Chin. Geogra. Sci. 2019 Vol. 29 No. 6 pp. 917–933
https://doi.org/10.1007/s11769-019-1081-8

Springer Science Press
www.springerlink.com/content/1002-0063

Spatiotemporal Variations and Regional Transport of Air Pollutants in Two Urban Agglomerations in Northeast China Plain

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Abstract: Characteristics of air pollution in Northeast China (NEC) received less research attention in the past comparing to other heavily polluted regions in China. Spatiotemporal variations of six criteria air pollutants (PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3 and CO) in Central Liaoning Urban Agglomeration (CLUA) and Harbin-Changchun Urban Agglomeration (HCUA) in NEC Plain were analyzed in this study based on three-year hourly observations of air pollutants and meteorological variables from 2015 to 2017. The results indicated that the annual mean concentrations of air pollutants are generally higher in the middle and southern regions in NEC Plain and lower in the northern region. Megacities such as Shenyang, Harbin and Changchun experience severe air pollution, with a three-year averaged air quality index (AQI) larger than 80, far exceeding the daily AQI standard at the first-level of 50 in China. The annual mean PM and SO_2 concentrations decrease most significantly in NEC urban agglomerations from 2015 to 2017, followed by CO and NO_2, while O_3 shows a slight increasing trend. All the six pollutants exhibit obvious seasonal and diurnal variations, and these variations are dictated by local emission and meteorological conditions. $PM_{2.5}$ and O_3 concentrations in NEC urban agglomerations strongly depend on wind conditions. High O_3 concentrations at different cities usually occur in presence of strong winds but are independent on wind direction (*WD*), while high $PM_{2.5}$ is usually accompanied by weak winds and poor dispersion condition, and sometimes also occur when the northerly or southerly winds are strong. Regional transport of air pollutants between NEC urban agglomerations is common. A severe haze event on November 1–4, 2017 is examined to demonstrate the role of regional transport on pollution.

Keywords: criteria air pollutant; meteorological condition; regional transport; urban agglomeration of Northeast China

Citation: LI Xiaolan, HU Xiao-Ming, SHI Shuaiyi, SHEN Lidu, LUAN Lan, MA Yanjun, 2019. Spatiotemporal Variations and Regional Transport of Air Pollutants in Two Urban Agglomerations in Northeast China Plain. *Chinese Geographical Science*, 29(6): 917–933. https://doi.org/10.1007/s11769-019-1081-8

1 Introduction

Over the past three decades, urban agglomerations that are made up of groups of large, nearly contiguous cities with many adjoining satellite cities and towns have played an important role in the economic growth of

Received date: 2018-11-01; accepted date: 2019-01-30

Foundation item: Under the auspices of National Key Research and Development Program of China (No. 2017YFC0212301, 2016YFC0203304), Basic Research Funds of Central Public Welfare Research Institutes (No. 2018SYIAEZD4), Program of Liaoning Meteorological Office (No. 201904, D201603), Key Program of National Natural Science Foundation of China (No. 41730647), Program of Laboratory of Atmospheric Chemistry, China Meteorological Administration (No. 2017B02), Key Program of Natural Science Foundation of Liaoning Province (No. 20170520359)

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China due to their collective economic capacity and interdependency (Shao et al., 2006). However, such economic boom is being overshadowed by the general decline in air quality. In every major urban agglomeration across China, concentrations of air pollutants greatly exceed standards recommended by the World Health Organization (WHO) (Zhang et al., 2015). Northeast China (NEC) have been regarded as the fifth most polluted region in China in terms of air quality, following the Beijing-Tianjin-Hebei megalopolis, the Pearl River Delta, the Yangtze River Delta, and the Sichuan Basin (Chen et al., 2017; Ma et al., 2017; Li et al., 2018a). Despite the economic recession in NEC in the past three decades, air quality still deteriorated in the region. Severe haze events occurred frequently in urban agglomerations in NEC Plain in recent years, especially in winter and occasionally in fall. Some of these haze events swept a large area in NEC. For example, an extreme haze event on October 20–23, 2013 in NEC Plain affected an extensive area (> 1×10^6 km^2) and caused dramatic negative impacts on the public (Chen et al., 2017).

Severe haze episodes in NEC can be caused by the combination of poor dispersion conditions, regional chemical transport, intensive coal combustion for house heating in winter, biomass burning, and other episodic emissions (Yang et al., 2012; 2017; Chen et al., 2017; Li et al., 2018b, 2019; Miao et al., 2018a; Ma et al., 2018). As one of the major regions for crop production in China, approximately 20% of the total arable land area in NEC is covered by crops (China Agricultural Yearbook Editorial Committee, 2013). Crop-straw burning in the harvest season, i.e., late fall, is a significant emission source in NEC (Chen et al., 2017). Beside this agriculture activity, house heating (through coal burning in cities and agricultural residue burning in rural area) during the prolonged winter season in NEC is another major contributor for the severe haze pollution in the region. The heating season (lasting for 4–6 months) in NEC is longer than any other Chinese regions because of its cold weather (Chen et al., 2018). In Harbin, a provincial capital in NEC, the heating season usually lasts from 15 October to 15 April, which is about 1 months longer than the heating season in Beijing. In addition to enhanced local emissions, regional transport of air pollutants from the North China Plain can exacerbate the air pollution in NEC at times (Li et al., 2018b; Miao et al., 2018a).

Since the Chinese Ministry of Environmental Protection (MEP) updated the Ambient Air Quality Standards in February 2012, concentrations of six criteria air pollutants ($PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO) have been monitored at the national air quality sites in some major cities in China. Using these air quality data and other measurements, spatiotemporal characteristics of some/ all of the six criteria air pollutants as well as their relationships with meteorological parameters have been investigated in dozens of major cities (Chai et al., 2014; Wang et al., 2015; Zhang et al., 2016; Zhao et al., 2016; He et al., 2017; Song et al., 2017) and several heavily polluted regions in China, including Sichuan Basin (Hu et al., 2016; Zhao et al., 2018), North China Plain (Zhang et al., 2015), Yangtze River Delta (Hu et al., 2014), and Northwest China such as Gansu and Shaanxi provinces (Guan et al., 2017; Xu et al., 2018). Similar studies have also been conducted in NEC, but were mostly limited to the provincial capital cities. Li et al. (2017) and Liu et al. (2019) analyzed temporal variation and spatial distribution of particulate matter (PM_{10} and $PM_{2.5}$) and O_3 during recent 2–3 yr in Shenyang, the provincial capital city of Liaoning Province in NEC. Ma et al. (2017) studied the statistical characteristics of the six criteria air pollutants and air quality index (AQI) during 2013–2015 in Changchun, the provincial capital city of Jilin Province in NEC. Their studies revealed that concentrations of particulate matter and gaseous pollutants (except O_3) in winter months/at urban sites were much higher than those in other months/at suburban sites. However, the current state and formation mechanism of regional pollution in NEC urban agglomerations are still unclear.

To understand the characteristics of regional pollution in urban agglomerations in NEC Plain, this work statistically studied the spatial distributions and temporal variations of the six criteria air pollutants and their regional transport in two urban agglomerations in NEC Plain from 2015 to 2017 using data at the national air quality sites, as well as surface meteorological observational data and satellite fire points data. The present study is meaningful for understanding the current state and formation mechanism of regional pollution in NEC Plain, and can provide references for policy making to alleviate air pollution in this region.

2 Data and Methods

2.1 Urban agglomerations in Northeast China Plain

The NEC Plain is surrounded by mountains, including the Da Hinggan Mountains in the west, the Xiao Hinggan Mountains in the north, and the Changbai Mountains in the east (Fig. 1a). Two urban agglomerations are located in NEC Plain, named as central Liaoning Urban Agglomeration (CLUA) that contains four cities in Liaoning Province and the Harbin-Changchun Urban Agglomeration (HCUA) that consists of five cities in Heilongjiang Province and six in Jilin Province (Fig. 1b). Table 1 summarizes the population, gross domestic product (GDP), and quantity of heat supplied at different cities in CLUA and HCUA in 2017, as well as the mean annual precipitation and air temperature from 2015 to 2017.

2.2 Data sources

Hourly air quality monitoring data used in this study include hourly mean mass concentrations of the six criteria atmospheric pollutants ($PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO) and AQI from 2015 to 2017 at 15 cities in NEC. The air quality data were downloaded from the real-time air quality publishing systems of Liaoning Province (http://211.137.19.74: 8089/), Jilin Province (http://hbj.jl.gov.cn/kqzljc/), and Heilongjiang Province (http://111.40.0.99:8081/). Eight-hour average O_3 (hereafter referred to as O_3-8h for short) is calculated to examine the upper end of daily O_3.

The corresponding hourly mean meteorological parameters such as wind speed (*WS*), wind direction (*WD*), air temperature (T_a), and relative humidity (*RH*) observed at national or basic meteorological weather stations in each city from 2015 to 2017 were obtained from China Meteorological Administration, to examine the dependence of air pollutants on meteorological condition. In addition, the European Centre for Medium Range Weather Forecasts (ECMWF) reanalysis data with a spatial resolution of 0.125° × 0.125° were used to retrieve the horizontal flow fields at 10 m above ground level (AGL) over NEC from November 1 to 4, 2017, which can be obtained four times a day at 02:00, 08:00, 14:00, and 20:00 Local Time (LT) (http://apps.ecmwf.int/datasets/data/interim-full-daily/levtype=sfc/).

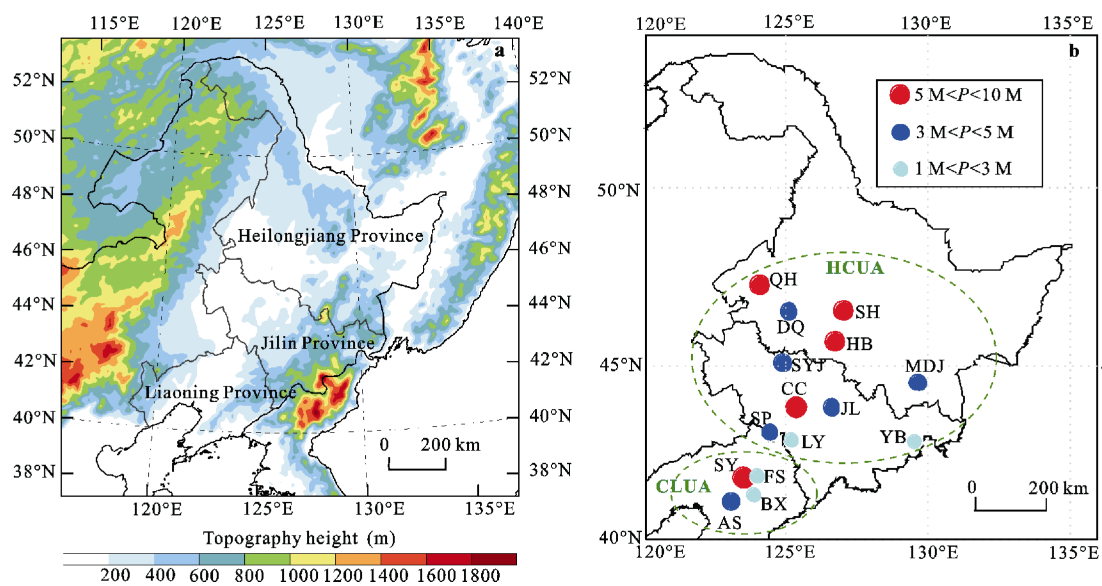

Fig. 1 Topography and provinces in Northeast China (a), and locations of 15 cities in the Central Liaoning Urban Agglomeration (CLUA) and in the Harbin-Changchun Urban Agglomeration (HCUA) of NEC (b), with dot size and color indicating population (*P*) size (M, million; SY, AS, FS and BX in the CLUA are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, respectively; CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH in HCUA represent Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively)

The satellite fire point data captured by the Moderate Resolution Imaging Spectroradiometer (MODIS) were acquired from the Level-1 and Atmosphere Archive & Distribution System (LAADS) Distributed Active Archive Center (DAAC) and MODIS-retrieved vegetation fraction for cropland were used to analyze the origination of air pollutants caused by fall agriculture residue burning. The data contains all fire point information in NEC on November 1–4, 2017 (https://ladsweb.modaps.eosdis.nasa.gov/).

3 Results

3.1 Spatial and temporal variations of air pollutants

3.1.1 Average air pollutant levels in urban agglomerations of NEC Plain

The mean concentration values of $PM_{2.5}$, PM_{10}, SO_2, NO_2, the daily maximum of O_3-8h, and CO from 2015 to 2017 at 15 cities of NEC Plain are listed in Table 2 and their distributions are shown in Fig. 2. Overall, air pollution in CLUA was observed heavier than that in HCUA. The three-year averaged AQI of 82.2 ± 53.6 in CLUA was higher than that of 71.2 ± 56.1 in HCUA (Table 2). The spatial distributions of concentrations of the six air pollutants all showed higher values in the southern and middle regions and lower in the northern and surrounding regions of NEC Plain. However, Harbin was exceptional due to possessing the largest population, GDP, and energy consumption among the whole 15 cities (Table 1). Big cities with population larger than 5 millions in NEC (in Fig. 1b) mostly experienced terrible air pollution (annual mean AQI > 80), while Qiqihar and Suihua were different, partly because of low local pollutant emissions related to small population density and GDP in the two cities as well as their geographic locations. The lowest AQI and pollutant concentrations were usually observed in Yanbian Korea Autonomous Prefecture (YB), which is located in the west edge of NEC and is adjacent to North Korea, far away from the core area of air pollution in NEC. Therefore, YB can be taken as an air pollution background city in NEC urban agglomerations.

Table 1 Population, gross domestic product (GDP), and annual quantity of heating supply during 2017 and annual mean precipitation and air temperature during 2015–2017 at different cities in CLUA and HCUA in Northeast China Plain

City cluster	City	Province	Population (million)	GDP (billion yuan RMB)	Heating supply (million GJ)	Precipitation (mm)	Mean temperature (℃)
Central Liaoning Urban Agglomeration	SY*	Liaoning	7.34	554.6	139.1	634.7	9.0
	AS	Liaoning	3.46	146.2	42.9	614.0	11.3
	FS	Liaoning	2.15	86.5	26.5	743.8	7.4
	BX	Liaoning	1.50	76.7	13.1	718.3	9.1
Harbin-Changchun Urban Agglomeration	HB*	Heilongjiang	9.62	610.2	158.3	457.9	5.2
	QH	Heilongjiang	5.44	132.5	28.63	417.8	4.8
	MDJ	Heilongjiang	2.52	123.1	22.93	590.4	4.8
	DQ	Heilongjiang	2.78	261.0	67.75	465.9	5.7
	SH	Heilongjiang	5.43	131.6	6.44	593.8	4.6
	CC*	Jilin	7.53	598.6	–	622.8	7.0
	JL	Jilin	4.22	245.4	–	763.2	6.7
	SP	Jilin	3.24	119.4	–	790.8	7.6
	LY	Jilin	1.20	76.5	–	690.0	6.5
	SYJ	Jilin	2.78	165.2	–	516.1	6.5
	YB	Jilin	2.12	87.6	–	606.5	6.3

Notes: * means provincial capital city, and '–' means lack of data; the data of Liaoning Province are obtained from http://www.ln.stats.gov.cn/tjsj/sjcx/ndsj/, Heilongjiang Province from http://www.hlj.stats.gov.cn/tjnj/, and Jilin Province from http://tjj.jl.gov.cn/tjnj/. SY, AS, FS and BX in the CLUA are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, respectively; CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH in HCUA represent Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively

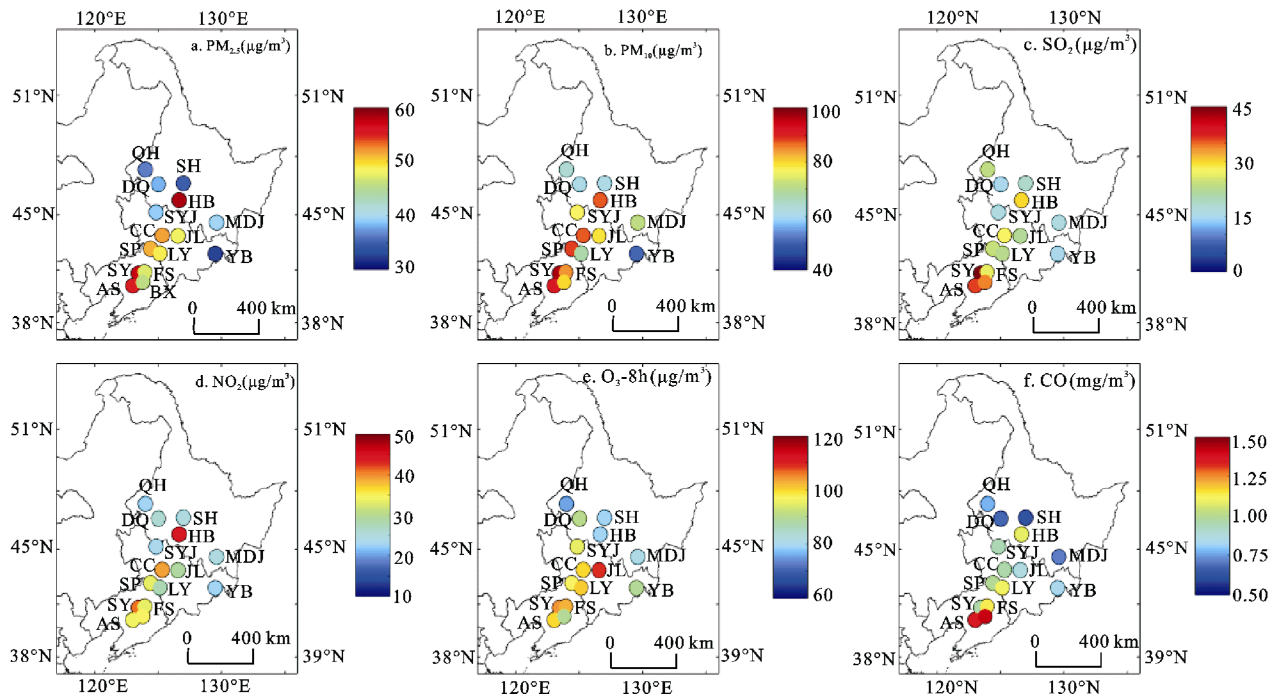

Fig. 2 Spatial distributions of mass concentrations of (a) $PM_{2.5}$, (b) PM_{10}, (c) SO_2, (d) NO_2, (e) daily maximum O_3-8h, and (f) CO averaged from 2015 to 2017 in 15 cities of Northeast China. SY, AS, FS, BX, CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively

In terms of individual air pollutant, PM ($PM_{2.5}$ and PM_{10}) pollution was the most severe in Shenyang, Harbin and Anshan, with the largest $PM_{2.5}$ value of (58.3 ± 74.1) μg/m^3 observed in Harbin and the largest PM_{10} of (96.2 ± 81.8) μg/m^3 in Shenyang. The mean SO_2 concentration in CLUA was (35.8 ± 38.6) μg/m^3, almost as twice as that in HCUA ((20.8 ± 23.3) μg/m^3). The annual mean concentrations of CO, NO_2, and daily maximum O_3-8h in CLUA were about 39%, 25% and 10% higher than that in HCUA, with the maximum CO, NO_2, and O_3-8h concentrations observed in Benxi ((1.42 ± 0.73) mg/m^3), Harbin ((40.9 ± 21.6) μg/m^3) and Jilin ((109.5 ± 39.6) μg/m^3), respectively.

3.1.2 Seasonal variation and distribution of air pollutant concentrations

Monthly mean AQI and mass concentrations of the six criteria air pollutants as well as the ratio of $PM_{2.5}/PM_{10}$ in CLUA and HCUA from 2015 to 2017 are shown in Fig. 3. Except for O_3, all the other variables had similar seasonal variations, decreasing from January to June at first, remaining a low level till September, and then increasing to the end of the year. High concentrations in winter months were directly caused by extra pollutant emissions such as domestic heating and agricultural burning (Li et al., 2018b) and also greatly exacerbated by poor dispersion conditions in winter (Ji et al., 2012; Miao et al., 2018b). In contrast, the highest O_3 concentrations usually occurred in summer due to photochemical O_3 formation, which was enhanced by strong radiation, and high temperature (Seinfeld and Pandis, 2006).

A decreasing trend can be described for most variables in Fig. 3, suggesting that air quality in NEC urban agglomerations is being gradually improved, especially during winter months. For example, the maximum monthly mean $PM_{2.5}$ and PM_{10} concentrations in CLUA during winter of 2017 declined by 35% and 22% approximately comparing to their levels during winter of 2015 (Figs. 3b, 3c). The ratio of $PM_{2.5}/PM_{10}$ can roughly reflect PM pollution types (fine PM pollution and coarse PM pollution) on some certain degree, and it did not change so much during the three years. The $PM_{2.5}/PM_{10}$ ratio was higher in fall and winter and was the lowest in spring mainly due to the increasing of coarse particles generated from frequent sand-dust events (Li et al., 2017). In contrast to CLUA, the $PM_{2.5}/PM_{10}$ ratio in HCUA was relatively higher in winter and lower in summer (Fig. 3d).

Table 2 List of three-year (2015–2017) averaged air quality index (AQI) and air pollutant concentrations in different cities of Northeast China. The numbers with bold font represent the average for each city cluster, and those with underlines represent the top three values for a given variable

City Cluster	City	AQI	$PM_{2.5}$ (μg/m^3)	PM_{10} (μg/m^3)	SO_2 (μg/m^3)	CO (mg/m^3)	NO_2 (μg/m^3)	O_3-8h (μg/m^3)
Central Liaoning Urban Agglomeration	SY	89.8±61.5	57.2±57.4	96.2±81.8	45.9±54.7	0.97±0.57	40.9±21.6	104.3±50.3
	AS	85.0±55.7	54.9±52.2	91.1±74.9	36.2±36.1	1.35±0.68	34.0±18.5	100.1±45.4
	FS	80.0±50.0	47.3±41.1	84.0±64.8	26.7±26.0	1.13±0.65	33.3±18.5	101.9±45.1
	BX	74.1±47.0	45.8±42.8	79.7±62.5	34.4±37.7	1.42±0.73	34.6±19.3	89.5±37.7
	Mean	82.2±53.6	51.3±48.4	87.8±71.0	35.8±38.6	1.21±0.66	35.7±19.5	99.0±44.5
Harbin-Changchun Urban Agglomeration	HB	86.5±74.7	58.3±74.1	86.9±82.5	29.9±36.1	1.08±0.59	44.3±24.9	79.1±35.2
	QH	64.0±49.5	36.4±41.6	63.6±57.7	23.3±23.1	0.78±0.44	22.8±14.0	75.4±29.9
	MDJ	66.6±43.7	39.7±36.2	70.9±54.0	16.2±17.2	0.71±0.54	24.9±14.1	81.5±29.1
	DQ	63.3±52.6	38.4±44.8	60.8±85.3	15.1±12.2	0.69±0.36	26.0±14.5	89.9±33.3
	SH	58.7±58.1	33.7±48.3	59.6±81.5	18.2±27.2	0.64±0.48	23.7±20.6	79.5±30.8
	CC	83.7±60.8	51.5±55.4	86.7±82.9	27.8±28.6	0.96±0.48	39.0±20.6	100.1±41.7
	JL	79.1±58.2	48.0±56.9	78.4±86.5	21.9±19.6	0.87±0.47	29.0±16.9	109.5±39.6
	SP	80.2±58.2	49.4±48.2	84.9±73.2	23.8±25.1	1.00±0.59	32.1±21.6	96.4±43.6
	LY	74.0±56.6	47.6±46.9	65.5±60.9	21.9±24.6	1.10±0.50	27.0±19.0	100.4±39.4
	SYJ	71.3±62.1	37.8±51.0	74.3±88.8	15.8±23.5	0.95±0.51	22.5±18.4	94.5±42.9
	YB	55.8±42.4	31.4±34.3	48.9±50.9	14.7±18.7	0.83±0.35	21.8±15.8	89.9±28.4
	Mean	71.2±56.1	42.9±48.9	71.0±73.1	20.8±23.3	0.87±0.48	28.5±18.2	90.6±20.1

Notes: SY, AS, FS and BX in the CLUA are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, respectively; CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH in HCUA represent Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively; The numbers with underlines represent the top three values for a given variable

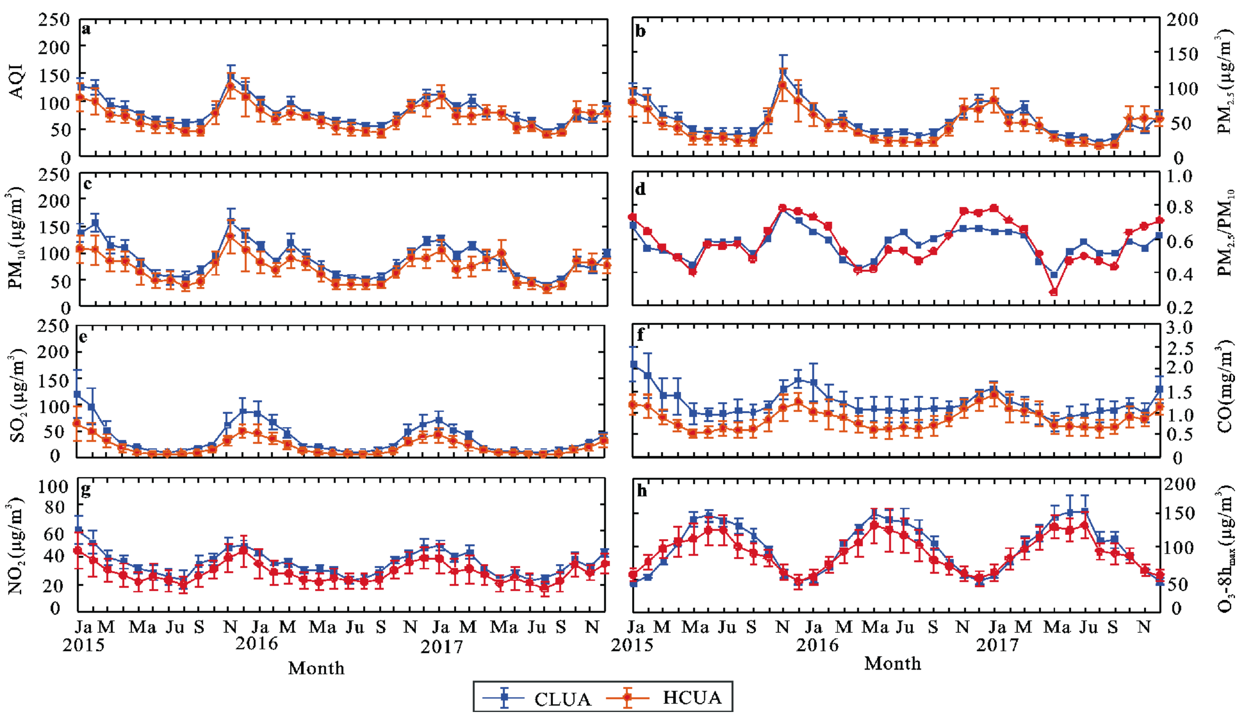

Fig. 3 Temporal variations of monthly mean (a) air quality index (AQI), mass concentrations of (b) $PM_{2.5}$, (c) PM_{10}, (d) SO_2, (e) NO_2, (f) daily maximum O_3-8h, and (g) CO, and (h) the ratio of $PM_{2.5}/PM_{10}$ from 2015 to 2017 in Central Liaoning Urban Agglomeration (CLUA) and Harbin-Changchun Urban Agglomeration (HCUA) in Northeast China Plain. The Ja, M, Ma, Ju, S, N represent January, March, May, July, September, and November, respectively.

Among airborne gaseous pollutants, SO_2 showed the most significant decreasing trend from 2015 to 2017. In CLUA, the peak of monthly mean SO_2 in 2015 reached 119 μg/m^3 and in 2017 decreased to 52 μg/m^3, approaching the SO_2 level in HCUA (Fig. 3e). In general, SO_2 in urban cities is greatly influenced by industrial emissions and coal burning for domestic heating. From April to October (without domestic heating) in each year, SO_2 in both urban agglomerations remained a quite low level. However, during the domestic heating season (from November to March), SO_2 pollution in CLUA was more severe than that in HCUA, and an extreme high SO_2 level occurred in CLUA at the beginning of 2015. Meanwhile, CO and NO_2 were also dropping during the study period (Figs. 3f, 3g), and they were usually related to vehicle emission and agriculture burning.

In contrast to primary pollutants, O_3 is not directly emitted, but formed through complex chemical reactions, which depends on meteorological conditions and its precursor such as NO_x (= NO + NO_2) and volatile organic compounds (VOCs) (Seinfeld and Pandis, 2006; Tang et al., 2012). The mean daily maximum O_3-8h concentration in both city clusters exhibited a small increasing trend, with the peak values in summer months increasing by 10–13 μg/m^3 approximately from 2015 to 2017 (Fig. 3h). Xu et al. (2016) found that the surface O_3 background concentration in the atmosphere gradually increased from 1994 to 2013 according to O_3 measurements at the baseline Global Atmospheric Watch (GAW) station, Waliguan, in China. It was reasonable that O_3 concentration in CLUA during summer seasons was higher than that in HCUA partly because much O_3 was tend to be formed in lower altitudes in presence of abundant sunlight and higher temperature. However, it was interesting to note that O_3 concentration in HCUA from January to March in 2015 exceeded that in CLUA during the same period. It is likely because that under high pollution conditions in CLUA from January to March in 2015, more O_3 tended to be consumed by NO due to the strong titration reaction (Xu et al., 2011a). In contrast to other regions in China, the three-year (2015–2017) averaged daily maximum O_3-8h concentration in Shenyang was 104.3 μg/m^3, which was higher than that observed at 22 major cities in Sichuan Basin during the same period (ranging from 53.4 to 99.7 μg/m^3) (Zhao et al., 2018); the highest daily maximum O_3-8h concentration from 2015 to 2017 in Shenyang (238.7 μg/m^3) was close to that in Beijing (245.2 μg/m^3) and lower than that in Shanghai (263.9 μg/m^3), and Guangzhou (286.7 μg/m^3) from March 2013 to February 2014 (Zhang et al., 2015). It should be paid more attention to the O_3 pollution in NEC in the future after $PM_{2.5}$ pollution has been alleviated.

In addition, the maximum and minimum values of the six criteria air pollutants averaged in different seasons from 2015 to 2017 and their corresponding cities are summarized in Table 3. Seasonal distributions of PM concentrations (Fig. 4) resembled the annual pattern (Fig. 2), with larger values in the southern and middle regions of NEC Plain and smaller values in the northern region. The city with the highest PM concentrations depends on season, such as spring maximum $PM_{2.5}$ (PM_{10}) of 54.9 (112.6) μg/m^3 in Shenyang, summer maximum $PM_{2.5}$ (PM_{10}) of 36.7 (58.2) μg/m^3 in Anshan (Siping), and fall maximum $PM_{2.5}$ (PM_{10}) of 66.4 (91.3) μg/m^3 in Harbin (Shenyang), and winter maximum $PM_{2.5}$ (PM_{10}) of 98.1 (125.7) μg/m^3 in Harbin (Harbin). Zhao et al. (2018) observed that the seasonal maximum $PM_{2.5}$ concentration in the urban agglomeration of Sichuan Basin all occurred in Zigong City that is located in southern part of Sichuan Basin, because PM pollutants in Sichuan Basin were influenced mostly by local emissions and the city with highest PM level did not change with seasons. However, in urban agglomerations in NEC Plain, PM seasonal distribution may be also determined by regional pollutant transport. NO_2 concentrations in Shenyang, Harbin and Changchun were relatively higher than that in the rest of cities during different seasons partly due to their higher GDP (Table 1) and probably also due to more motor vehicles. Zhao et al. (2018) showed that the NO_2 concentration in different cities in Sichuan Basin increased significantly as increase of vehicle numbers and GDP with a correlation coefficient value of 0.43.

The O_3 level at different cities was much higher in spring and summer than in fall and winter, with the highest daily O_3-8h varying from 57.7 μg/m^3 at Jilin in winter to 149.3 μg/m^3 at Shenyang in summer. In contrast to other seasons, SO_2 concentration was quite low during summer at all cities. In winter, SO_2 concentration was high at all cities of CLUA but only at Harbin, Changchun, Siping and Qiqihar in HCUA, which was also the reason why the mean SO_2 level in CLUA appeared much higher than that in HCUA. In addition,

Table 3 Summary of maximum and minimum values of the six criteria air pollutants averaged in different seasons from 2015 to 2017 and corresponding cities in the two urban agglomerations of Northeast China

Season	Spring		Summer		Fall		Winter	
	Max/City	Min/City	Max/City	Min/City	Max/City	Min/City	Max/City	Min/City
$PM_{2.5}$	54.9/SY	30.3/SH	36.6/AS	14.6/YB	66.4/HB	32.5/YB	98.1/HB	49.2/SH
PM_{10}	112.6/SY	59.6/YB	60.2/SP	26.7/YB	91.3/SY	49.5/YB	125.7/HB	66.4/YB
SO_2	33.4/SY	11.6/SYJ	13.7/SY	4.79/SYJ	35.0/SY	11.0/SH	101.8/SY	25.9/DQ
NO_2	40.8/HB	19.7/SYJ	34.3/CC	15.7/SYJ	43.1/SY	23.3/QH	62.0/HB	29.0/BX
O_3-8h	138.2/JL	85.8/SH	149.3/SY	94.7/MDJ	89.9/LY	57.3/QH	57.7/JL	50.9/QH
CO	1.35/BX	0.59/DQ	1.23/AS	0.33/SH	1.29/AS	0.62/SH	1.87/BX	0.90/DQ

Notes: Units of all air pollutants are $\mu g/m^3$ except for CO with the unit of mg/m^3. SY, AS, FS, BX, CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively

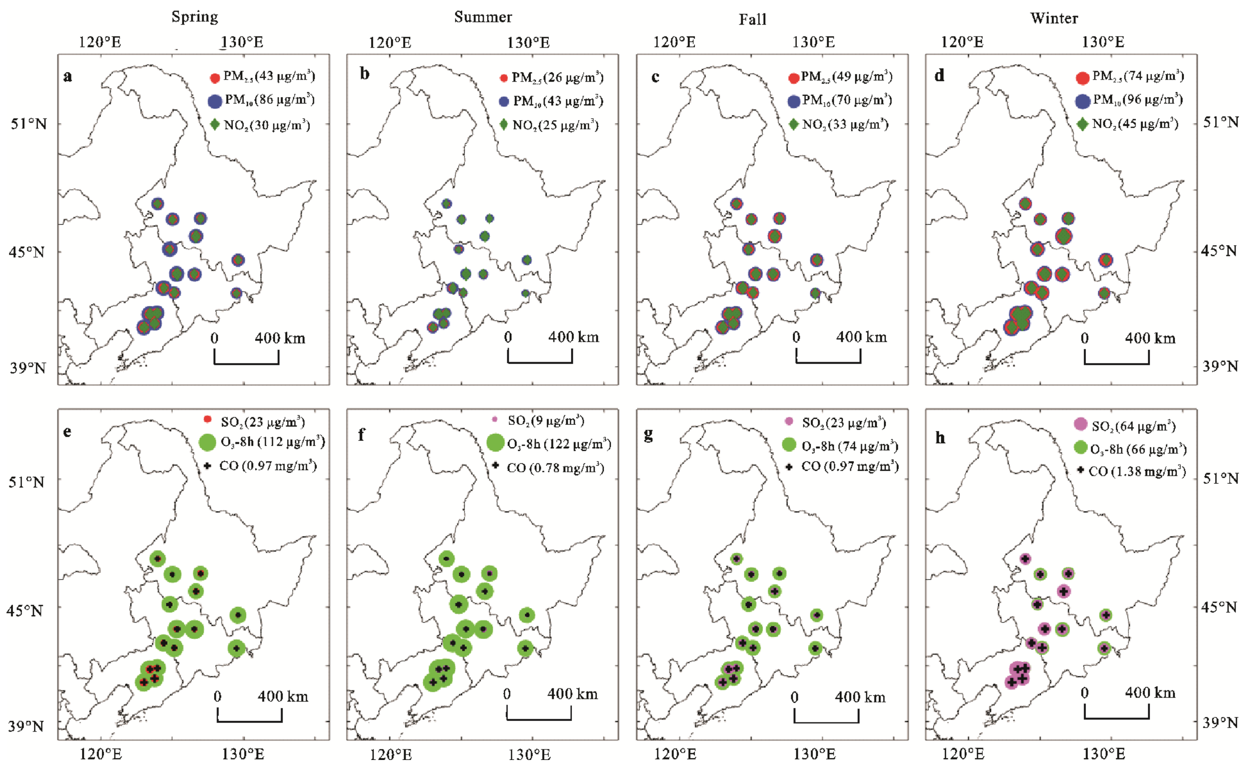

Fig. 4 Spatial distributions of mass concentrations of $PM_{2.5}$, PM_{10} and NO_2 averaged in (a) spring, (b) summer, (c) fall, and (d) winter of 2015 to 2017, and those of SO_2, daily maximum O_3-8h and CO in different seasons (e–h) in city clusters of Northeast China. The reference values of each air pollutant shown in the right upper corner represent the average of maximum and minimum values. SY, AS, FS, BX, CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively

Shenyang had the largest SO_2 concentration in all seasons, which indicates the great influence of local emission of SO_2. Besides terrible SO_2 pollution, CLUA was also characterized by high CO, with the seasonal maximum CO concentration observed in Benxi in spring and winter, and in Anshan in summer and fall (Fig. 4). The above analysis suggested that compound air pollution characteristic existed in CLUA and megacities in HCUA with simultaneously high concentrations of $PM_{2.5}$ and O_3 due to joint effects of more primary emissions.

3.1.3 Diurnal variation of air pollutants

Diurnal variations of air pollutant concentrations at each city averaged from 2015 to 2017 are showed in Fig. 5. The hourly mean O_3 concentration was used here instead of the daily maximum O_3-8h. The O_3 at all cities showed the highest value in middle of the day, while the other pollutants generally exhibited two peaks in one day, with the first one occurring in the period of 07:00–10:00 LT and the second around 18:00–20:00 LT, basically corresponding to the morning and evening rush hours. Moreover, the change in atmospheric boundary layer (ABL) also influences the diurnal variation of air pollutants. It should note that such diurnal variations of air pollutants among some cities showed subtle difference. First, as a background city, Yanbian had lower concentrations than other cities at most of time in a day. Especially during nighttime, pollutant concentrations in Yanbian declined faster than that in other cities, mainly due to less pollutant emissions and its local topography. Second, the first peak of SO_2 in Shenyang and Changchun occurred 1–2 h earlier than that in other cities, which was observed in all seasons (figure not shown). This should be related to stable SO_2 emissions like industry emissions in Shenyang and Changchun because the peaks of other pollutant concentrations at different cities were consistent. Third, SO_2 and NO_2 concentrations in Shenyang were higher than that in other cities of Liaoning Province at all hours except for the period of 10:00–16:00 LT (Fig. 5c), partly indicating the dilution of surface pollutants by the increase of ABL height during daytime. At last, CO concentration in Shenyang was obviously lower than that in other cities of CLUA, partly due to more biomass burning for heating and cooking in small cities. Similarly, CO in Changchun was lower than most other cities in Jilin Province; while Harbin was exceptional because it had larger rural areas and cropland (Fig. 5f).

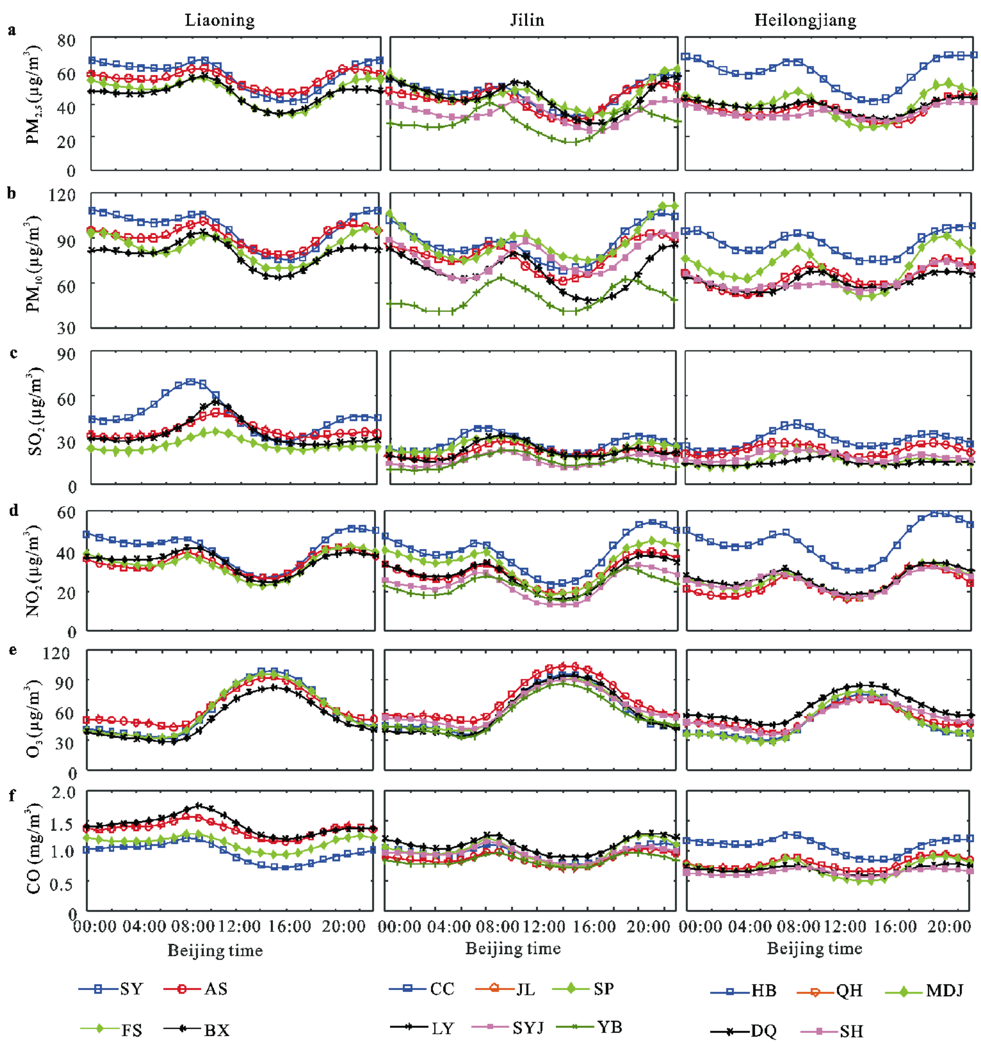

Fig. 5 Diurnal variations of mass concentrations of (a) $PM_{2.5}$, (b) PM_{10}, (c) SO_2, (d) NO_2, (e) O_3, and (f) CO averaged from 2015 to 2017 observed at cities in Liaoning , Jilin and Heilongjiang provinces. SY, AS, FS, BX, CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively

3.2 Regional transport of air pollutants between NEC city clusters

3.2.1 Dependence of $PM_{2.5}$ and O_3 on winds

The dependence of hourly mean O_3 in summer (represented by June 2017) and $PM_{2.5}$ in fall (represented by November 2017) on *WS* and *WD* at all cities are shown in Fig. 6 and Fig. 7, respectively. Wind roses (represented by the solid lines) indicated that the prevailing winds at most cities originated from the south and the north during June and November due to the effect of East Asian monsoon and local topography, while at some cities in Heilongjiang Province such as Harbin, Mudanjiang and Suihua, the prevailing *WD* had a wide range.

As shown in Fig. 6, the distributions of the mean O_3 (dash-dot line) in June at most cities were quite even at different *WD*, which means the distribution of O_3 concentration was almost independent on *WD*. The highest O_3 at all cities occurred in the presence of *WS* > 3 m/s and was also independent on *WD*. Xu et al. (2011b) observed that low O_3 concentrations occurred in presence of slow winds for all *WD*s at Wuqing, a suburban site in North China Plain, which because high NO accumulation consumed O_3 through chemical reactions under calm wind conditions. Liu et al. (2019) calculated that the correlation coefficient between hourly mean O_3 and *WS* was 0.49 approximately at Shenyang during summers from 2012 to 2015, which indicated that O_3 can be transformed by its precursors and transported along the prevailing wind transport path. Overall, the highest O_3 in the CLUA was much higher than that in the HCUA and gradually decreased at cities from the south to the north.

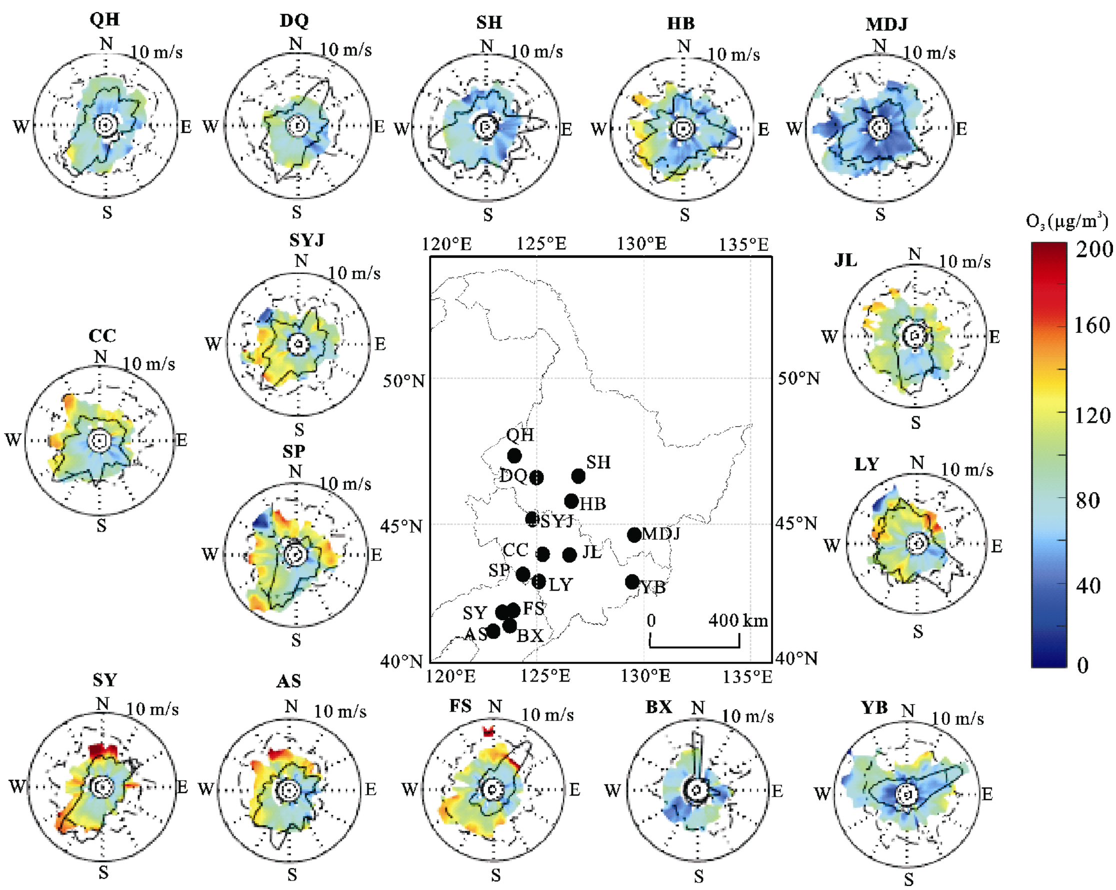

Fig. 6 Wind speed (*WS*) and direction (*WD*) dependence of hourly mean O_3 concentration during June 2017 at different cities of Northeast China. In each picture, the shaded contours indicate the hourly mean O_3 concentration for different *WS*s (radial direction) and *WD*s (transverse direction). The dash-dot lines indicate the relative mean values at each *WD*, and the solid lines represent a wind rose. SY, AS, FS, BX, CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively

Compared with O_3, the distribution of mean $PM_{2.5}$ in November 2017 at all cities was dependent on both of *WS* and *WD*, and the highest $PM_{2.5}$ at cities in the HCUA was larger than that in the CLUA (Fig. 7). The highest $PM_{2.5}$ concentration at cities in the northern part of HCUA (mainly refer to Heilongjiang Province) mostly occurred in presence of calm winds (*WS* < 2 m/s) at all *WD*s, and sometimes also occurred when winds were strong, especially southerly winds. For instance, at Harbin, the highest $PM_{2.5}$ (hourly mean value > 1500 $\mu g/m^3$) at the N-NE directions occurred as winds were weak, mainly due to large local emissions caused by agricultural residue burning activities (Fig. 8). While in the S direction, the highest $PM_{2.5}$ at Harbin was accompanied by extremely strong winds (*WS* > 9 m/s approximately) and apparently associated with pollutant transport from upwind areas. In contrast to the northern region of HCUA, the highest $PM_{2.5}$ in the southern HCUA region (mainly refer to Jilin Province) and in CLUA mostly occurred in presence of strong winds at the N-NE directions.

3.2.2 Inter-regional transport of air pollutants during a severe haze event

A wide area of agricultural residue burning occurred during November 1–4, 2017 in NEC, resulting in a severe regional and persistent haze event in NEC urban agglomerations. Massive fire points during this haze event have been captured by the MODIS satellite, as shown in Fig. 8; more fire points in HCUA and less in CLUA and most are located in areas with cropland fraction larger than 0.8, which confirmed the activity of crop residues burning. It should note that some fire points can

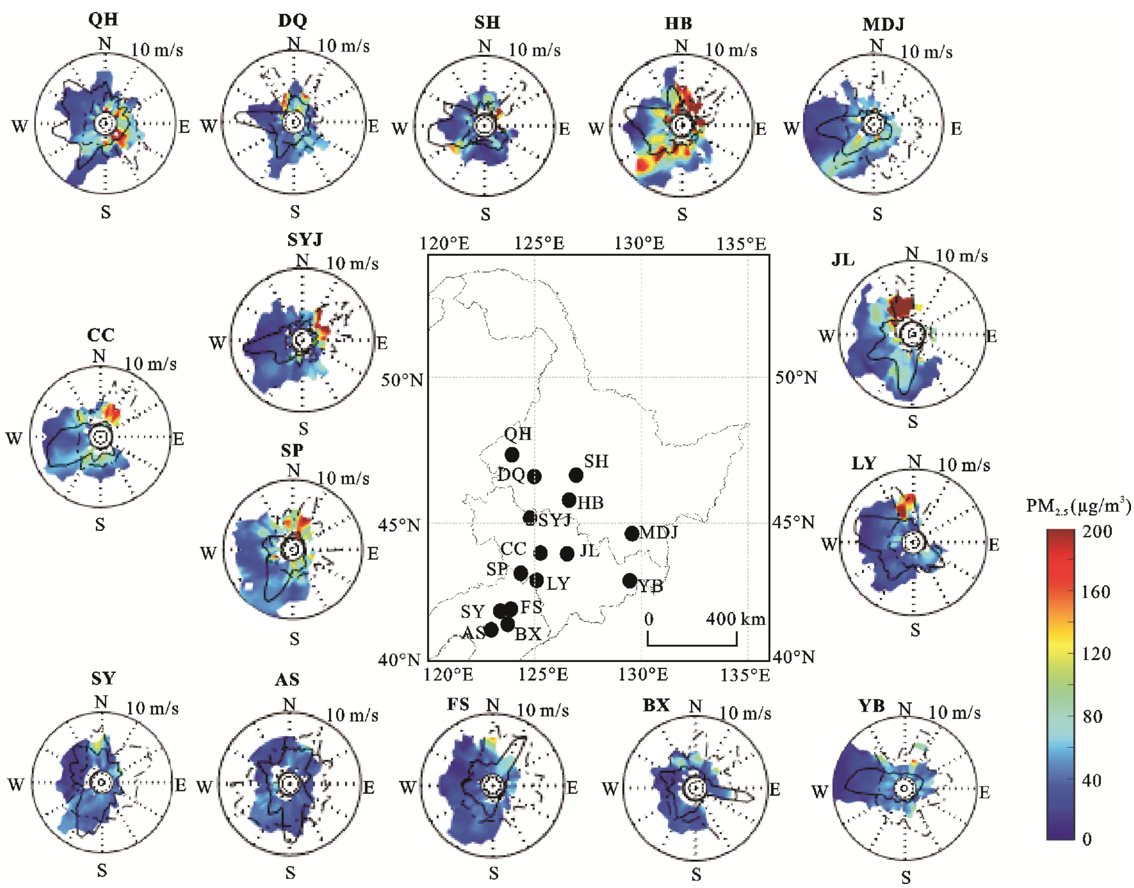

Fig. 7 Wind speed (*WS*) and direction (*WD*) dependence of hourly mean $PM_{2.5}$ concentration during November 2017 at different cities of Northeast China. The shaded contours indicate the hourly mean $PM_{2.5}$ concentration for different *WS*s (radial direction) and *WD*s (transverse direction). The dash-dot lines indicate the relative mean values at each *WD*, and the solid lines represent a wind rose. SY, AS, FS, BX, CC, JL, SP, LY, SYJ, YB, HB, QH, MDJ, DQ and SH are the abbreviations of Shenyang, Anshan, Fushun, and Benxi, Changchun, Jilin, Siping, Liaoyuan, Songyuan, and Yanbian Korean Autonomous Prefecture, Harbin, Qiqihar, Mudanjiang, Daqing, and Suihua, respectively

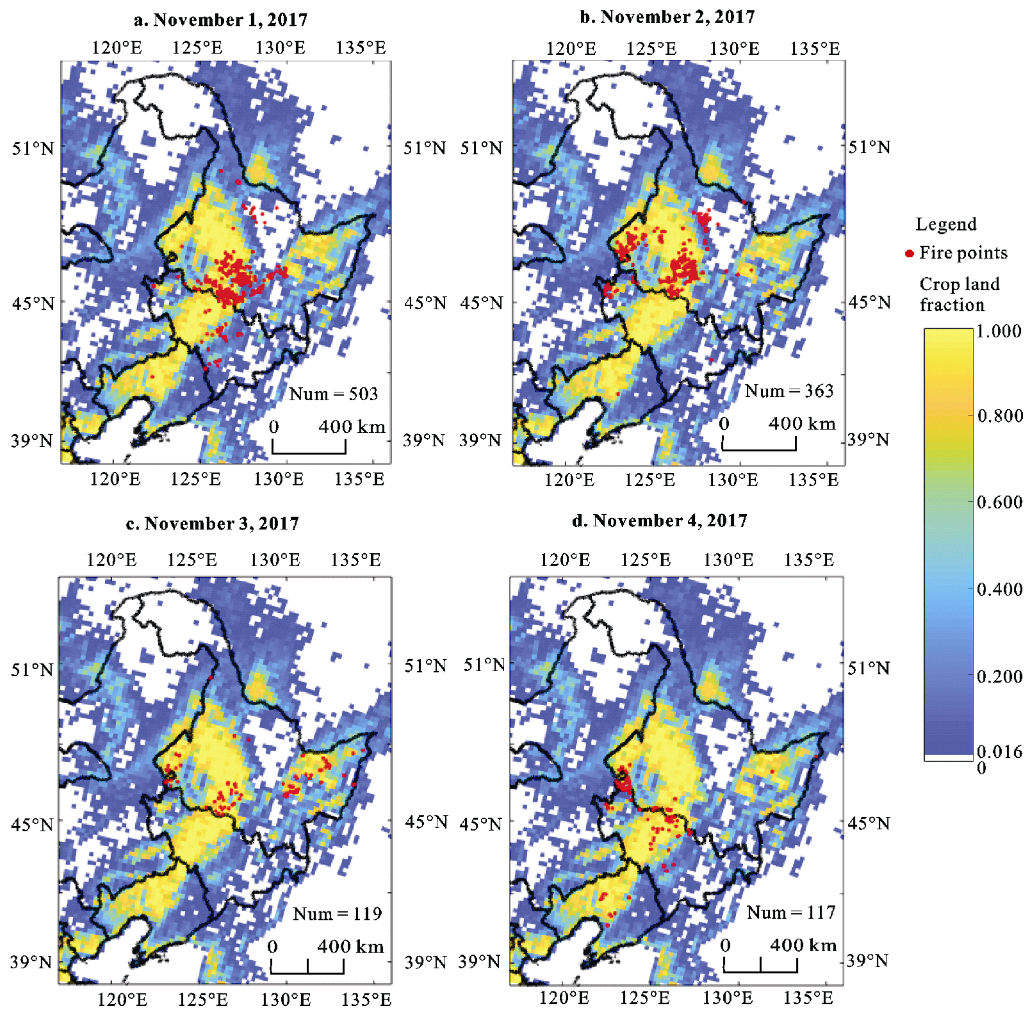

Fig. 8 Distribution of fire points and cropland fraction in Northeast China on (a) November 1, (b) November 2, (c) November 3, and (d) November 4, 2017 retrieved from the MODIS satellite. The total number (Num) of fire points on each day is also shown in each panel

not be detected due to clouds covering over Jilin and Liaoning provinces on November 2 and Heilongjiang Province on November 4. Fig. 9 shows temporal variations of hourly mean $PM_{2.5}$ concentration and some meteorological parameters including *WS* and *WD* at 10 m height, *RH* and T_a at 2 m height observed at meteorological stations in Harbin, Changchun and Shenyang during the hazy episode.

In Fig. 9a, $PM_{2.5}$ at Harbin increased significantly after 20:00 LT each day from November 1 to 4, 2017. This is because corp residue burning activities are prohibited strictly by local government for reducing air pollution, and farmers had to do this at late night to avoid supervision of local environmental protection department. The highest $PM_{2.5}$ at Harbin occurred 01:00 LT on November 2, exceeding 1500 μg/m^3 and accompanied by relatively small southerly wind (*WS* < 4 m/s). At the same time, a $PM_{2.5}$ peak was also observed in Changchun, but much lower than that in Harbin (Fig. 9b). Several hours later, $PM_{2.5}$ at Shenyang increased significantly, and the same phenomenon occurred on November 3 when strong northerly flows (*WS* > 8 m/s) controlled all cities (Fig. 9c).

During the haze event, daily T_a at all cities showed a decreasing trend. In Harbin, T_a had an abnormal small peak around 00:00 LT on November 2 due to the extreme high aerosol concentrations in the atmosphere and heating from crop residue burning. The values of *RH* changed oppositely with the variation of T_a. When $PM_{2.5}$ was high, *RH* at Harbin and Changchun was usually smaller than 70%, while that at Shenyang was mostly larger than 80%.

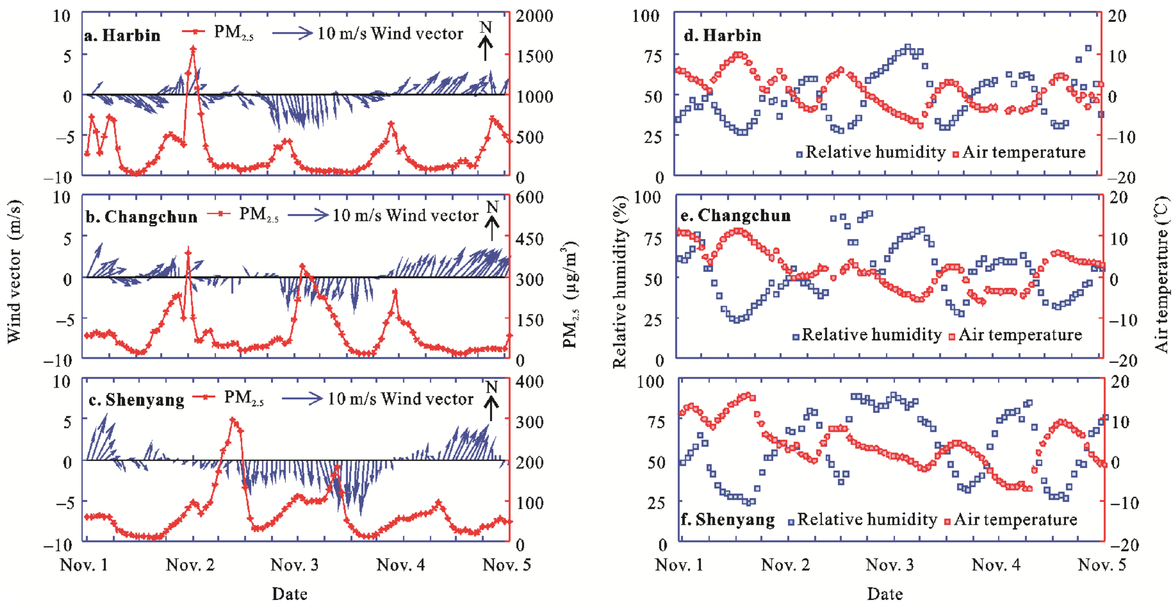

Fig. 9 Temporal variation of hourly mean $PM_{2.5}$ concentration and wind vector at 10 m height at meteorological stations in (a) Harbin, (b) Changchun and (c) Shenyang, and (d–f) relative humidity and air temperature on November 1–4, 2017

Wind field at 10 m height was retrieved from the ECMWF reanalysis data and overlaid by $PM_{2.5}$ concentration at different cities in NEC (Fig. 10), to clearly examine the inter-regional transport of air pollutants during this haze event. At 20:00 LT on November 1 (Fig. 10a), $PM_{2.5}$ concentration was higher in cities of Heilongjiang Province, followed by Jilin Province and least in CLUA. The cities with $PM_{2.5} > 200$ μg/m^3 included Changchun, Jilin and Harbin. Because of wind convergence in the southern region of Heilongjiang and in CLUA, $PM_{2.5}$ at Harbin and Suihua (in Heilongjiang Province) sharply increased to 1543 and 866 μg/m^3 till to 02:00 LT on November 2, and that in CLUA also increased significantly (Fig. 10b). After that, *WS* continually increased over the whole NEC region, and $PM_{2.5}$ in Heilongjiang Province and in the western region of Jilin Province decreased significantly from 08:00 to 14:00 LT on November 2 due to strong westerly flows. In contrast, $PM_{2.5}$ in CLUA continually increased because strong northerly flows carried massive air pollutants from HCUA to this region (Figs. 10c, 10d). This case clearly revealed the regional transport of air pollutants among cities in NEC urban agglomerations, which suggests that the inter-regional prevention and control of air pollutants are essential to atmospheric environmental management.

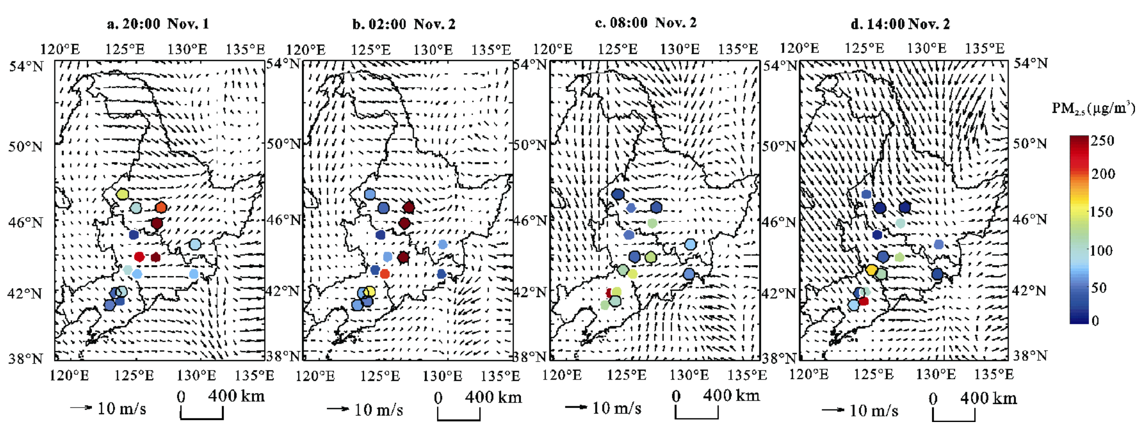

Fig. 10 Wind fields at 10 m height retrieved from the European Centre for Medium Range Weather Forecasts (ECMWF) reanalysis data and $PM_{2.5}$ mass concentration in NEC urban agglomerations at (a) 20:00 LT on November 1, (b) 02:00 LT, (c) 08:00 LT and (d) 14:00 LT on November 2, 2017

4 Discussion

4.1 Effective control options to reduce local emissions

Strict emission control measures have been launched by local governments in NEC in recent years, in order to improve air quality and reduce extreme air pollution events. These control measures include prohibiting crop residues burning outdoors, improving central heating systems, adjusting energy structure such as using gas instead of coal, *etc*.. Taken Shenyang as an example, the proportion of clean energy heating area is supposed to increase up to 81% of the total domestic heating area of 4.02×10^{10} km^2 in 2020 according to a clean energy heating plan released by the Liaoning Province government. These control policies can effectively reduce emissions of fine particles and gaseous pollutants such as SO_2, CO and NO_2 in NEC urban agglomerations. Chen et al. (2018) suggested that making pollution control policies should consider the dependence of primary emission sources on seasons in NEC. The primary emission sources in late-fall are crop residue burning, coal burning in winter, and dust emission in spring. Therefore, the most efficient ways to reduce severe air pollution events in different seasons are probably the complete prohibition of crop residues burning, improving combustion efficiency and reducing the emission of pollutant from coal burning based on technological improvement, as well as controlling major anthropogenic dust emission sources (mainly due to road dust emissions and soil tilling-induced dust emissions) (Chen et al., 2018).

Although PM and SO_2 pollution has been alleviated nationwide during the past decade, O_3 pollution has worsen in major city clusters in China, including the CLUA and HCUA. According to monitoring results from 74 Chinese cities, the mean daily 8-h maximum concentrations of O_3 increased from approximately 69.5 ppbv (~139 μg/m^3) to 75.0 ppbv (~150 μg/m^3) from 2013 to 2015, while the percentage of non-compliant cities increased from 23% to 38% (Wang et al., 2017). The elevated O_3 levels have been reported to have adverse impacts on agricultural crops (Feng et al., 2015), forest (Li P et al., 2018), and human health (Brauer et al., 2016).

To reduce O_3 pollution in urban agglomerations, the emissions of volatile organic compounds (VOCs) and oxides of nitrogen (NO_x) should be controlled, because the ground-level O_3 in urban atmospheres is typically formed through a series of free radical reactions involving VOCs and NO_x in the presence of sunlight (Haagen-Smit et al., 1953). NO_x emissions in major developed regions in China have decreased since 2011 due to implementation of NO_x control in China's 12th Year Plan (2011–2015) and a slowdown in manufacturing activities (Wang et al., 2017), while the emission of VOCs has continuously increased in mainland China since the 1980s (Lu et al., 2013). Among VOCs, reactive aromatics are found to be the predominant contributor to O_3 formation in many urban areas in China (Xue et al., 2014), and these compounds are mainly from solvent use and vehicle exhausts (Guo et al., 2006). Therefore, control measures targeting solvent use along with vehicular emissions are advised to reduce O_3 pollution (Wang et al., 2017).

4.2 Coping with regional air pollution events

Long-range transport has been shown to be an important cause of regional air pollution (including high PM and O_3) events in NEC Plain. For example, Miao et al. (2018a) reported that the cross-boundary transport of aerosols from the North China Plain contributed approximately 20% of the near-surface $PM_{2.5}$ during a heavy haze event on December 2–3, 2016 in Shenyang, which was caused by the nocturnal low-level jets and convective turbulence at daytime (Li et al., 2019). This implicated that control of emissions in one city cluster is not sufficient to reduce regional air pollution events, and joint efforts among city clusters are essential (Wang et al., 2014).

In order to cope with regional air pollution throughout city clusters, it is necessary to establish a joint prevention and control system, which requires closing cooperation among governments, meteorological offices, and environment protection administrations in different regions. Once heavy air pollution occurs in some region, the down-stream regions should pay attention to meteorological conditions and control local emissions in time. Additionally, it is also important to carry on joint observational experiments over large areas for better understanding the formation mechanisms of regional heavy air pollution.

5 Conclusion

The spatiotemporal characteristics of the six criteria air pollutants at 15 cities in the two urban agglomerations CLUA and HCUA, Northeast China are investigated using their real-time measurements of hourly mean mass concentrations from 2015 to 2017, and their relationships with meteorological conditions were studied based on meteorological data from surface meteorological stations and the ECMWF reanalysis data.

The mean concentrations of the six air pollutants in CLUA were higher than that in HCUA. Provincial capital cities (Shenyang, Harbin and Changchun) had more severe air pollution than other cities in NEC. The highest $PM_{2.5}$ value averaged from 2015 to 2017 was observed at Harbin ((58.3 ± 74.1) μg/m^3), the highest PM_{10} at Shenyang ((96.2 ± 81.8) μg/m^3), CO at Benxi ((1.42 ± 0.73) mg/m^3), NO_2 at Harbin ((44.3 ± 24.9) μg/m^3), and O_3 at Jilin ((70.6 ± 39.7) μg/m^3), respectively. The cities with low pollutant levels were mostly located in the surrounding areas of HCUA, such as Suihua and Yanbian. Air pollutants (except for O_3) at cities in NEC regularly exhibited an annual variation, with the largest concentrations in winter and the lowest in summer, and their diurnal variations were generally characterized by two peaks, corresponding to rush hours at 07:00–10:00 LT and 18:00–20:00 LT, respectively. O_3 varied oppositely with other pollutants because it is formed from photochemical reactions in the presence of heat and sunlight. Both of the annual mean PM and SO_2 decreased from 2015 to 2017, particularly in CLUA, while O_3 had a slightly increasing trend.

Air pollutant concentrations are strongly dependent on meteorological conditions. The dependence of $PM_{2.5}$ in fall and O_3 in summer at all cities on wind speed and wind direction are comparatively analyzed. The highest O_3 at all cities occurred in presence of strong winds (WS > 3 m/s) and was independent on WD. While the relationship between the highest $PM_{2.5}$ and winds depended on regions. In the northern HCUA such as Harbin, the highest $PM_{2.5}$ mostly occurred in presence of calm winds (WS < 2 m/s) at all WDs, and sometimes also occurred when southerly winds were strong; In the southern HCUA and in CLUA, besides accompanied by weak winds, the highest $PM_{2.5}$ mostly occurred as the northerly and northeasterly flows were strong.

The inter-regional transport of air pollutants between CLUA and HCUA was clearly examined during a severe regional haze event on November 1–4, 2017, which was caused by a wide area of crop residue burning in NEC. The burning activities concentrated in HCUA at nighttime, therefore several $PM_{2.5}$ peaks were observed in Harbin and Changchun at nighttime each day during the haze episode. However, the $PM_{2.5}$ peaks at Shenyang usually occurred 6–10 hours later than that in Harbin and Changchun. Wind (WS < 4 m/s) convergence in pollutant emission regions (HCUA) favored rapid accumulation of air pollutants, and then after several hours, strong northerly winds transported massive pollutants to CLUA.

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