Comparative analysis of the variation characteristics of O₃, NO₂ and CO in the boundary layer in coastal cities and inland cities in the middle latitude regions in China

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Abstract. Based on the hourly monitoring data of O_3 concentration, and its closely related NO_2 and CO concentration in four coastal cities (Qingdao, Yantai, Weihai, Rizhao) and four inland cities (Dezhou, Liaocheng, Heze and Jining) in Shandong Province from 2009 to 2017, we compared and analyzed variations of O_3, NO_2 and CO pollutants in the boundary layer in both coastal and inland cities in mid-latitude regions, and explored their causes. It is found that O_3 pollution showed an aggravating tendency in coastal and inland cities from 2012 to 2017, especially remarkably in inland cities. However, NO_2 and CO pollution showed an opposite tendency. Seasonal variation of O_3 pollution in the boundary layer of coastal cities in the mid-latitude showed an M-shaped bimodal structure, similar to that in low latitude cities, while NO_2 and CO show a V-shaped structure with a maximum in winter and a minimum in summer. The diurnal variation of O_3 concentration is featured with a high concentration in the daytime and a low one in the night, while NO_2 and CO concentration show a bimodal curve with a high concentration both in the morning and evening.

1. Introduction

O_3 is a trace component of the atmosphere and one of the main components of photochemical pollution. O_3 and $PM_{2.5}$ are considered to be the two most important atmospheric pollutants affecting the environmental quality in the current era [1]. O_3 has a strong oxidability, and its existence can affect the oxidability of the atmosphere. Its high concentration near the surface layer may do harm to human health, and affect plant growth, resulting in crop yield reduction [2-11]. With regard to the study on O_3 in the troposphere, long-term continuous observation points were set up in major cities and background regions in North America and Europe quite long ago, and many studies on O_3 and its related precursors and meteorological conditions have been conducted. The results show that the atmospheric O_3 concentration in the troposphere in the northern hemisphere increases at an average rate of approximately 1.0% annually, and at an even faster rate in some industrially developed areas with a dense population [12-15].

In recent years, China has beefed up its control of SO_2 and PM_{10} and other pollutants, and has achieved initial results. However, secondary pollution incidents, such as photochemical smog and

excessive ozone concentration in the boundary layer caused by the development of densely populated cities, are occurring at an increasingly high frequency. The study on the concentration of O_3 in the troposphere started late in China. The study on the concentration of ozone in the boundary layer have been conducted in Beijing, Guangzhou and Shanghai and other major cities since 2000 [16]-[21], focusing on the characteristics of ozone pollution in different cities. Preliminary results show that there are differences to varying degrees between different regions in terms of annual, seasonal and diurnal variations, and there are also large differences between the south and the north, cities and suburbs [22-30]. Ozone in the city is mainly generated by the reaction under suitable meteorological conditions of precursors such as NOx, CO and VOVs emitted in relation to transportation by human beings [31-32], in which process, differences in emission quantity, emission source, pollutant transport, geographical environment and meteorological conditions result in different characteristics of ozone distribution [33]. There is little literature on the comparative study of the distribution characteristics of ozone pollution in coastal and inland cities.

2. Data

This study investigated the hourly monitoring data of O_3 concentration in four coastal cities (Qingdao, Yantai, Weihai, Rizhao) and four inland cities (Dezhou, Liaocheng, Heze, Jining) (their respective regional range is as shown in Fig. 1) in Shandong Province from 2009 to 2017, and the NO_2 and CO concentrations which are closely associated with O_3 concentration. Based on this, we compared and analyzed the distribution characteristics of O_3, NO_2 and CO concentrations in the boundary layer in coastal cities and inland cities on different time scales, namely, annual, seasonal and diurnal, etc., and studied the correlation between the three pollutants. The results can provide a basis for exploring the ozone pollution mechanism in mid-latitude regions and control of urban photochemical pollution.
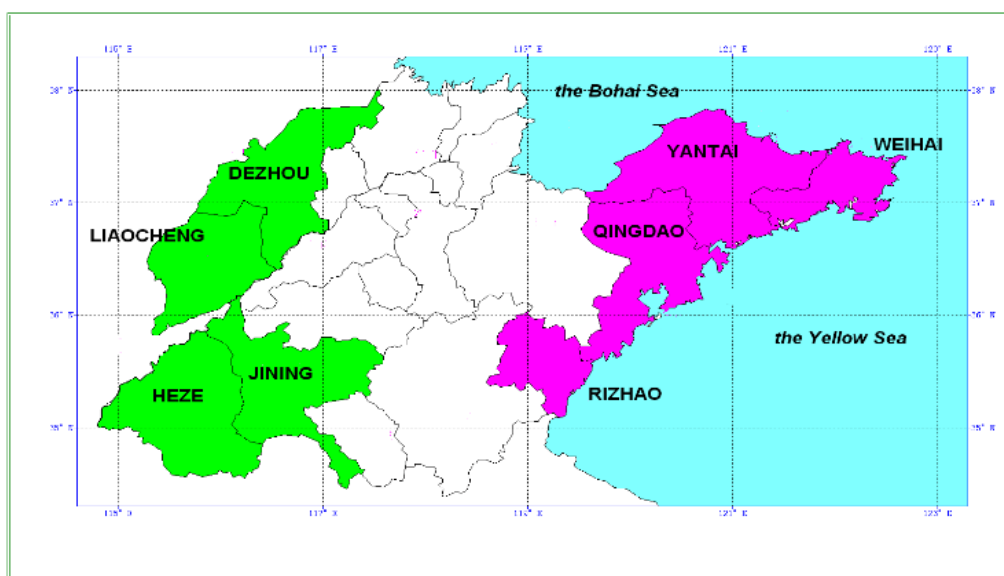

Fig. 1 Areas covered in this study: coastal cities (Qingdao, Yantai, Weihai, Rizhao) and inland cities (Dezhou, Liaocheng, Heze, Jining) in mid-latitude (Shandong Province)

3. Analysis of results

3.1. Annual variations

3.1.1. O_3. It can be seen from the annual variation curve of the average O_3 concentration in the mid-latitude coastal and inland cities between 2009 and 2017 (Fig. 2) that the annual average concentration of O_3 in coastal cities is higher than that in inland cities, indicating that O_3 pollution in coastal cities is

more serious than that in inland cities, which is consistent with the research results in Jiangsu Province [23]. From 2009 to 2017, the average concentration of O_3 in coastal and inland cities showed a rising trend, both hitting a maximum in 2017. The upward trend in inland cities was more evident: It grew year upon year after experiencing a phased low in 2010 (46.02 $\mu g/m^3$), and hit a maximum in 2017 (72.96 $\mu g/m^3$). The rising trend in coastal cities was weaker than that in inland cities. After reaching a phased high (75.81 $\mu g/m^3$) in 2010, it continued to decline to a phased low (58.50 $\mu g/m^3$) in 2011 and 2012. It only began to show an upward tendency in 2013, and experienced continuous decline in 2015 and 2016. Again, it hit a maximum (80.33 $\mu g/m^3$) in 2017. Between 2009 and 2017, the average difference of O_3 concentration between coastal and inland cities was significantly narrowed (with a difference of 22.43 $\mu g/m^3$ in 2009, and 7.37 $\mu g/m^3$ in 2017).

3.1.2. NO_2 and CO. The annual variation curve of the average concentration of NO_2 and CO in coastal and inland cities from 2009 to 2017 (Fig. 2) shows a different tendency from that of average O_3 concentration in that the annual average concentration of the former two in coastal cities is lower than that in inland cities, showing a downward tendency during the study period.

NO_2 concentrations in both coastal and inland cities reached their maximum (40.14 $\mu g/m^3$ and 50.88 $\mu g/m^3$, respectively) in 2011, and fell to a smaller value (31.44 $\mu g/m^3$) and a minimum (39.91 $\mu g/m^3$) in 2017, respectively. The difference in average concentration of NO_2 between coastal cities and inland cities was also on the decrease (10.74 $\mu g/m^3$ and 8.47 $\mu g/m^3$, respectively), and the decrease in inland cities was more obvious. The fluctuations of annual average concentration of NO_2 in coastal and inland areas were roughly in the same direction.

Between 2009 and 2017, the annual average CO concentration in coastal and inland cities showed a monotonous decreasing trend, among which, the annual average CO concentration in inland cities in 2010-2013 declined significantly, decreasing from 3.27 mg/m^3 in 2010 to 1.66 mg/m^3 in 2013, and then gradually slowing down its declining magnitude year by year after 2013. Differences between coastal and inland cities were significantly narrowed down (from a difference of 3.14 mg/m^3 in 2009 to 0.53 mg/m^3 in 2017).

3.1.3. Summary. The comparative analysis of the annual average concentration of O_3, NO_2 and CO in coastal and inland cities from 2009 to 2017 shows that the average concentration of O_3 in coastal cities was higher than that in inland cities. Although it showed an upward trend in both coastal and inland cities, it was more evident in inland cities. The annual average concentrations of NO_2 and CO in coastal cities were lower than those in inland cities. Although they both showed a downward trend in both coastal and inland cities, the down trend was more evident in inland cities. The difference in annual average concentration of O_3, NO_2 and CO in coastal and inland cities decreased significantly. The annual average concentration of O_3 showed an opposite trend as against those of NO_2 and CO.

The difference in the average concentration of O_3, NO_2 and CO in coastal and inland cities pointed to the important impact of sea-land distribution on their concentrations. The pollution caused by SO_2, PM_{10}, $PM_{2.5}$, NO_2 and CO and other particulate matter in inland cities was more serious than that in coastal cities. The concentrations of NO_2 and CO in inland cities were higher than those in coastal cities, and as a result, affected visibility and solar radiation. The photochemical reaction in coastal cities was more active than that in inland cities, and the average concentration of O_3 was higher than its counterpart in inland cities. The annual variations in O_3, NO_2 and CO concentrations indicate that the large-scale atmospheric environmental control measures taken in China since 2010 have achieved notable results. The narrowed difference in pollutant concentrations between inland cities and coastal cities and the rise in photochemical pollutant O_3 concentration have both demonstrated this. In this respect, inland cities' achievements are particularly notable.
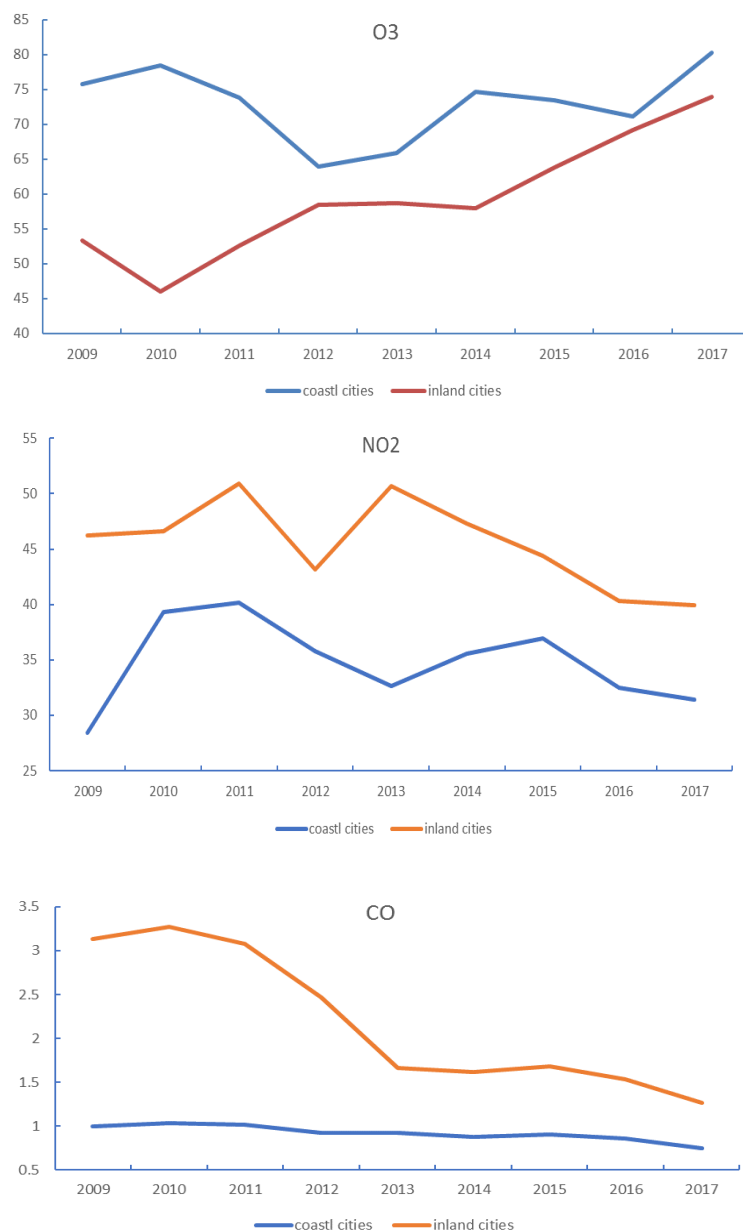
Fig. 2 Distribution of annual average concentrations of O_3($\mu g/m^3$), NO_2 ($\mu g/m^3$) and CO(mg/m^3) in coastal and inland cities in the mid-latitude regions

3.2. Monthly variation

3.2.1. O₃. An analysis of the variation curve of O_3 average concentration in the mid-latitude coastal and inland cities (Fig. 3) reveals that there is a significant seasonal difference in O_3 pollution, high in spring and autumn, and low in winter. Except for June, the monthly average concentration of O_3 in coastal cities was higher than that in inland cities at other months. The annual change took on a M-shaped bimodal (May and September) distribution structure, similar to Shanghai and other cities in the Yangtze River Basin [21] [24] [30], with a higher concentration in spring and autumn and a lower one in winter. The maximum appeared in May (97.15 $\mu g/m^3$), and the second peak appeared in September (88.51 $\mu g/m^3$). July and August were relative valley, and December witnessed the lowest (42.02

$\mu g/m^3$). By contrast, inland cities showed an inverted V-shaped unimodal distribution, consistent with the characteristics of northern cities such as Beijing [18] [19] [25] and Zhengzhou [22][25], with a high concentration of pollution in early summer and a low one in winter. The maximum appeared in June (96.15 $\mu g/m^3$), followed by May (84.23 $\mu g/m^3$), July (81.75 $\mu g/m^3$), and with December being the lowest (27.23 $\mu g/m^3$). The highest concentration of O_3 found in cities of Guangxi and Guangdong appeared in winter [16-17] [28-29].

Ozone is the product of the photochemical reaction, while solar radiation and temperature are the main influencing factors [36]. From winter to early summer (June), with the increase in the solar radiation and rise in temperature, the photochemical reaction becomes active, and ozone concentration rises as a result. The temperature in inland areas is generally higher than that in coastal areas in June, and ozone concentration there reaches the pinnacle. Due to increase in precipitation during July and August, solar radiation is reduced instead, and consequently, ozone concentration also decreases. Into the autumn, as the solar radiation and temperature begin to decrease and the photochemical reaction is inhibited to a certain degree, ozone concentration declines accordingly. Ozone concentration is also lowest when the solar radiation and temperature hit the lowest in winter. According to Li et al., the valley of ozone concentration appearing in summer (July and August) was closely related to the summer monsoon circulation [34], and thereby this phenomenon is also found in many cities in the Yangtze River Basin in China [30]. This study found that this phenomenon also exists in the mid-latitude coastal cities, yet is absent in inland cities of the same latitude, indicating that the mid-latitude coastal areas are more significantly affected by the summer monsoon compared with inland areas of the same latitude.

3.2.2. NO$_2$ and CO. Analysis of the monthly average variation curve of NO$_2$ and CO concentrations in coastal and inland cities during 2009 and 2017 (Fig. 3) reveals that the monthly average concentrations of NO$_2$ and CO were all lower in coastal cities than those in inland cities. Both the coastal and inland cities were characterized by a light pollution in summer and a heavy one in winter, exhibiting a V-shaped unimodal structure. Inland cities experienced a fluctuation marked by a slight increase in spring (April and May) first, and then a continued decline. There was no such phenomenon in coastal cities. NO$_2$ was at lowest in July (28.83 $\mu g/m^3$) and highest in January (63.09 $\mu g/m^3$); CO was at lowest in June (1.69 mg/m^3) and highest in January (3.01 g/m^3) in inland cities. By contrast, NO$_2$ was at lowest in July (23.95 $\mu g/m^3$), and highest in January (44.80 $\mu g/m^3$); CO was at lowest in June (0.74 mg/m^3) and highest in January (1.30 mg/m^3) in coastal cities.

The seasonal variation of NO$_2$ and CO concentrations was mainly affected by precipitation subsidence and the boundary layer pressure field [35]. The monthly precipitation in China's cities was inversely correlated with NO$_2$. High air pressure has a continuous accumulative effect on pollutants such as NO$_2$ and CO. In summer, continental high pressure is characterized by weak intensity, small range, and short duration. Therefore, the accumulation time for pollutants such as NO$_2$ and CO is short, resulting in light pollution. In autumn and winter, most of China is under the control of continental high pressure. The high pressure system is marked by its strength, wide range of influence, and long duration, exerting a long-lasting effect on the continuous accumulation of pollutants. The pollution in December and January is the most serious in the whole year, with the largest pollution range.

3.2.3. Summary. A comparison of the monthly average concentrations of O$_3$, NO$_2$ and CO in coastal and inland cities shows that except for June, the average ozone concentration in coastal cities was higher than that in inland cities, displaying an obvious seasonal variation. The annual variation in coastal cities showed a M-shaped bimodal distribution structure, high in spring and autumn and low in winter. By contrast, the monthly variation in inland cities showed an inverted V-shaped unimodal distribution, high in early summer and low in winter. The monthly average concentrations of NO$_2$ and CO in coastal cities were all lower than those in inland cities, with also an obvious seasonal variation. It is a V-shaped unimodal structure, with a light pollution level in summer and a heavy one in winter.

The maximum and minimum of NO_2 appeared in January and July, respectively, while those of CO appeared in January and June, respectively. Inland cities experienced a fluctuation of slightly increasing first in spring (April and May), and then decreasing.
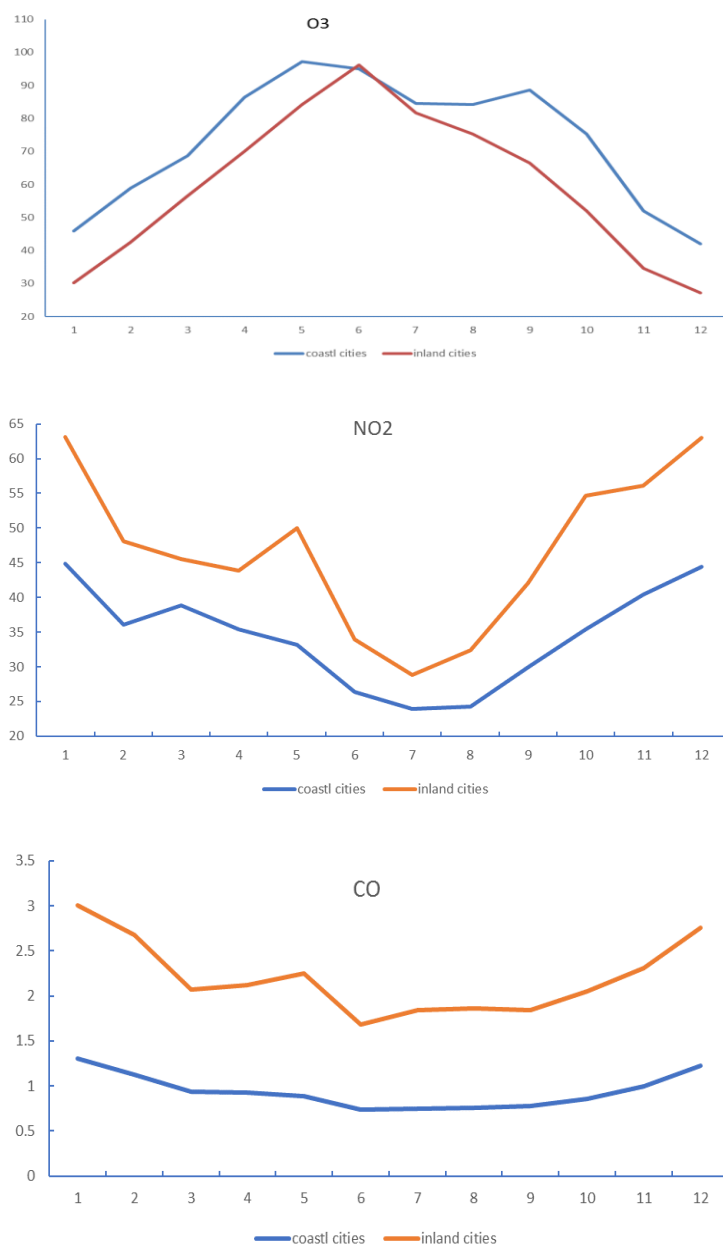

Fig. 3 Distribution of monthly average concentrations of O_3 (µg/m³), NO_2 (µg/m³) and CO (mg/m³) in coastal and inland cities in the mid-latitude regions

3.3. Hourly variation

3.3.1. O_3. It can be seen from the variation curve (Fig. 4) that the hourly average concentration of O_3 in coastal and inland cities shared similar characteristics of the diurnal variation, displaying a unimodal structure characteristic of being low in the morning and high in the afternoon. The maximum of O_3 concentration in coastal cities appeared at 14 o'clock (97.65 µg/m³), and the minimum (54.92

$\mu g/m^3$) at 7 o'clock. The maximum in inland cities appeared at 15 o'clock (94.49 $\mu g/m^3$), later than that in coastal cities, and the minimum (35.09 $\mu g/m^3$) appeared at 6 o'clock, earlier than that in coastal cities. At 16 o'clock, the average concentrations of O_3 were approximate in both coastal and inland cities.

The sunrise time in coastal cities is earlier than that in inland cities, and the peak of hourly average ozone concentration also appears a little earlier. There must be some other causes responsible for the late appearance of the lowest value of ozone concentration in coastal cities compared with inland cities, which merits further study.

3.3.2. NO₂ and CO. It can be seen from the variation curves (Fig. 4) that the hourly average concentration of NO_2 and CO in both coastal and inland cities shared the characteristics of the diurnal variation, displaying a bimodal structure characteristic of being high both in the morning and evening. The maximum (40.20 $\mu g/m^3$) of the average concentration of NO_2 in coastal cities appeared at 8:00 in the morning, and the minimum (26.36 $\mu g/m^3$) appeared at 13 o'clock; The maximum (55.66 $\mu g/m^3$) in inland cities appeared at 19 o'clock in the evening, while the minimum (31.73 $\mu g/m^3$), same as that in coastal cities, also appeared at 13 o'clock. The maximum (1.09mg/m^3) of average concentration of CO in coastal cities appeared at 8:00 in the morning, while the minimum (0.85 $\mu g/m^3$) appeared at 15 o'clock. The maximum (2.37 mg/m^3) of average concentration of CO in inland cities appeared at 8:00 in the morning, while the minimum (1.92 mg/m^3), same as that in coastal cities, also appeared at 15 o'clock.

The hourly average concentrations of NO_2 and CO sharing characteristics of the bimodal diurnal variation corresponded to morning and evening traffic peaks, and the inland evening peak was significantly higher than others.

3.3.3. Summary. A comparative analysis of hourly average concentration of O_3, NO_2 and CO in coastal and inland cities reveals that the hourly average concentration of O_3 in coastal and inland cities both showed a unimodal structure of being low in morning and high in the afternoon. The maximum of O_3 concentration in coastal cities appeared one hour earlier than that in inland cities, while the minimum appeared one hour later than that in inland cities. The hourly average concentration of NO_2 and CO in coastal and inland cities exhibited characteristics of bimodal diurnal variation featuring a peak either in the morning and evening. The maximum of NO_2 concentration in coastal cities appeared in the morning, whereas that in inland cities appeared in the evening. The minimum in both coastal and inland cities appeared in the afternoon. The maximum and minimum of CO also appeared at the same time, at 8 and 15 o'clock, respectively.
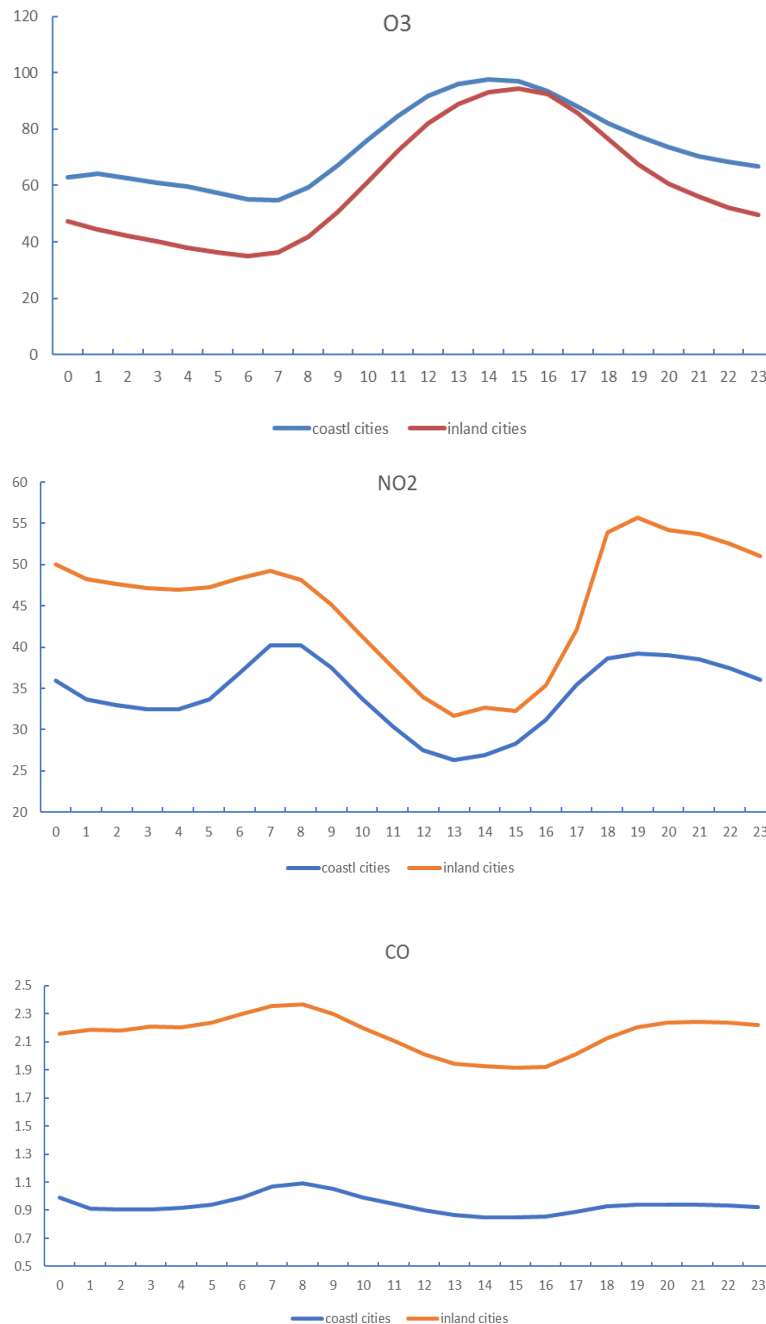
Fig. 4 Distribution of hourly average concentrations of O3 ($\mu g/m^3$), NO2 ($\mu g/m^3$) and CO (mg/m^3) in coastal and inland cities in the mid-latitude regions

4. Conclusion

(1). The average concentration of O3 in coastal cities was higher than that in inland cities. and both showed an upward trend during 2009 and 2017, especially prominently in inland cities. The annual average concentrations of NO_2 and CO in coastal cities were lower than those in inland cities, both of which showed a downward trend during 2009 and 2017, with a more prominent downward trend for inland cities. The difference in annual average concentration of O_3, NO_2 and CO in coastal and inland cities decreased significantly. The average concentration of O_3 showed an opposite trend with those of NO_2 and CO. The sea-land distribution has an important impact on O_3, NO_2 and CO concentrations.

During the study period, the narrowed difference in pollutant concentrations between inland cities and coastal cities and the rise in photochemical pollutant O_3 concentration indicate that China's large-scale measures to address atmospheric environmental pollution have achieved considerable results, especially in inland cities.

(2). The average concentration of O_3 had an obvious seasonal variation. Its annual variation in coastal cities showed a M-shaped bimodal structure featuring highest in spring and autumn and lowest in winter. The average concentration of O_3 in inland cities exhibited an inverted V-shaped unimodal distribution of with a peak in early summer and valley in winter. The major influencing factors were solar radiation and temperature. The monthly average concentrations of NO_2 and CO also showed an obvious seasonal variation, both of which showed a V-shaped unimodal structure with the pollution being light in summer and heavy in winter. The maximum and minimum of NO_2 appeared in January and July, respectively, while the maximum and minimum of CO appeared in January and June, respectively. The major influencing factors were precipitation and the boundary layer pressure field. Inland cities experienced a fluctuation by increasing slightly first in spring (April and May) , and then decreasing.

(3). The hourly average concentrations of O_3 in coastal and inland cities both showed a unimodal structure with a low level of pollution in the morning and a high one in the afternoon. As the sunrise time in coastal cities is earlier than that in inland cities, the maximum average value of O_3 hours also appears a little earlier. There must be other causes for a later appearance of the minimum of ozone concentration in coastal cities than in inland cities, which remains a further study topic. Hourly average concentrations of NO_2 and CO sharing characteristics of the bimodal diurnal variation corresponded to morning and evening traffic peaks, and the inland evening peak was significantly higher than others.

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