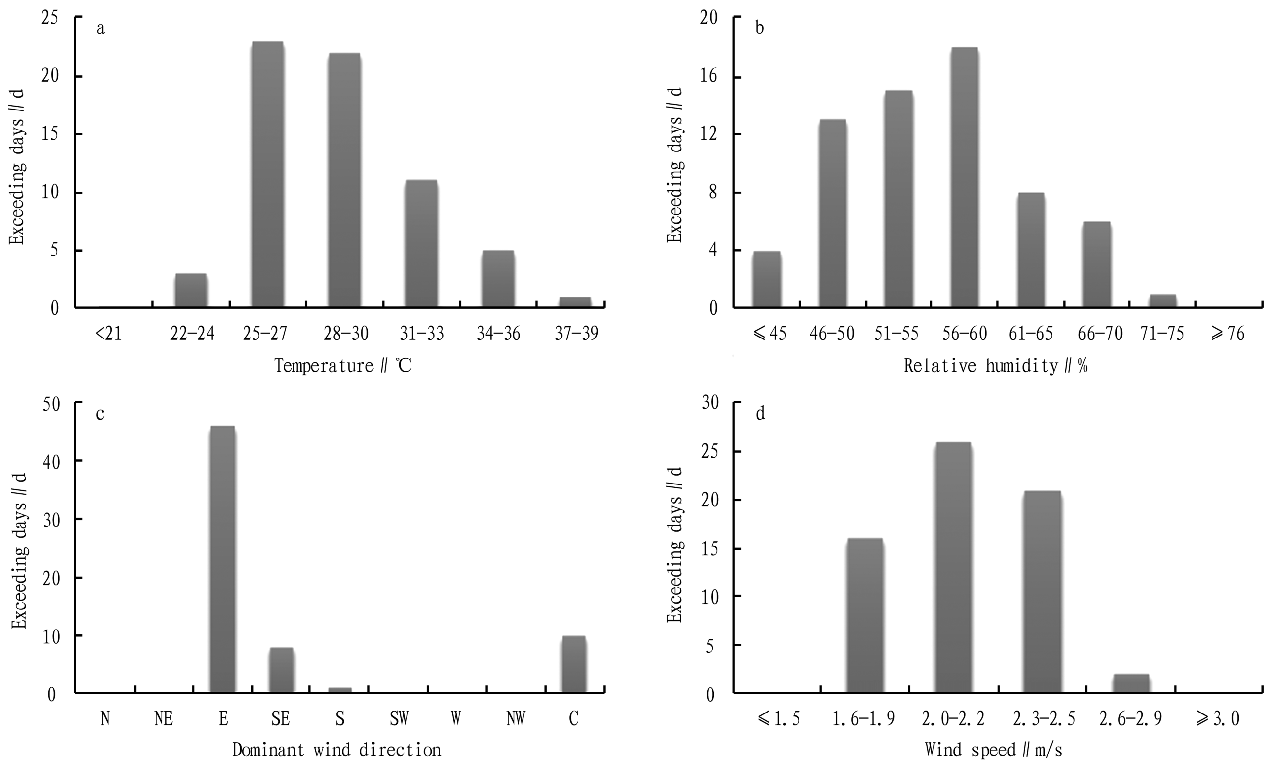
2.2 Meteorological conditions of O_3 pollution days Meteorological condition is one of the important factors affecting local air quality[13–14], and plays an important role in the formation of O_3.

It could affect O_3 concentration in the atmosphere by affecting atmospheric circulation, photochemical environment, and diffusion of O_3 precursors[9, 15]. Shantou City is in the border of Guangdong Province, Fujian Province and Taiwan, with obvious regional pollution characteristics, and pollution occurrence is closely related to weather situation in the period. Temperature, relative humidity, dominant wind direction and average wind speed in Shantou City at daytime (08:00 – 18:00) during 2015 – 2019 were counted, and the results were shown in Fig. 8.



Note: a. April 28, 2018; b. April 29, 2018.

Fig. 7 Diurnal variation of O_3 initial high concentration type in pollution days



Note: a. Temperature; b. Relative humidity; c. Dominant wind direction; d. Average wind speed. C showed that four or more of the six national control stations had different dominant wind directions on that day.

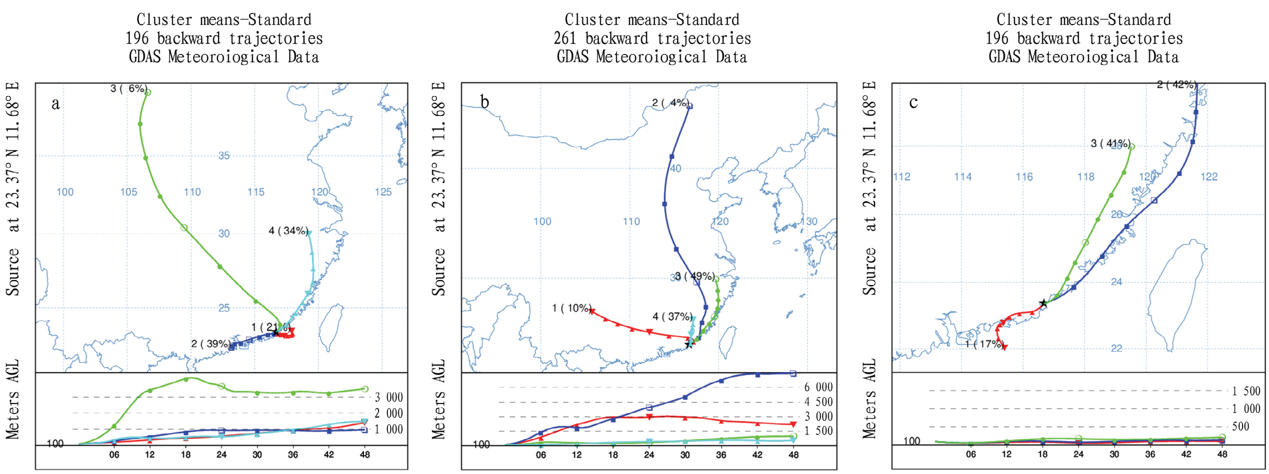
Fig. 8 The relationship between O_3 pollution distribution and meteorological parameters near the ground

Solar radiation is one of the important factors affecting the formation of O_3. There is lack of solar radiation data, but temperature variation could better reflect the variation of solar radiation intensity[10]. Seen from Fig. 8a, exceeding days in Shantou City did not increase with temperature rise, but concentrated between 25 and 30 ℃. It illustrated that temperature was an important but not the only meteorological factor affecting O_3 concentration, which may be related to geographical location characteristics of Shantou City. High temperature mostly appeared in summer, and it was mainly affected by ocean background wind and was accompanied by strong rainfall. Average wind speed was also larger, which affected O_3 concentration.

Exceeding days of O_3 in Shantou City concentrated in relative humidity lower than 75% (Fig. 8b). When relative humidity is higher, water vapor in the atmosphere will affect the role of ultraviolet radiation in photochemical reactions, thereby weakening photochemical reactions in the atmosphere, and it was not favorable for the generation of O_3[16].

The main influence of wind speed on O_3 pollution is migration and diffusion, and wind could transport O_3 from the area with serious pollution to downwind. When wind speed enhances significantly, the horizontal transport will dilute the concentration of O_3 and its precursors[17]. Meanwhile, wind direction also has important influence on concentration of O_3 and its precursors. Seen from meteorological data in Shantou City over the years, O_3 exceeding situation mostly occurred in that dominant wind was east – south wind radian or there was no obvious dominant wind direction (Fig. 8c), and wind speed was between 1.6 and 2.9 m/s (Fig. 8d), dominant by level-two light wind.

2.3 Transmission trajectory characteristics of polluted air mass High concentration days of O_3 during 2016 – 2018 (O_3 mean in 3 hours or above exceeded 160 μg/m^3 at observation site) were selected, and 48-h backward trajectory at O_3 exceeding time was drawn and clustered, and pollution transmission route of high-concentration O_3 days was distinguished (Fig. 9). In O_3 pollution days, the polluted air mass in Shantou City was mainly from northeast direction (northeast coast accounted for 38%, and northeast inland accounted for 46%), northwest inland (5%) and southwest coast (11%), and it was mainly introduced from east – northeast of Shantou City. In 2016, it originated from northeast coastal provinces, and trajectory proportion of the polluted air mass introduced into Shantou City through Zhejiang and Fujian was 55%. In 2017 and 2018, trajectory of the polluted air mass in northeast coast increased to more than 80%. It was clear that transmission trajectory of O_3 polluted air mass was mainly coastal transmission of northeast.



Note: a. 2016; b. 2017; c. 2018.

Fig. 9 Hourly backward trajectory clustering of high-concentration O_3 during 2016 – 2018

3 Conclusions

(1) At present, the main atmospheric environmental problem faced by Shantou City is O_3 pollution, and O_3 pollution had the development trend from offshore island Nao'ao Island to urban district. Monthly variation of O_3 in Shantou showed "M shape", and O_3 pollution all could occur in March – November. Among them, O_3 pollution was the most serious in Shantou City in spring and autumn, and the pollution days was the most in October. Maybe it was because that easterly was dominant in Shantou City in autumn, and it was easy to be affected by air mass from north inland and coast. The brought O_3 from long distance transmission of the north or sea superposed photochemically generated O_3, which commonly improved O_3 concentration in Shantou City.

(2) Average diurnal variation characteristics of O_3 in Shantou urban district and Nan'aozi Station overall showed "single-peak type", and O_3 concentration reached the peak at 14:00. Via comparison of hourly extreme high value, it further illustrated that there existed O_3 long-distance transportation between Shantou urban district and Nan'ao Island.

(3) Diurnal variation of O_3 in the pollution days in Shantou City was afternoon wide peak type, showing as that O_3 concentration declined slowly after the noon. The days that O_3 concentration still maintained more than 150 μg/m^3 after 22:00 accounted for 44.2% of O_3 pollution days in statistical years. By the influence of transmission and residue, O_3 concentration at night was higher, which further affected initial concentration of O_3 before photochemical reaction activity in the second day, and caused the rise of O_3 peak, and it was easy to cause continuous pollution of O_3.

(4) Meteorological conditions of O_3 pollution in Shantou City had the characteristics of high temperature and low humidity. Among them, there was 25 – 30 ℃ of temperature range and 46% – 60% of relative humidity range, and it was accompanied with level-two easterly wind, and O_3 pollution was easy to occur.

(5) Combining backward trajectory analysis of O_3 pollution days, it was verified that transmission trajectory of O_3 pollution air mass in Shantou City was mainly affected by coastal transmis-

sion of northerly polluted air mass, which was introduced from east – northeast.

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(From page 18)

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