Characteristics and Formation Mechanism of Surface Ozone in a Coastal Island of Southeast China: Influence of Sea-land Breezes and Regional Transport

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ABSTRACT

The ozone (O₃) concentrations in island cities are low due to the relatively low concentrations of O₃ precursors. However, Pingtan, a typical island city along the southeast coast of China, suffers from frequent O₃ pollution. In this study, one year of hourly O₃ concentration data collected from three sites (rural, suburban, and urban) on Pingtan were used to investigate the characteristics and formation mechanism of O₃ pollution. The results showed that the minimum O₃ concentrations measured at the three sites were larger than 55.53 μg m⁻³, which was likely caused by low NO titration over Pingtan. The O₃ concentrations in summer were low because of the East Asian monsoon. The daily maximum O₃ concentrations with sea and land breezes (SLBs) were higher than those without SLBs at all three sites (8.52, 9.84, and 14.30 μg m⁻³ at the rural, suburban, and urban sites, respectively). In addition, SLBs amplified the diurnal variation in O₃ concentrations. Cluster analysis and the analysis of an episode of high O₃ concentration indicated that the developed Yangtze River Delta is the main source of O₃ in Pingtan. This study helps reveal the characteristics and formation mechanism of O₃ pollution in island cities.

Keywords: Ozone (O₃); Spatiotemporal variation; Transport; Sea and land breezes; Island.

INTRODUCTION

Surface ozone (O₃) is a key component of photochemical smog and is harmful to public health, vegetation (NRC, 1991), and crop yield (Lal *et al.*, 2017). Strict emission measures enacted in the 1990s have alleviated serious O₃ pollution in many European and American urban areas (Simon *et al.*, 2015; Lin *et al.*, 2017). Meanwhile, rapid urbanization and industrialization have taken place in South and East Asia, resulting in significant increases in the anthropogenic emissions of O₃ precursors (Duncan *et al.*, 2015) and possibly shifting global hot spots of air pollution to these densely populated regions (Zhang *et al.*, 2016). The number of photochemical studies focused on

the mainland of China has increased substantially since 2005, especially in the Beijing-Tianjin-Hebei (BTH) region, the Pearl River Delta (PRD) region, and the Yangtze River Delta (YRD) region (Wang *et al.*, 2017). However, the number of photochemical studies focused on island cities are limited.

Generally, the meteorological conditions of island cities are complex and are easily affected by sea and land breezes (SLBs). SLBs play important roles in O₃ distribution and concentration in island cities (Hsu and Cheng, 2019). For example, Wang *et al.* (2018) demonstrated that O₃ episodes can be affected by SLBs. Ding *et al.* (2004) used MM5, a mesoscale model, to simulate SLBs-related O₃ episode. Using the WRF-CHEM model, Bei *et al.* (2018) found that land breeze transports pollutants from the inland to coastal areas, causing O₃ episodes over the gulf, while sea breeze recirculates the pollutants, aggravating air pollution over the gulf. Studying SLBs is helpful to understand the interactions between continental and marine atmospheres. To the best of our knowledge, few studies have investigated the effects of SLBs on islands of southeast China.

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Therefore, the effects of SLBs on O₃ concentration in subtropical monsoon climates are not clear, severely limiting our understanding of the interactions between continental and oceanic air masses. An island with minimal contributions from local emissions is ideal for studying the effect of regional transport on O₃ pollution.

Pingtán, a southeast coastal island of China, has low levels of primary pollutant emissions but high O₃ concentrations. Pingtán has a population of only 0.42 million, a land area of 371.91 km², and a sea area of 6,064 km². Despite having no major industries, the annual average O₃ concentration in Pingtán is 1.5 to 2.5 times higher than those of other cities in Fujian Province (Wang *et al.*, 2018). Wind speed in this region is relatively high throughout the year because of the “narrow tube effect” in the Taiwan Strait. Excluding typhoon weather, the maximum wind speed in Pingtán is 30–40 m s⁻¹ at 10 m above ground. Since the influence of local pollutants in Pingtán is minimal, Pingtán is easily affected by long-distance transport. Comparing field studies on the atmospheric environment in regions with low primary pollutant emissions with those in regions with high primary pollutant emissions can provide important information about the effects of human activities and improve our understanding of O₃ pollution in those regions.

Given the limited number of studies on O₃ levels conducted on islands along with the urban focus of existing studies, O₃ levels on Pingtán were measured at three sites (rural, suburban, and urban) to better represent the geographical

features of O₃. First, the spatiotemporal variations in O₃ levels were evaluated based on field measurements. Second, the effects of SLBs on O₃ concentration and its diurnal variations were explored. Finally, the effects of regional transport on O₃ pollution in Pingtán were discussed.

METHODOLOGY

Study Area

Pingtán, which is located in Southeast China and has a subtropical climate, is one of the five largest islands in China. The annual average precipitation and temperature are 1196.2 mm and 19.5°C, respectively. As shown on the left side of Fig. 1, Pingtán is located in the Taiwan Strait. Due to the “narrow tube effect” of the Taiwan Strait, the annual average wind speed of Pingtán is 8 m s⁻¹ at 10 m above ground. Therefore, Pingtán is easily affected by regional transport. Pingtán is also affected by SLBs, as indicated by data collected by the Pingtán Ocean Observation Station (POOS). Thus, we choose Pingtán as a site to study the effects of regional transport and SLBs on O₃ concentration. Unlike the Pingtán Weather Station, POOS is located on the eastern coastline of Pingtán, which is less affected by topography and adverse environmental factors. The location of POOS is indicated by the star symbol on the right side of Fig. 1. Based on the O₃ data available for January to December 2015, the following three air quality monitoring stations in different areas of Pingtán were

Fig. 1. Location of Pingtán in China (left) and locations of the three monitoring sites and the Pingtán Ocean Observation Station (POOS) in Pingtán (right).

selected for this study: (1) 36-Degree Reservoir (36DR), a rural site, is situated in the center of Pingtan and is surrounded by forests and lakes; (2) Jinjing Bay (JJB), a suburban site, is located in the southwestern region of Pingtan and is a developing area with considerable ongoing construction; and (3) Government of County (GC), an urban site, is located in downtown Pingtan and is surrounded by the main road, which has a high volume of motor vehicles. The locations of the monitoring sites (round symbols) are also presented on the right side of Fig. 1.

Collection and Analysis of O₃ Data and Other Parameters

The air pollution data recorded in Pingtan from January to December 2015 [local time (UTC+8)] included hourly average concentrations of O₃, NO_x, CO, SO₂, PM₁₀, and PM_{2.5}. Meteorological parameters were also collected from each site.

The surface O₃ concentration was measured using an ultraviolet spectrophotometric ozone analyzer (Thermo Model 49i). The concentrations of NO₂ and NO were measured using a NO-NO₂-NO_x analyzer based on chemiluminescence (Thermo Model 42i). The CO concentration was measured based on the absorption of infrared radiation (Thermo Model 48i). The SO₂ concentration was determined by a pulse fluorescence SO₂ analyzer (Thermo Model 43i). These instruments were automatically calibrated on a daily basis using a dynamic gas calibrator (TE 146C). The PM_{2.5} (SHARP 5030) and PM₁₀ (FH62C14) concentrations were measured using β -ray methods. Meteorological parameters were determined continuously by microwave induction measurements using a Lufft WS500-UMB Compact Weather Station.

The eight-hour maximum concentrations of O₃ on mainland China from October 10–17, 2015, were downloaded from datacenter.mep.gov.cn/ (in Chinese). The data used to calculate the trajectories from January to December 2015 were downloaded from the Air Resources Laboratory (ARL) of the National Oceanic and Atmospheric Administration (NOAA) (<ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1>).

Analysis of Trajectories

To identify the sources of air parcels reaching the monitoring sites, the atmospheric trajectory was studied through backward trajectory analysis. The three-day backward trajectories were computed at an elevation of 500 m above the center of Pingtan (119.78°E, 25.51°N) from 00:00 to 23:00 (UTC) throughout 2015. This 72-h time scale is appropriate for capturing the long-distance transport of air pollutants because the majority of pollutants fall within a few days. The elevation of 500 m ensures that the atmospheric boundary layer is the origin of the backward trajectories (Latif *et al.*, 2012). The Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT, version 4), which was developed by the NOAA ARL (Rolph, 2003), was used to calculate the backward trajectories. To compare the origins of air masses between different months, the clustering method was applied to cluster the obtained trajectories according to transport characteristics, length, and curvature. Data for the entire year were used to

compare the O₃ concentrations of different clustering trajectories. The HYSPLIT model and Ward method were used for cluster analysis (Romesburg, 1984). The algorithm minimizes the total spatial variance (TSV), where the spatial variance of the cluster is the sum of the squared distances between the endpoints of the trajectory (Stunder, 1996). The TSV plot is applied to determine the cut-off point of the algorithm and the optimal cluster number based on the detection of a sharp increase in the TSV plot. A sharp increase in TSV indicates the combination various clusters. Thus, the clustering algorithm should be ended at this moment (Eva and Lambin, 1998).

Potential Source Contribution Function (PSCF) Analysis

PSCF analysis is a statistical method to assess the contributions of certain air masses to observed pollutant concentrations that exceed a given threshold (Hopke *et al.*, 1995). In this study, PSCF values were computed to distinguish potential source zones contributing to the high concentrations of O₃ and CO in Pingtan. The study area was split into $i \times j$ small cells of equal size. The PSCF values were normalized using the following equation:

$$PSCF_{ij} = n_{ij}/m_{ij}, \quad (1)$$

where n_{ij} is the number of endpoints included in the ij^{th} cell with pollutant concentration above a given threshold, and m_{ij} is the total number of endpoints for the ij^{th} cell. Cells with high PSCF values are associated with air masses in which a pollutant exceeds the threshold value. This suggests that the corresponding regions are potential contributors to receptor contamination.

A total of 5,184 72-h backward trajectory endpoints (3 d $\times$ 72 h $\times$ 24 h) were used in this study. The PSCFs were calculated within a defined area (25°N, 80°E to 58°N, 130°E) comprising 6,600 grid cells with latitude/longitude dimensions of $0.5^\circ \times 0.5^\circ$. Thus, the mean number of endpoints per cell (n_{ave}) was $5,184/6,600 \cong 1$. The average concentration of each pollutant from October 12–14, 2015 was evaluated with respect to the threshold criteria. The PSCF values were multiplied by an arbitrary weighting function W_{ij} (Polissar *et al.*, 1999) to reduce uncertainty in cells with $n_{ij} \leq 3n_{ave}$. W_{ij} was determined as follows:

$$W_{ij} = \begin{cases} 1.00 & n_{ij} > 3n_{ave} \\ 0.70 & 1.5n_{ave} < n_{ij} \leq 3n_{ave} \\ 0.42 & n_{ave} < n_{ij} \leq 1.5n_{ave} \\ 0.17 & n_{ij} \leq n_{ave} \end{cases}. \quad (2)$$

RESULTS AND DISCUSSION

General Characteristics of O₃ Concentration

The O₃ concentrations in Pingtan were 67.5, 66.2, and 79.2 $\mu\text{g m}^{-3}$ at the urban, suburban, and rural sites, respectively (Table 1). Compared to the concentrations of O₃ at the other two sites, the O₃ concentration at the rural site was significantly higher, likely due to the lack of NO

Table 1. Comparison of annual average O₃ concentrations determined in this study with those reported elsewhere.

Location	Site type	Time	O ₃ (μg m ⁻³)	Reference
Anantapur, India	Rural	Jan.–Dec. 2010	87.2	(Reddy <i>et al.</i> , 2012)
New Jersey, USA	Urban	Jun. 2007–May 2008	46.9	(Roberts-Semple <i>et al.</i> , 2012)
Andalusia, Spain	Urban (traffic)	2003–2008	51.0	(Adame <i>et al.</i> , 2014)
	Urban background	2003–2008	55.9	
	Suburban background	2003–2008	66.0	
	Rural background	2003–2008	72.0	
	Urban background	2013–2014	68.1	
Delhi, India	Urban (traffic)	2013–2014	62.4	(Kumar <i>et al.</i> , 2014)
	Rural	2013–2014	84.4	
	Urban	2013	42.2	
Seoul, Korea	Urban	2013	42.2	(Vellingiri <i>et al.</i> , 2016)
Shanghai, China	Urban	2015	55.5	(Gao <i>et al.</i> , 2017)
Guangzhou, China	Urban	2007	38.6	(Zheng <i>et al.</i> , 2010)
	Suburban	2007	57.9	
Huizhou, China	Rural	2007	81.4	
Hongkong, China	coastal	2007	47.1	
Nanjing, China	Urban	Jan. 2000–Feb. 2003	43.7	(Tu <i>et al.</i> , 2007)
Ningbo, China	Urban	2012–2015	48.6	(Tong <i>et al.</i> , 2017)
	Suburban		77.4	
	Rural		80.8	
	Rural		91.71	
Zhoushan, China	Rural	Apr.–Oct. 2015	91.71	(Tong <i>et al.</i> , 2018)
Pingtan, China	Urban	Jan.–Dec. 2015	67.5	Present study
	Suburban	Jan.–Dec. 2015	66.2	
	Rural	Jan.–Dec. 2015	79.2	

If concentration in other articles was reported in ppb, it was converted into μg m⁻³ using the following formula:
concentration (μg m⁻³) = concentration (ppbv) × 48/22.4.

titration at the rural site (Kiros *et al.*, 2017; Kulkarni *et al.*, 2017). The annual mean O₃ concentration in Pingtan was compared to the concentrations reported for other cities (Table 1). The O₃ concentration at the urban site on Pingtan island (67.5 μg m⁻³) was similar to the urban background concentration reported in Delhi (68.1 μg m⁻³) but obviously higher than those observed in the urban areas of New Jersey (46.9 μg m⁻³), Andalusia (51.0 μg m⁻³), Shanghai (55.5 μg m⁻³), Guangzhou (38.6 μg m⁻³), Nanjing (43.7 μg m⁻³), Ningbo (48.6 μg m⁻³), and Seoul (42.2 μg m⁻³) along with Andalusia (55.9 μg m⁻³). The O₃ at these urban sites with high traffic density might be consumed by NO titration from traffic emissions. The concentration of O₃ at the suburban site on Pingtan island (66.2 μg m⁻³) was nearly equal to that of the suburban background of Andalusia (66.0 μg m⁻³), approximately 8.3 μg m⁻³ higher than that reported for Guangzhou, and approximately 11.2 μg m⁻³ lower than that reported for Ningbo. The O₃ concentration at the rural site of Pingtan (79.2 μg m⁻³) was similar to those reported for rural sites in other locations (Zheng *et al.*, 2010; Reddy *et al.*, 2012; Kumar *et al.*, 2014; Tong *et al.*, 2017). However, the O₃ concentration of the rural site of Zhoushan (91.71 μg m⁻³), which has few commercial and residential buildings nearby and similar climatic conditions to Pingtan, was much higher than that of the rural site of Pingtan (Tong *et al.*, 2018). This can be attributed to the fact that Zhoushan is located in the YRD and is easily affected by the YRD. However, Pingtan is affected by YRD in the condition of regional transport. Overall, the O₃ concentration at the urban site of Pingtan, an island in

Southeast China with few anthropogenic activities, was higher than those reported for metropolitan areas.

Diurnal variations in pollutant concentrations provide insights into the interactions between emissions and daily chemical/physical processes. Fig. 2(a) presents the mean annual variations in diurnal O₃ concentration. O₃ concentration began to increase after 08:00 and peaked at 15:00–16:00 at all three monitoring sites. This trend was opposite that observed for NO₂ in the daytime (Fig. S1). This phenomenon suggests that NO₂ was mainly transformed into O₃ in the daytime (Khoder, 2009; Hassan *et al.*, 2013). The maximum O₃ concentrations at the rural, suburban, and urban sites were 93.29, 79.09, and 77.18 μg m⁻³, respectively. The O₃ concentration at the rural site was obviously higher than those at the urban and suburban sites, and the diurnal O₃ maximum appeared one hour later at rural site compared to at the other two sites. Among the three sites, the O₃ concentration was highest at the rural site, which can be explained as follows. The air masses transported from the urban site, upwind of the rural site, had high concentrations of O₃ precursors. These air masses led to elevated levels of O₃ over the rural site, which was characterized by low NO titration (Banan *et al.*, 2013). The rural site is located in an area that is protected for drinking water, and anthropogenic sources of pollution such as vehicle exhaust and industrial emission are few. Therefore, the NO concentration was low at the rural site (0.51 μg m⁻³). The delayed time of peak O₃ concentration at the rural site compared with the other two sites indicates that the photochemically aged plume is a key source of O₃ at the rural site (Xu *et al.*, 2011). The minimum

Fig. 2. (a) Diurnal variations in O₃ concentration and (b) monthly variation in O₃ concentration for the three monitoring sites.

O₃ concentrations measured at the rural (65.60 μg m⁻³), suburban (55.77 μg m⁻³), and urban sites (55.53 μg m⁻³) were higher than those reported in other cities, which was likely caused by low NO titration over the three sites in this study. The NO concentrations for the rural and the urban sites were only 0.51 and 1.63 μg m⁻³, respectively. Unfortunately, the NO data for the suburban site were invalid.

The O₃ concentrations in Pingtan were higher in spring (March, April, and May) and early autumn (September and October) compared to in summer (June, July, and August; Fig. 2(c)). This seasonal trend is widespread at mid-latitudes in the northern hemisphere (Monks, 2000). Notably, the seasonal variations in O₃ concentration in Pingtan are consistent with those reported for PRD, Taipei, and Japan (Chou *et al.*, 2006), which are located in the East Asian monsoon region. The maximum concentration of O₃ was observed in the spring due to the transport of air masses over long distances from contaminated areas on the continent (Gong *et al.*, 2018). The minimum O₃ concentration was observed in summer, which is attributed to the transport of air masses with relatively low O₃ concentrations from the ocean (Pochanart *et al.*, 1999; Chou *et al.*, 2006; Latif *et al.*, 2012). Fig. S2 shows the monthly mean 72-h backward trajectory cluster along with the contribution of each trajectory cluster to the total trajectory cluster. All clusters in June, July, and August are from the ocean, which might explain why the O₃ concentration was low in the summer. The relatively rainy weather in the summer might also contribute to this phenomenon by restricting O₃ formation, which requires solar radiation, and accelerating O₃ removal via wet deposition (Jo and Park, 2005; Tong *et al.*, 2017). The precipitation in summer was significantly higher than in the other three seasons; the precipitation amounts in spring, summer, autumn, and winter were 82.53, 176.90, 88.86, and 75.93 mm, respectively.

Effect of SLBs on O₃

SLBs is a mesoscale local circulation system caused by

the difference of thermal power between land and sea. SLBs are key area of research related to the atmospheric boundary layer in coastal regions. The mesoscale circulation caused by SLBs is directly related to the diffusion and transport of atmospheric pollutants in coastal cities (Wagner *et al.*, 2012). Therefore, SLBs play important roles in the distribution of O₃ concentration in island cities (Wang *et al.*, 2018). In this study, SLBs were considered to be present when the absolute value of variation in wind direction between approximately 02:00 and 14:00 ranged from 90° to 270°, and both sea breeze and land breeze lasted for longer than 3 h. The sea breeze always blows in the daytime, while land breeze occurs in the evening. SLBs are driven by temperature differences between land and sea along with topographic conditions, and SLBs usually occurs when the winds are weak (Lu *et al.*, 2009; Wang *et al.*, 2018). As shown in Table 2, SLBs were recorded on 44 days in Pingtan in 2015, including two days of pollution. The month with the maximum number of days with SLBs (eight days) was March followed by May (six days), January, February, April, August, and November (four days each), July and October (three days), September (two days), and June and December (one day). The daily mean wind speed fluctuated between 1.2 and 7.4 m s⁻¹. As shown in Fig. 3, the maximum daily O₃ concentration was always higher with SLBs than without SLBs at the three sites (8.52, 9.84, and 14.30 μg m⁻³ for the rural, suburban, and urban sites, respectively). When SLBs occurred, the wind speed was lower than when SLBs were absent; this lower wind speed is conducive to the accumulation of O₃, resulting in a higher O₃ concentration. The analysis of observational data combined with simulations using the WRF-CHEM model indicated that SLBs promoted the accumulation of O₃, resulting in the deterioration of air quality (Bei *et al.*, 2018). Hsu and Cheng (2019) applied cluster analysis also finding that SLBs were favorable to O₃ accumulation.

The diurnal variations in O₃ concentration with and without SLBs were inconsistent (Fig. 4). On days without SLBs, the differences between the minimum and maximum

Table 2. Number of days with SLBs each month.

Number of days	Month	Day	Nighttime wind direction	Daytime wind direction	Daily average wind speed (00–23 hour) (m s^{-1})	Time when the land breeze turns to sea breeze	Time when the sea breeze turns to land breeze
4	1	4	NNW	SE	2.5	10 ^h	19 ^h
		1	NW	E	1.2	12 ^h	22 ^h
		1	NW	E	2.1	11 ^h	21 ^h
		1	SSW	ENE	2.3	11 ^h	22 ^h
4	2	11–14	SW	E	1.8	12 ^h	22 ^h
8	3	3	W	E	5.3	13 ^h	00 ^h
		3	W	NE	7.4	06 ^h	–
		3	14–15	NW	3.3	04 ^h	22 ^h
		3	17	N	2.5	11 ^h	00 ^h
	3	19	WSW	NNE	6.6	11 ^h	–
		3	NNW	ESE	3.6	09 ^h	–
		3	NNW	ESE	2.6	08 ^h	19 ^h
		4	WSW	SSE	3.4	05 ^h	20 ^h
4	4	6	WSW	NNE	6.8	04 ^h	–
		13	SW	ENE	3.3	10 ^h	19 ^h
		27	N	ESE	3.4	09 ^h	22 ^h
		27	N	ESE	3.4	09 ^h	22 ^h
6	5	3	SW	E	3.3	12 ^h	–
		9	SW	NE	4.7	14 ^h	00 ^h
		11	N	ESE	3.7	09 ^h	20 ^h
		12	WNW	E	5.0	09 ^h	21 ^h
	5	13	NNW	SSE	2.7	08 ^h	22 ^h
		31	WSW	NE	4.7	09 ^h	03 ^h
		31	WSW	NE	4.7	09 ^h	03 ^h
		31	WSW	NE	4.7	09 ^h	03 ^h
1	6	1	N	SSE	3.4	08 ^h	19 ^h
3	7	15	N	ESE	2.6	07 ^h	23 ^h
		16	N	ESE	3.3	07 ^h	19 ^h
		25	SW	ENE	4.9	13 ^h	22 ^h
		25	SW	ENE	4.9	13 ^h	22 ^h
4	8	5	N	ESE	2.8	09 ^h	20 ^h
		13	WSW	E	4.4	08 ^h	–
		17	SW	ESE	3.2	10 ^h	19 ^h
		25	NNW	SE	2.8	04 ^h	01 ^h
2	9	2	WSW	SE	2.0	13 ^h	23 ^h
		24	WNW	NE	5.3	11 ^h	23 ^h
		24	WNW	NE	5.3	11 ^h	23 ^h
		24	WNW	NE	5.3	11 ^h	23 ^h
3	10	8	NNW	SE	3.3	12 ^h	20 ^h
		27	WNW	SE	3.5	10 ^h	05 ^h
		30	SW	NNE	7.1	11 ^h	–
		30	SW	NNE	7.1	11 ^h	–
4	11	7	N	ESE	3.0	12 ^h	20 ^h
		8	NNW	SE	1.8	08 ^h	00 ^h
		13	N	SSE	3.4	10 ^h	23 ^h
		17	NW	WSW	1.9	10 ^h	21 ^h
1	12	23	WSW	E	3.3	11 ^h	19 ^h

Note: “h” in Table 2 means hour.

O₃ concentrations were 24.59, 20.77, and 20.63 $\mu\text{g m}^{-3}$ at the rural, suburban, and urban sites, respectively. In contrast, larger differences between the minimum and maximum O₃ concentrations were observed on days with SLBs (47.50, 28.71, and 47.47 $\mu\text{g m}^{-3}$ at the rural, suburban, and urban sites, respectively). Sea breeze, which always occurs after sunrise, brings cold and moist air from over the ocean into contact with hot and dry air over land. This results in a thermal interior boundary layer over coastal land and produces an inversion near the convergence line between sea and land breezes. Atmospheric stratification is relatively stable due to the inversion, resulting in weakened convective

motion; these conditions are not conducive to O₃ diffusion (Stauffer *et al.*, 2015). In addition, sea breeze carries polluted air masses over the sea, which aggravates the cycle of pollution between the sea and inland (Darby *et al.*, 2007). SLBs form in the absence of large-scale weather systems, which further makes the atmosphere calm. Therefore, in the absence of SLBs, the O₃ concentration was primarily affected by regional transport. In contrast, in the presence of SLBs, local photochemical reactions were dominant in O₃ formation. The suburban Pingtan site experienced high winds throughout the year; thus, SLBs had little influence at this site.

Fig. 3. Differences in O_3 concentration between days with SLBs (solid columns) and days without SLBs (hollow columns) at the three monitoring sites.

Fig. 4. Diurnal variations in O_3 concentration at the three monitoring sites on days with and without SLBs.

Effect of Regional Transport on O_3 Concentration

Wind speed in the study region is relatively high throughout the year because of the “narrow tube effect” in the Taiwan Strait. Since the influence of local pollutants in Pingtan is minimal, Pingtan is easily affected by long-distance

transport. Fig. 5 shows the 72-h backward trajectory clusters along with the percentage contribution of each cluster to annual air masses. The mean O_3 concentration of each trajectory cluster is also given. The trajectories in Pingtan were divided into six categories according to their

Fig. 5. Average backward trajectory clusters with the corresponding O₃ concentrations at Pingtan in 2015.

origin and transport path: a continental mesoscale cluster (Cluster 1) from western China; a mesoscale northeastern marine cluster (Cluster 2) from northeastern China; a continental long-range cluster (Cluster 3) from northwestern China; a mesoscale southwestern marine cluster (Cluster 4) from the South China Sea; a short-range southern marine cluster (Cluster 5) originating from the Taiwan Strait; and a long-distance cluster (Cluster 6) from the North China Plain and passing over the YRD. The mesoscale westerly continental Cluster 1 was the largest contributor, accounting for 27.68% of all air masses, followed by Cluster 5 (24.66%), Cluster 4 (19.61%), Cluster 2 (15.44%), Cluster 3 (7.67%), and Cluster 6 (4.84%).

Cluster 6 had the highest concentration of O₃ (109.71 μg m⁻³), which might be associated with the high levels of primary pollutants in the surrounding industrial and densely populated areas (Cao *et al.*, 2011). Cluster 4 from the South China Sea had the lowest O₃ concentration (56.73 μg m⁻³) because it did not pass over any populated areas. Cluster 5, the short-range southern marine cluster originating from the Taiwan Strait, had the second lowest of O₃ concentration (68.98 μg m⁻³). Cluster 2, which originated from the East China Sea, had the second highest O₃ concentration (91.82 μg m⁻³). The O₃ concentrations of Cluster 3 from northwestern China and Cluster 1 from western China were similar (88.26 and 88.32 μg m⁻³, respectively).

Based on cluster analysis, we obtained six main transport pathways, demonstrating that O₃ concentration was influenced by regional transport, especially from the surrounding industrial and densely populated areas.

Effect of Regional Transport on O₃ Episodes

Cases of regional transport leading to O₃ episodes are discussed in detail in this section. The calendar figure was used to identify instances when the O₃ concentration at Pingtan exceeded the Chinese grade II level in 2015 based on the China GB 3095-2012 criteria. The O₃ concentrations at the rural and suburban sites exceeded the grade II standard from October 12–14, 2015 (Fig. S3). Only the rural and

suburban sites were selected for case study because of the lack of valid data for the urban site during that time period.

Fig. 6 presents the time series of hourly mean O₃ concentration, relative humidity, temperature, and atmospheric pressure at the rural and suburban sites from October 10–17, 2015. From October 12–14, hourly O₃ concentrations exceeding 200 μg m⁻³ were recorded at the rural site, and the concentrations at the rural site were always higher than those at the suburban site. The temperature and atmospheric pressures increased during the episode of increased O₃ concentration (October 12–14), while the relative humidity was low. This indicates that high temperature, high atmospheric pressure, and low relative humidity promote O₃ formation. O₃ concentration was significantly positively correlated with the concentrations of other air pollutants (Table 3). Fig. 7 shows the O₃ concentrations recorded on mainland China during the case study period. Before the episode in Pingtan, the O₃ concentrations in most mainland cities were below the Chinese ambient air quality standard. Most cities on the southeastern coast suffered from light or moderate pollution from October 12–14, 2015. The pollution spread throughout eastern China from October 15–17, 2015. These results indicate that O₃ pollution in Pingtan during this period was seriously affected by regional transport.

To determine the cause of the regional pollution, the weather patterns at the surface and 500 hPa were obtained for October 10–17, 2015. Fig. 8 shows the climatological isolines and wind fields at the surface and at 500 hPa over Pingtan from October 10–17, 2015. A semi-permanent, subtropical anticyclonic high-pressure system over the western North Pacific called the Western Pacific Subtropical High (WPSH) extends from the ground to the middle of the troposphere (Zhou *et al.*, 2009). In 500-hPa weather patterns, the WPSH is usually indicated by an isoline at 5880 gpm (Wang, 2006). In this study, the winds changed from southwesterly to northwesterly at 500 hPa. Approaching the surface, the WPSH shifted toward the northeast, driving a large amount of pollutants over Pingtan. During

Fig. 6. Time series of hourly average O_3 concentration, relatively humidity, temperature, and pressure at the rural and suburban Pingtan sites from October 10–17, 2015.

Table 3. Pearson correlation coefficients for the relationships between the concentrations of O_3 and other pollutants from October 12–14, 2015.

	NO_2	CO	SO_2	PM_{10}	$PM_{2.5}$
Rural	0.370**	0.831**	0.532**	0.882**	0.528**
Suburban	—	0.854**	—	0.854**	0.943**

** : Significant correlation was found at the 0.01 level.

* : Significant correlation was found at the 0.05 level.

— : No valid data were available for correlation analysis.

the episode of high O_3 concentration evaluated in the case study, all of Pingtan was under the control of a strong WPSH, which might have directly led to the abnormal high pressure observed in Fig. 8. The downdraft acted as a dome that trapped pollutants at the surface, and little convection occurred due to the lack of air lift. In addition, as the typhoon approached from October 14–17, the near-surface wind speed over Pingtan gradually increased, mitigating the O_3 pollution. Moreover, as the typhoon approached, the high-pressure system was compelled to disconnect and move southward. Finally, the high-pressure system was removed, and the O_3 pollution was mitigated.

The spatial distributions of potential O_3 sources and transport paths along with those of the precursor CO were determined for Pingtan by combining backward trajectories with the pollutant concentrations (Fig. 9). CO is an important O_3 precursor emitted by anthropogenic activities. Furthermore, CO has a long lifetime (1–2 months) and can serve as an indicator for anthropogenic pollution in the study region (Pochanart *et al.*, 1999). Thus, simultaneous measurements of O_3 and CO provide an opportunity to study

the anthropogenic effects on O_3 . Pochanart *et al.* (1999) continuously measured the O_3 and CO concentrations on the Oki Islands to characterize the variation in O_3 under the influence of the regional-scale anthropogenic emission of air pollutants in the northeast Asian Pacific rim region. Therefore, we evaluated the anthropogenic contribution to O_3 pollution by simultaneously evaluating the PSCFs of O_3 and CO. The potential source locations were similar for CO and O_3 . The potential source regions with high PSCF values for O_3 and CO were primarily distributed in the northwestern region of Pingtan. This indicates that O_3 pollution in Pingtan largely originates from anthropogenic sources located in industrialized areas of the YRD. The industries that emit O_3 precursors, such as NO_x and VOCs, have important effects on the O_3 concentration in Pingtan. Industrial sources of VOCs mainly include VOC production, storage, and transport; processes that use VOCs as raw materials; and the use of products containing VOCs (Liang, 2017). Industrial sources of NO_x mainly include the non-metallic mineral products, steel, and biomedical manufacturing industries (Wu, 2009).

Fig. 7. Plots of the eight-hour maximum values of O_3 concentration on mainland China from October 10–17, 2015.

Fig. 8. Weather patterns at 500 hPa and at the surface for October 10–17, 2015.

Fig. 9. Spatial distributions of PSCF for O₃ and CO at Pingtan from October 10–17, 2015.

CONCLUSION

An island city under the control of the East Asian monsoon that is not strongly affected by anthropogenic sources of pollution was studied to evaluate the spatial and temporal variations in O₃ concentration along with the effects of SLBs and regional transport. The minimum O₃ concentrations measured at the rural ($65.60 \mu\text{g m}^{-3}$), suburban ($55.77 \mu\text{g m}^{-3}$), and urban sites ($55.53 \mu\text{g m}^{-3}$) were higher than those reported in other cities, which was likely caused by low NO titration over Pingtan. The O₃ concentrations at each site in summer, which should be high, were low due to the effect of the East Asian monsoon. SLBs increased the daily maximum O₃ concentration at each site by approximately $10 \mu\text{g m}^{-3}$. The diurnal variation in O₃ concentration was larger when SLBs was present compared to when SLBs was absent. Cluster analysis and the analysis of an episode of high O₃ concentration indicated that O₃ primarily originated from the transport of O₃ precursors from the industrialized YRD region. These findings have implications for O₃ pollution in similar areas.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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