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Regional Characteristics and Causes of Haze Events in Northeast China

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Abstract: Northeast China experiences severe atmospheric pollution, with an increasing occurrence of heavy haze episodes. However, the underlying forces driving haze formation during different seasons are poorly understood. In this study, we explored the spatio-temporal characteristics and causes of haze events in Northeast China by combining a range of data sources (i.e., ground monitoring, satellite-based products, and meteorological products). It was found that the 'Shenyang-Changchun-Harbin (SCH)'city belt was the most polluted area in the region on an annual scale. The spatial distribution of air quality index (AQI) values had a clear seasonality, with the worst pollution occurring in winter, an approximately oval-shaped polluted area around western Jilin Province in spring, and the best air quality occurring in summer and most of the autumn. The three periods that typically experienced intense haze events were Period I from mid-October to mid-November (i.e., late autumn and early winter), Period II from late-December to February (i.e., the coldest time in winter), and Period III from April to mid-May (i.e., spring). During Period I, strong $PM_{2.5}$ emissions from seasonal crop residue burning and coal burning for winter heating were the dominant reasons for the occurrence of extreme haze events (AQI > 300). Period II had frequent heavy haze events (200 < AQI < 300) in the coldest months of January and February, which were due to high $PM_{2.5}$ emissions from coal burning and vehicle fuel consumption, a lower atmospheric boundary layer, and stagnant atmospheric conditions. Haze events in Period III, with high PM_{10} concentrations, were primarily caused by the regional transportation of windblown dust from degraded grassland in central Inner Mongolia and bare soil in western Jilin Province. Local agricultural tilling could also release PM_{10} and enhance the levels of windblown dust from tilled soil. Better control of coal burning, fuel consumption, and crop residue burning in winter and autumn is urgently needed to address the haze problem in Northeast China.

Keywords: air quality; PM_{10}; $PM_{2.5}$; dust; agricultural activity; coal burning; fuel consumption

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1 Introduction

Regional atmospheric pollution has generated widespread concern in China (Xu et al., 2013). Frequent and increasingly heavy haze events have caused serious harm to human health and the environment, and thus affect the sustainable development of the regional economy (He et al., 2002; Chan and Yao, 2008; Heal et al., 2012). Understanding the spatio-temporal characteristics, pollution sources, and underlying mechanisms of

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these complex regional atmospheric pollution events would help to identify key emission sources and to support regional joint prevention and control measures.

Northeast China experiences high levels of atmospheric pollution, with an increasing occurrence of heavy haze episodes in recent years (Chen et al., 2017). Air quality index (AQI) values in the major cities in the region (e.g., Harbin, Changchun, and Shenyang) during autumn and winter are often the highest of any location in the country (http://www.zhb.gov.cn/gkml/hbb/qt/201602/t20160204_329886.htm). However, efforts to control air pollution in the region have not been as effective as those in the Beijing-Tianjin-Hebei region, the Pearl River Delta, and the Yangtze River Delta regions. In the 'Air Pollution Control Action Plan' (Atmosphere 10), there were no explicit requirements established to control coal use and reduce Particulate Matter 2.5 ($PM_{2.5}$) in Northeast China (http://www.gov.cn/zwgk/2013-09/12/content_2486773.htm). Most provinces or cities have achieved or exceeded the anticipated medium-term goals set in 'Atmosphere 10', but annual Particulate Matter 10 (PM_{10}) concentrations in the provinces of Northeast China have still increased (http://www.gov.cn/xinwen/2016-07/06/content_5088795.htm). Although annual $PM_{2.5}$ concentrations in the 40 cities at prefecture level and above (among them there are one prefecture, one autonomous prefecture, and one league, hereafter referred to as 40 cities) in Northeast China in 2015 have reduced by 12.5% compared to those in 2013, the extent of the reduction was lower than the average (23.6%) of 74 nationally important cities (hereafter referred to as 74 cities), in which new air quality standards (GB 3095-2012) were firstly implemented (http://www.zhb.gov.cn/gkml/hbb/bwj/201203/t20120302_224147.htm). Furthermore, the average extent of the $PM_{2.5}$ reduction in provincial capitals (i.e., Shenyang, Changchun, and Harbin) was only 1.7%. On an annual scale, the proportion of days experiencing heavy pollution in the 40 cities of Northeast China was equal to the national average (3.2%), but the corresponding values in late autumn and early winter (from September 20 to November 20) increased by up to 7%–8% based on our data analysis. There is an urgent need to reduce particulate matter (PM) concentrations and eliminate heavy haze episodes in autumn and winter to achieve the goals of 'Atmosphere 10' in Northeast China.

Compared with other regions, Northeast China has unique characteristics in terms of its geographical location, climatic conditions, industrial base, and economic development. As a traditional old industrial base, important breadbasket (i.e., 30% of the total land area is agricultural), and the area of the country with the longest central-heating season in buildings (i.e., 4–6 months) (National Agricultural Work Conference, 2013; National Bureau of Statistics of the People's Republic China, 2013), the sources of regional atmospheric pollutants are diverse and complex. The regional topography is characterized by the so-called 'three mountains with plains'. The western, northern, and eastern borders of the region are delimited by the Da Hinggan Mountains, the Xiao Hinggan Mountains, and the Changbai Mountains, respectively, whereas the south is a flat area adjacent to the Beijing-Tianjin-Hebei region, which is a possible source of atmospheric pollutants for Northeast China. Thus, there is a range of emission sources, pollutant components, and haze formation mechanisms in Northeast China. Previous studies of the atmospheric environment in Northeast China have mainly included surface Particulate Matter (PM) concentration monitoring, the analysis of pollutant physicochemical properties, source apportionment in the central cities of the Liaoning and Jilin provinces, assessments of agricultural PM emissions, and numeric simulations of air quality (Huang et al., 2010; Liu, 2015; Long et al., 2016; Fang et al., 2017). However, these studies have not been able to determine the physico-chemical mechanisms involved in the formation of heavy haze events, and thus a full understanding of regional atmospheric pollution and the required governance has not been achieved.

In this study, we analyzed the spatio-temporal characteristics of air quality and explored the possible underlying forces driving haze formation during different seasons in Northeast China. A diverse range of data sources were used, including the local ground-monitored AQI and gaseous pollutant concentrations, satellite-based aerosol optical depth (AOD) and fire-point products from National Aeronautics and Space Administration (NASA), and regional meteorological data from National Oceanic and Atmospheric Administration (NOAA). The aim was to provide a basic understanding of haze events at the regional scale and the inter-regional mechanisms responsible for their prevention, enabling a determination of the control measures required to meet the needs of 'Atmosphere 10'.

2 Methodology

2.1 Study area

The geographical region of Northeast China consists of the three provinces of Liaoning, Jilin, and Heilongjiang, and the eastern part of Inner Mongolia Autonomous Region (i.e., Tongliao City, Chifeng City, Hulun Buir City, and Hinggan League) (Fig. 1). The region is separated from the Russian Far East to the north largely by the Heilong, Ergun, and Ussuri rivers, from North Korea to the south by the Yalu and Tumen rivers, and from the Inner Mongolian Autonomous Region to the west. The heartland of the region is the northeastern China Plain (40°25′N–48°40′N, 118°40′E–128°00′E), which extends north to the crown of the 'Chinese rooster', near where the Da Hinggan Mountains and Xiao Hinggan Mountains converge. The Changbai Mountains in the east separate China from the Korean peninsula. This topography is referred to as 'three mountains with plains'. The Northeast China Plain is a major breadbasket region, with a total area of 3.5×10^5 km^2, and is one of the three black earth terrains in the world. The growing season in this area generally runs from May to September, while the non-growing season lasts more than half a year from October to the following April. The conventional planting pattern is single cropping, with staple crops of corn, soybean, rice, and wheat. Due to the severely cold climate, with the lowest monthly temperatures in the range of –12℃ to –19℃, the winter heating season lasts for 4–6 months. Northeast China has a population of about 0.12 billion people, accounting for 8% of China's total population. In addition, Northeast China is a major industrial base of the country. However, in recent years, Northeast China's heavy-industry-based economy has stagnated, as China's economy continues to liberalize and privatize. The government has initiated the 'Revitalize the Northeast' campaign to counter this problem, and established the northeastern Summitto improve policy coordination and integration.

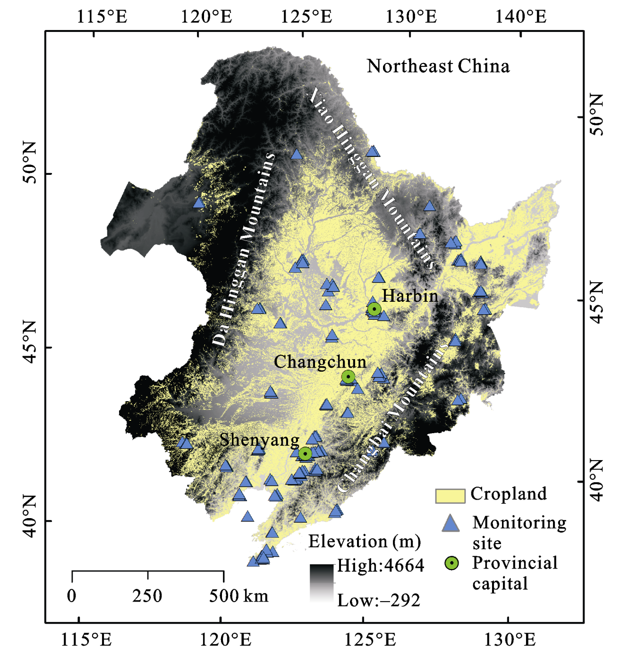

Fig. 1 Regional topography and atmospheric monitoring sites in Northeast China

2.2 Data sources and processing

2.2.1 Ground monitoring data for atmospheric pollutants

Ground-monitored hourly AQI values and standard atmospheric pollutant (i.e., PM_{10}, $PM_{2.5}$, O_3, SO_2, NO_2, and CO) concentrations in 40 cities during 2014–2015 were obtained from the Qingyue Open Environmental Data Center (https://data.epmap.org/) (Fig. 1). In each city, there were three to twelve monitoring sites distributed in various urban areas, and there were 183 monitoring sites in total. The average AQI values and atmospheric pollutant concentrations at these sites were used as the representative value for each city.

Annual and seasonal AQI values for all cities in 2015 were spatially interpolated to provide a regional distribution for Northeast China using Cokriging interpolation method. Based on local climate characteristics, spring consists of the months of April and May; summer is June, July, and August; autumn is September and October; and winter lasts from November to the following March. Data from 11 major cities (i.e., Shenyang, Fushun, Dandong, and Huludao in Liaoning Province; Harbin, Qiqihar, Mudanjiang, and Daqing in Heilongjiang Province; Changchun and Jilin in Jilin Province; and Chifeng in Inner Mongolia) were used to analyze the temporal variation of the AQI in 2014 and 2015 at a regional scale. The AQI and atmospheric pollutant concentrations in provincial capitals (i.e., Shenyang, Changchun, and Harbin) were used in an analysis of the causes of haze events in Northeast China.

2.2.2 Aerosol optimal depth

The AOD products used in the study were the daily combined dark target and deep blue AOD at 0.55 μm for land and ocean, which are observed and processed based on the Moderate Resolution Imaging Spectroradiometer (MODIS) carried in the Terra and Aqua satellite. The resolution of AOD products is 0.5° × 0.5°. These data were retrieved from the NASA Giovanni website (http://giovanni.sci.gsfc.nasa.gov/giovanni).

The MODIS AOD products were used to confirm the feasibility of monitoring atmospheric pollutants and to quantitatively evaluate regional air quality classes. Regional AOD data were defined by the boundary of Northeast China. The differences between each haze period were analyzed using the average daily AOD data during the pre-haze, haze, and post-haze periods.

2.2.3 Satellite-based fire points

Fire point information could be acquired from the Fire Information for Resource Management System (FIRMS) https://firms.modaps.eosdis.nasa.gov/download/request.php), which is also processed from MODIS Terra and Aqua. The fire point products provide the latitude and longitude of observed fire information during the given time period.

As with the AOD, regional fire point information was defined by the boundary of Northeast China. The fire point distributions during the pre-burning, burning, and post-burning periods were separated to identify their contribution to haze periods.

2.2.4 Vertical profile of aerosol subtypes

High-resolution vertical profiles of aerosols and their corresponding subtypes (i.e., clean marine, dust, polluted continental, clean continental, polluted dust and smoke) were provided by Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) (https://www-calipso.larc.nasa.gov/tools/data_avail/). CALIPSO is a joint NASA (USA) and Centre National d'Etudes Spatiales (CNES) (France) environmental satellite, built in the Cannes Mandelieu Space Center, which was launched atop a Delta II rocket on April 28, 2006. CALIPSO is part of the 'A Train', flying in formation with several other satellites (Aqua, Aura, and CloudSat). Therefore, we can compare or combine the observed data collected at similar times and locations from these satellites, which will improve our understanding of the horizontal and vertical distribution of different aerosols.

During haze periods, we collected vertical aerosol subtype data from CALIPSO when the satellite was passing over Northeast China. These aerosol subtypes enabled the identification of major or possible emission sources, in combination with ground monitoring data for atmospheric pollutants.

2.2.5 Meteorological data

Daily meteorological indexes (e.g., air temperature, precipitation, visibility, relative humidity, wind speed, and wind direction) in each city during 2014–2015 were acquired from the State Meteorological Administration. Regional meteorological indexes (e.g., wind speed and pressure) during specified periods were acquired and plotted based on the National Centers for Environmental Prediction (NCEP) data analysis (http://www.esrl.noaa.gov/psd/data/histdata/). The meteorological data were used as ancillary information for haze formation during haze events.

2.3 Statistical analysis

Pearson correlations were obtained for the relations among the AQI and atmospheric pollutants on an annual, seasonal, and monthly scale or during haze periods. The significances of the differences in the AQI values or atmospheric pollutant concentrations during different periods were investigated using the independent-samples t-test. All statistical procedures and plotting were performed using the software SigmaPlot 10.0 (SPSS Inc., Chicago, IL, USA) and ArcGIS 10.2 (Esri, Redlands, CA, USA).

3 Results and Discussion

3.1 Overview of air quality

Compared to 2013, the mean annual concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, and CO of 40 cities in 2015 had decreased by 14.5%–30.6%, while O_3_1h (i.e., 1-hour average concentration) and O_3_8h (i.e., daily maximum 8-hour average concentration) concentrations had increased by 8.0% and 14.1%, respectively (Table 1). In 2013, only Harbin, Changchun, Shenyang, and Dalian had almost complete pollution records, while the four cities of Huludao, Panjin, Yingkou, and Dandong had data only for the period between September 30 and December 31 (91 days in total). In the provincial capitals, most of the data were complete for the autumn and winter seasons. These would lead to an overestimation of the mean concentrations in 2013 compared to 2015. According to the percentage content of each pollutant (Fig. 2), compared to 2013, the number of days classed as moderately polluted or worse decreased, while the number classed as good and moderate increased in

2015. The percentage of polluted days reduced from 30.8% (2013) to 21.2% (2015). However, the percentage of polluted days, especially heavily and severely polluted days, in the provincial capitals was significantly higher than that in prefecture-level cities, with little apparent improvement in the air quality. In contrast, air quality in the prefecture-level cities investigated here was much better in 2015.

The primary pollutant of concern in Northeast China was $PM_{2.5}$, followed by PM_{10}. The mean $PM_{2.5}$ concentration of the 40 cities (47.9 μg/m^3) was slightly lower than that of 74 cities in China (55.0 μg/m^3) (Mid-term Assessment Report of Air Pollution Prevention and Control Action Plan). The $PM_{2.5}$ levels in 2015 were reduced by 23.4% compared to the concentration in 2013, which was similar to the reduction observed for the 74 important cities (23.6%), as shown in the mid-term assessment report. However, the annual mean $PM_{2.5}$ concentration (67.7 μg/m^3) in the provincial capitals (Shenyang, Changchun, and Harbin) of Northeast China was much higher than that in the 74 important cities (Chen et al., 2016), with little obvious improvement (rate of decrease was only 1.7%) from 2013 (68.9 μg/m^3) to 2015 (67.7 μg/m^3). In addition, the pollutant concentrations in the provincial capitals were all higher than those in prefecture-level cities, except for O_3.

Table 1 Air quality index (AQI) values and gaseous pollutant mean concentrations in all 40 cities, provincial capitals, and prefecture-level cities in Northeast China from 2013 to 2015

City	Year	AQI	$PM_{2.5}$ (μg/m^3)	PM_{10} (μg/m^3)	SO_2 (μg/m^3)	NO_2 (μg/m^3)	O_3_1h (μg/m^3)	O_3_8h (μg/m^3)	CO (mg/m^3)
NE	2013	95±60 (22–500)	63±56 (7–747)	103±70 (17–847)	44±45 (2–392)	40±19 (7–160)	83±46 (13–390)	69±34 (9–228)	1.1±0.7 (0.2–6)
	2014	88±49 (20–500)	56±45 (4–640)	96±59 (11–834)	41±38 (2–342)	36±17 (5–221)	94±43 (5–328)	82±39 (3–284)	1.2±0.7 (0.1–5.7)
	2015	79±50 (14–500)	48±44 (2–891)	80±59 (5–939)	28±31 (1–348)	28±16 (2–166)	90±42 (1–322)	79±38 (1–291)	1±0.6 (0–6.1)
PC	2013	102±69 (22–500)	69±65 (8–747)	113±79 (18–847)	47±51 (2–392)	44±20 (12–160)	81±40 (13–259)	72±38 (9–228)	1.1±0.6 (0.3–5.9)
	2014	104±63 (26–500)	70±59 (9–640)	115±71 (16–834)	55±54 (2–342)	49±18 (16–134)	93±44 (17–261)	81±40 (13–242)	1±0.4 (0.4–3.2)
	2015	103±70 (25–500)	68±67 (9–891)	106±84 (16–920)	44±52 (3–321)	46±19 (11–146)	91±46 (9–257)	80±43 (9–242)	1±0.5 (0.3–4.1)
PLC	2013	85±43 (23–302)	53±37 (7–252)	89±51 (17–324)	40±35 (3–258)	34±14 (7–108)	87±53 (14–390)	65±27 (10–211)	1.1±0.8 (0.2–6)
	2014	85±45 (20–500)	53±40 (4–606)	92±56 (11–782)	33±15 (5–221)	33±15 (5–221)	94±43 (5–328)	82±39 (3–284)	0.6±0.2 (0.2–1.1)
	2015	77±47 (14–500)	46±41 (2–755)	78±57 (5–939)	27±29 (1–348)	26±15 (2–166)	90±41 (1–322)	79±38 (1–291)	1±0.6 (0–6.1)

Notes: NE: Northeast China, i.e., all 40 cities; PC: provincial capital cities of Shenyang, Changchun, and Harbin; PLC: prefecture-level cities. The figures outside the parentheses are the mean and standard deviation of the corresponding cities and the figures inside the parentheses are their range

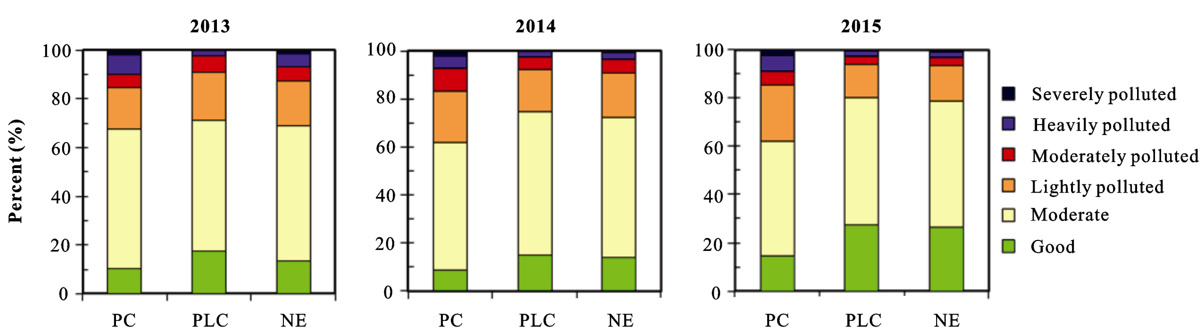

Fig. 2 Percentage of days with different pollution levels from 2013 to 2015 in Northeast China. NE: Northeast China, i.e., all 40 cities; PC: provincial capital cities of Shenyang, Changchun and Harbin; PLC: prefecture-level cities

3.2 Spatio-temporal characteristics of air quality

On an annual scale, high AQI values (> 80) were concentrated in the 'Shenyang-Changchun-Harbin (SCH)' city belt, covering three provincial cities and their surroundings (Fig. 3a), indicating that this was the most polluted area in Northeast China. Of these cities, Shenyang had the highest annual AQI value (103). The further those measurements were made from this area, the lower the AQI became. Air quality in cities (e.g., Tongliao, Siping, and Fuxin) around the trans-border region in the three provincial areas (Jilin, Liaoning, and Inner Mongolia) was worst in spring, with AQI values of 80–100 (Fig. 3b). Most of this region is degraded grassland, with saline soils. The best air quality in most regions was experienced in summer, with AQI values lower than 60, with only Tongliao and Baicheng having higher AQI values (60–80) (Fig. 3c). In autumn, air quality (AQI: 60–80) in cities on he northeastern Plain started to deteriorate due to crop residue burning and winter coal burning (Fig. 3d). Throughout the winter, the AQI values in Northeast China were greatly increased and the average values in the SCH region exceeded 100 (Fig. 3e). Harbin and Shenyang were the most polluted cities, with AQI values of 130 and 140, respectively, in winter. On a seasonal scale, air pollution levels in Northeast China followed the order of winter > spring > autumn > summer. The analysis revealed that annual AQI values in the cities had stronger correlations (n = 41, r^2 = 0.79, P < 0.01) with the winter values in Northeast China than with the values for the other seasons. In addition, the totals of the winter daily $PM_{2.5}$ concentrations as a proportion of the annual totals were 55% ± 7%, 64% ± 3%, and 53% ± 8% at the regional, provincial, and prefecture-level city scales in Northeast China, respectively, indicating the seriousness of wintertime air pollution in provincial cities.

From the daily AQI variations in 11 major cities in 2014 and 2015, three typical periods when heavy haze events occurred were identified in Northeast China (Fig. 4). The first period (Period I) was the most polluted period, and occurred in the late autumn and early winter from mid-October to mid-November. The

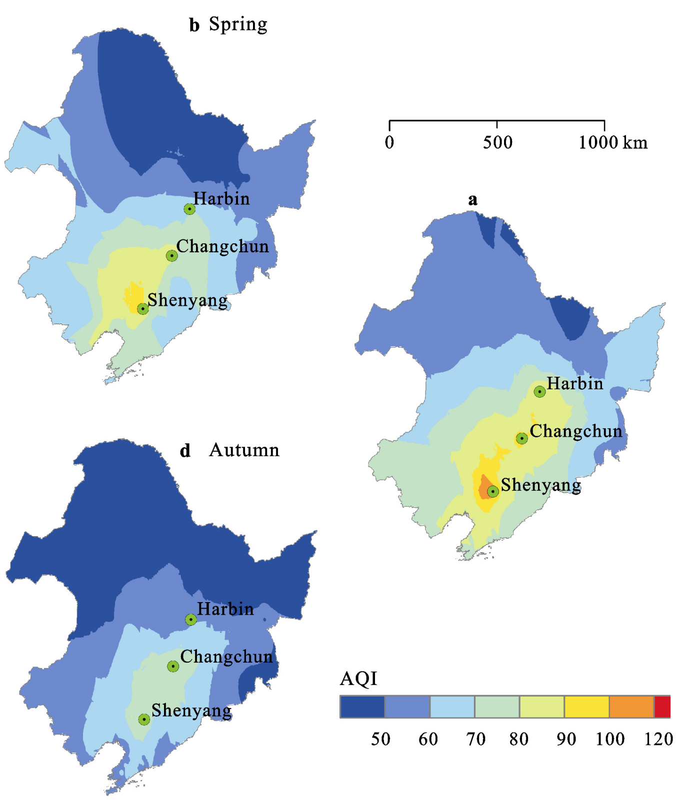

Fig. 3 Spatial characteristics of air quality index (AQI) in whole year (a), spring (April and May) (b), summer (from June to August) (c), autumn (September and October) (d), and winter (from November to next March) (e) in 2015

average frequencies of heavy haze events (AQI > 200) and extreme haze events (AQI > 300) in the 11 cities were 15% ± 13% and 7% ± 7% of all days during this period of 2014 and 2015, respectively. In provincial capitals (i.e., Shenyang, Changchun, and Harbin), 30% ± 8% and 16% ± 7% of all days during the same period experienced heavy and extreme haze events, respectively. Crop burning and coal burning for winter heating are two major seasonal emission sources of atmospheric pollutants during this period. The second period (Period II) covered the coldest time in winter, i.e., from January to February, with 9% ± 8% and 1% ± 1% of all days during the period experiencing heavy and extreme haze events, respectively, in the prefecture-level cities in 2014–2015. The corresponding figures in provincial capitals were 20% ± 5% and 2% ± 2% for heavy haze events and extreme haze events. Atmospheric pollutant emissions from coal burning were enhanced by the increase in winter heating. The third period (Period III) included most of the spring, i.e., generally from April to mid-May. Although heavy haze events did not occur in the prefecture-level cities on most spring days, extreme haze could happen in specific cities (e.g., Chifeng and Changchun) during periods with strong wind (Fig. 4). Wind-blown dust and agricultural dust (i.e., released during land preparation) in farmlands could be transported long distances during this period.

3.3 Cause analysis of typical haze periods

3.3.1 Crop residue burning period (*Period I*)

According to local conventional practices in Northeast China and actual weather conditions in 2015, we divided the whole crop harvesting period into pre-burning (Oct. 6–20), burning (Oct. 21–Nov. 10), and post-burning (Nov. 11–20) stages. The typical agricultural activities during the pre-burning stage were crop harvesting, grain handling and a small amount of straw burning. The burning stage involved two activities, i.e., most crop residue burning and a winter heat supply for various cities. The formal start dates of the heat supply to Harbin, Changchun, and Shenyang were October 15, October 25, and November 5, respectively. In the post-burning stage, nearly all crop residue burning events were completed, and the intensity of the heating supply was not high. Ground monitoring data clearly showed that the AQI across the Northeast Plain during the burning stage was much higher than in the post-burning and pre-burning

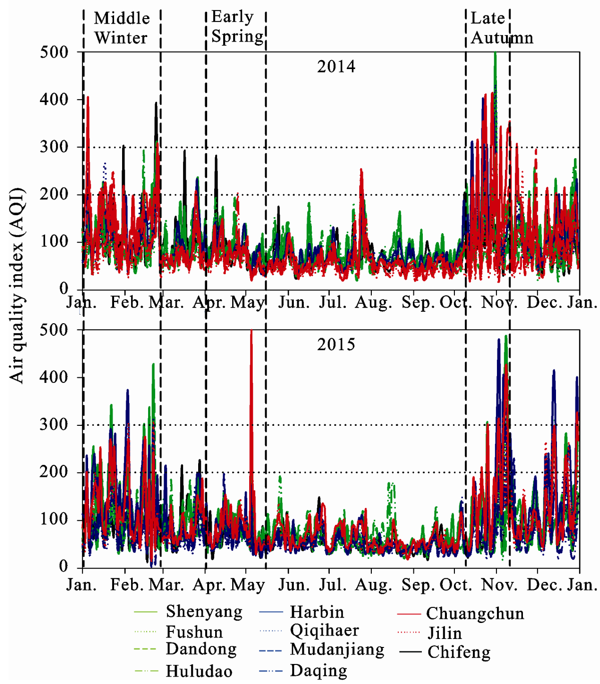

Fig. 4 Daily variations of air quality index (AQI) in 11 major cities of Northeast China in 2014 and 2015. The horizontal dotted lines represent a very unhealthy AQI level (i.e., 200) and hazardous AQI level (i.e., 300), respectively. The vertical dashed lines divided three important and typical haze periods, i.e., middle winter (January and February), early spring (April to mid-May), and late autumn (October 10 to November 10)

stages (Fig. 5a). The belt area of the SCH had a significantly higher AQI (> 160) than other regions, with the highest AQI (> 180) detected in the region centered on Changchun. Similarly, the AOD data also confirmed the spatial distribution of air quality during each stage (Fig. 5b), with significantly higher AOD values in the belt area of SCH and the Sanjiang Plain during the burning stage.

Regional air quality is closely linked to atmospheric pollutant emissions (including direct primary emissions and indirect secondary formation), meteorological conditions (e.g., atmospheric diffusion and boundary layer height), and inter-regional transportation (Marcazzan et al, 2001; Shen et al., 2014; Zhou et al., 2014; Chen et al., 2017). Local emissions, the internal cause of haze events, are considered to be due to general emissions (e.g., industrial, traffic, residential, and power plant sources), periodic emissions (e.g., agricultural activities and winter heating supply), and burst emissions (e.g., natural eruptions and exploration). In 2015, the number of cars, the value of industrial output, and the population

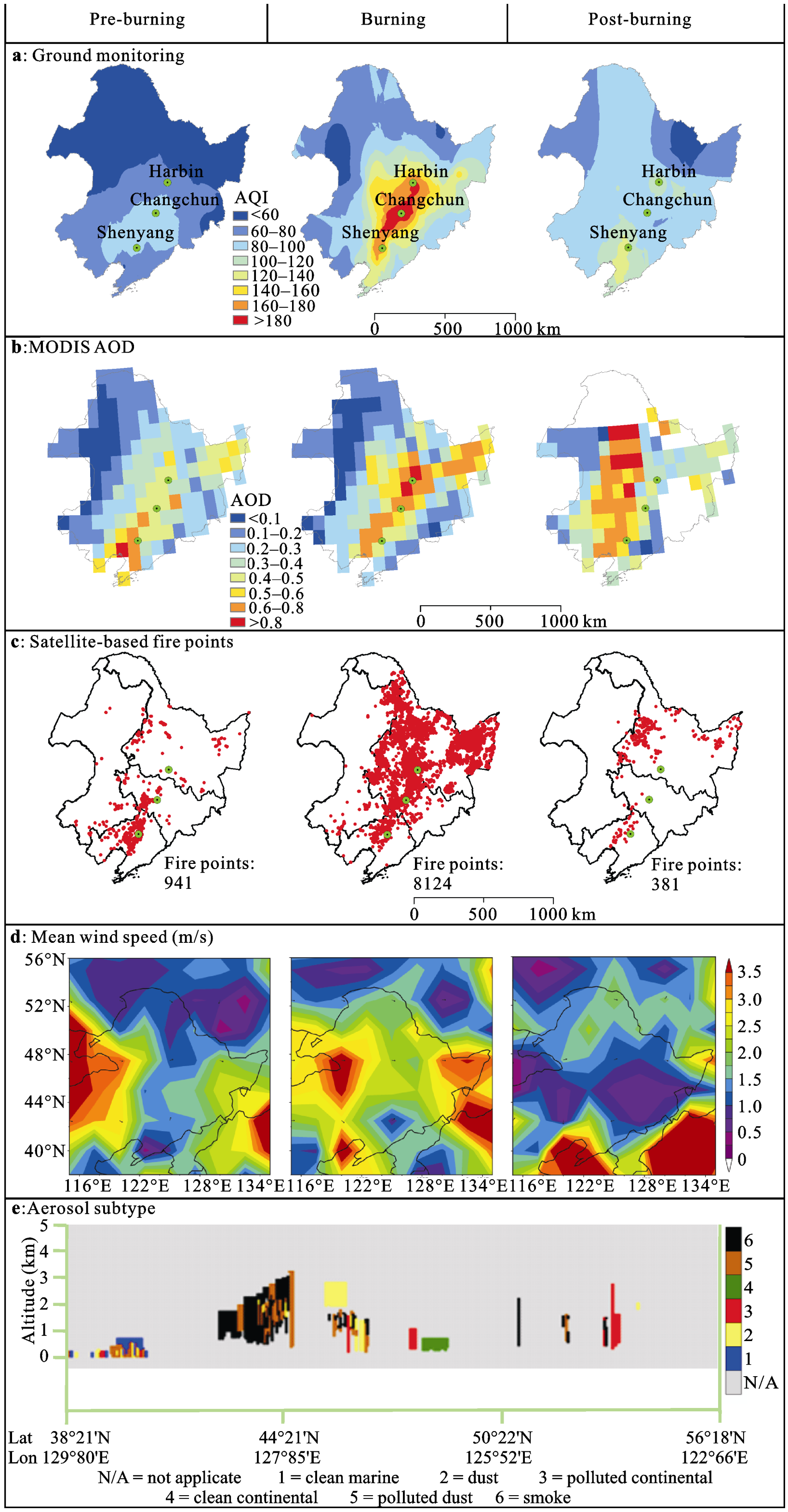

Fig. 5 Ground-monitored air quality index (AQI), satellite-based aerosol optical depth (AOD), satellite-based fire points, mean wind speed and vertical profile of aerosol subtypes retrieved by CALIPSO during pre-burning (Oct. 6–20), burning (Oct. 21–Nov. 10), and post-burning (Nov. 11–20) stages of crop harvesting period in late autumn and early winter in 2015. CALIPSO: Cloud-aerosol Lidar and Infrared Pathfinder Satellite Observation

in towns in the provinces of Liaoning, Jilin, and Heilongjiang increased on average by 198% ± 19%, 100% ± 37%, and 12% ± 6% compared to 2008. Such rapid increases suggest that the emission potential of traffic, industrial, and residential sources were substantially enhanced in the seven-year period. Therefore, background concentrations of atmospheric pollutants might be significantly increased and the atmospheric holding capacity could be close to saturation, which is the basis of heavy haze events throughout the year.

For Period I, several reasons may have jointly contributed to the significant increase in heavy or extreme haze events. First, crop residue burning could release a large amount of PM and gaseous pollutants (Zhao et al., 2015). The production of crop straw in Heilongjiang, Jilin, and Liaoning provinces increased by 58%, 33%, and 26%, respectively, from 2008 to 2015, indicating that crop residue burning would also have increased. We found that fire points during the burning period accounted for 86% of all agricultural activities in the fall and nearly covered all of the Sanjiang, Songnei, and Liaohe plains (Fig. 5c). The CALIPSO data also indicated that the aerosol type in the region was smoke when the data were gathered on November 4, 2015 (Fig. 5e). When residue burning began, the $PM_{2.5}$ and CO concentrations significantly increased, especially before rainfall days (Chen et al., 2017). The crop harvesting procedure and agricultural machinery could contribute to PM emissions by disturbing soil and plants and increasing fuel consumption (Li et al., 2016). The increase in suspended soil dust was most likely to form secondary road dust in cities or the countryside (Chen et al., 2017). Coal burning for heat is another important emission source of $PM_{2.5}$ and gaseous pollutants (e.g., SO_2 and CO) in winter. As an indicator of coal burning, SO_2 concentration rapidly increased by 2.4–2.8-fold when the formal heat supply commenced. Concentrated urban heating, self-determined coal burning in urban and suburban locations (e.g., bungalows and small factories), and rural indoor crop residue burning are the major types of winter heating in Northeast China. According to our preliminary estimation, combined with population density and the different types of heating, $PM_{2.5}$ emissions were mainly distributed in the city zone of SCH. Unrestrained fugitive dust emissions from coal storage piles and construction processes also increased in winter. Thus, although the intensity of coal burning was not as strong as in the coldest time of the year, the overall emissions due to crop residue burning, coal burning, and other emissions were largest in this period on an annual scale. The calm meteorological conditions that prevailed at this time prevented the diffusion of atmospheric pollutants during heavy and extreme haze periods. Although the regional average wind speed during the crop residue burning stage was slightly higher than during the other two stages (Fig. 5d), the daily wind speed was generally lower than 2 m/s on heavy haze days (e.g., October 25, November 3, and November 9, 2015) (Chen et al., 2017). Furthermore, there was also an inversion layer present on these days, as revealed by the vertical profile of air temperature.

3.3.2 Winter coal burning (Period II)

In Northeast China, the winter period can be divided into three sub-periods, i.e., early winter (November 11 to December 15), mid-winter (December 16 to February 28, and late winter (March 1 to March 31). At the regional scale, ground data confirmed the order of the average AQI to be early winter > mid-winter > late winter (Fig. 6a). However, snowfall in the northeastern region resulted in a large number of gaps in MODIS data from November to March, and therefore the average AOD in winter was likely lower than the actual value. In early winter, although most field activities have finished, there are still some crop residue burning and autumn soil preparation practices undertaken, and agricultural transportation, handling, and processing continues. Due to the close linkage between winter coal burning intensity and ambient air temperature, atmospheric pollutant emissions are generally highest during mid-winter, followed by early winter and late winter. There was a high frequency of heavy haze events observed throughout early winter and mid-winter, with $PM_{2.5}/PM_{10}$ ratios of 29%–91% (average: 70% ± 11%) (Fig. 6a and Fig. 6b), indicating fine particle emission sources. In the late winter period, lower coal burning emissions were accompanied by an increase in windblown dust events in some places, which was indicated by the lower $PM_{2.5}/PM_{10}$ ratios of 16%–81% (average: 49% ± 15%) (Fig. 6a and Fig. 6b). Meteorological data confirmed higher wind speeds in mid-winter and later winter compared to early winter (Fig. 6c). Thus, the higher AQIs in early winter were mainly caused by both large emissions of atmospheric pollutants and unfavorable diffusion conditions, with both lower emissions and favorable

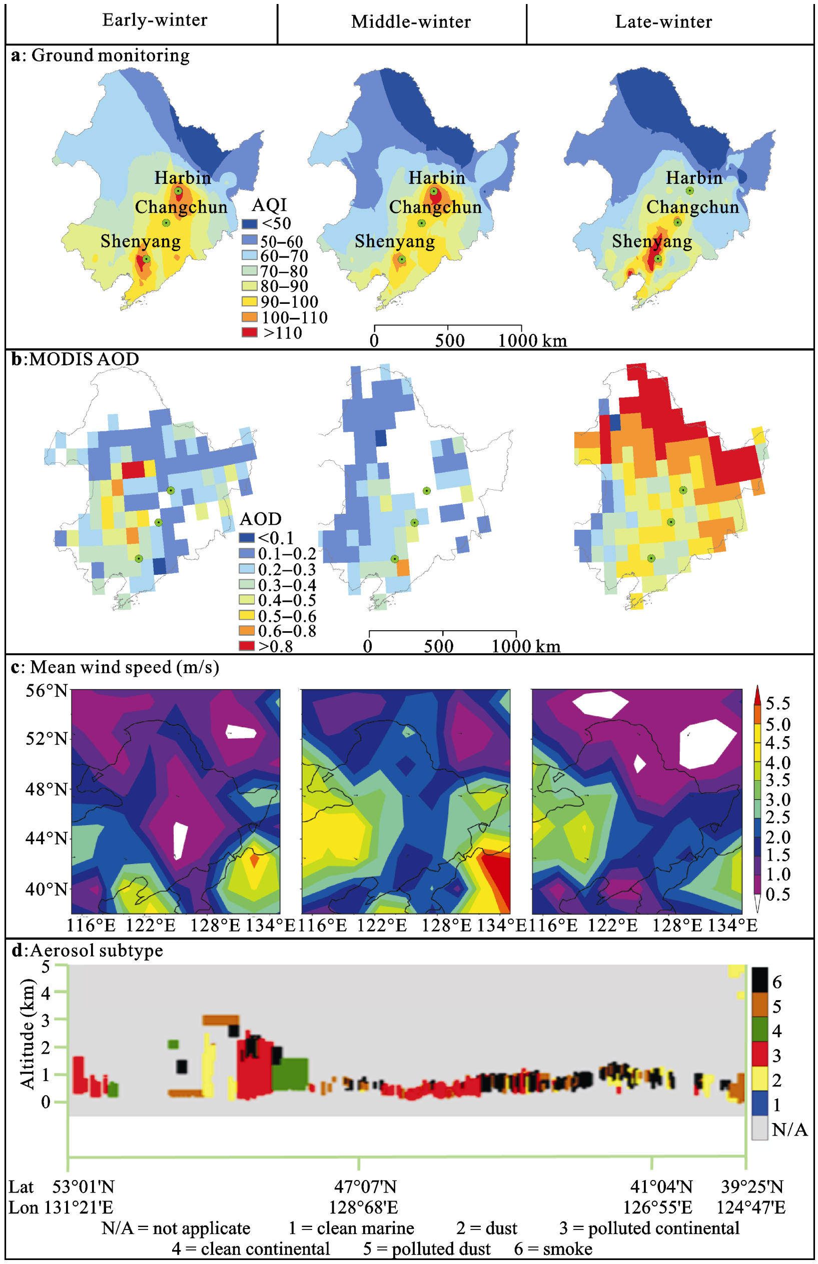

Fig. 6 Ground-monitored air quality index (AQI), satellite-based aerosol optical depth (AOD), mean wind speed and vertical profile of aerosol subtypes retrieved by CALIPSO during early winter (Nov. 11–Dec. 15), mid-winter (Dec. 16–Feb. 28), and late winter stage (Mar. 1–31) in 2015 and 2016. CALIPSO: Cloud-aerosol Lidar and Infrared Pathfinder Satellite Observation

diffusion conditions contributing to the better air quality in late winter.

Middle winter in Northeast China is mainly characterized by cold weather with snow, which means high levels of atmospheric pollutant emissions from coal burning and a higher relative humidity on snowy and foggy days. The CALIPSO data showed that the main aerosol subtypes in mid-winter were smoke and polluted continental dust below a height of 1.5 km (Fig. 6d), and ground monitoring data revealed the highest SO_2 and CO concentrations during this period. These indicators suggest that coal burning is the strongest seasonal emission source. With the acceleration of urbanization, the demand for winter heating has increased rapidly, which is reflected in the population size and in the towns and urban areas of Northeast China. For example, the population of towns and urban areas in Liaoning Province has increased by 33% and 17% from 2008 to 2015, respectively. Coal burning sources in cities can be divided into four subtypes, i.e., large power plants, industrial production, small heating boilers, and scattered coal burning. In mid-winter, regional air quality has a close relationship with emissions from large power plants and industrial production due to their high-intensity continuous operation, while small heating boilers and scattered coal burning can directly affect local air quality, especially in suburban areas where their use is centralized. In most of the countryside, agricultural straw is stored and burned for heating and cooking, and therefore contributes to smoke pollution. Automobile exhaust is another important emission source of atmospheric pollutants, although its contribution to heavy haze events is still controversial. In mid-winter, fuel consumption by vehicles is higher than in other periods because of the need to circulate heated air inside vehicles, the lower efficiency of gasoline diesel engines, the longer running time required for vehicle engines to warm-up, and the use of ice and snow tires. Additionally, automobile exhaust is released near the ground and is unlikely to be well diffused in the higher atmosphere considering the presence of unfavorable diffusion conditions near the ground and the street canyon effect. If wind speeds are low and the street canyon effect prevails, automobile exhaust emissions can cause local hotspots of atmospheric pollutants in urban areas. The atmospheric boundary layer is generally lowest in mid-winter, which will trap and concentrate pollutants, assuming the emission intensity remains constant. The higher relative humidity on foggy or snowy days is conducive to the hygroscopic growth of fine particles. Most heavy haze events in mid-winter occurred under conditions with temperature inversions and low wind speeds. Such unfavorable diffusion conditions hinder the diffusion of atmospheric pollutants in the vertical and horizontal directions, and thus atmospheric pollutants continuously accumulate, reaching high concentrations.

3.3.3 Spring dust (*Period III*)

The spring dust period in Northeast China generally starts after the snow thaws in April and ends when plant growth commences in May or June. A strong windblown dust event occurred on May 5, 2015 and was used to analyze the causes of haze (Fig. 7). The dust storm could be divided into a pre-dust-storm day (May 4), dust storm day (May 5), and post-dust-storm day (May 6). Ground data identified two severely affected areas, with an AQI higher than 300 (Fig. 7a). One was located in eastern Inner Mongolia and western Jilin Province, where there is widespread degraded grassland and saline-alkaline soil farmland, while the other was located in southern Heilongjiang Province and northern Jilin Province, where there is a vast area of farmland on the Songnei Plain. High AOD values extended over similar areas (Fig. 7b). In this case, the wind speed gradually increased during the pre-dust day and was highest (> 10 m/s) on the dust day (Fig. 7c), indicating the time of maximum dust occurrence. The CALIPSO data clearly showed that dust was the main aerosol subtype in this area on May 5, 2015 (Fig. 7d).

As the main driving factor of spring dust events, high wind speeds can increase local dust emissions from farmland, river beaches, roads, and construction sites, and is also responsible for long-distance regional dust transport from desert areas. The occurrence of high PM_{10} concentrations and a large coarse particle ratio (i.e., $(PM_{10}–PM_{2.5})/PM_{10}$) during these periods reflects the obvious dust emissions. In Northeast China, the natural dust sources are predominantly the degraded grassland or sand dunes in Inner Mongolia and saline-alkaline soil in western Jilin Province. Compared with black soils, the threshold friction velocity in deserts and areas with saline-alkaline soil is significantly lower, resulting in a high frequency of dusty weather. Soil disturbance by agricultural activities (e.g., tillage, land preparation) is an important anthropogenic source of dust. Previous studies have suggested that the total amount of dust emitted from agricultural activities was higher than the amount of natural windblown dust, although these activities only occurred over a few days or weeks. Moreover, dust from tilled bare soils can be readily suspended at low wind speeds. Thus, the combination of emissions from natural windblown dust and anthropogenic dust from agricultural practices in spring will increase the frequency and intensity of dusty weather in Northeast China.

3.4 Comprehensive control options to reduce extreme haze events

Strong emissions of atmospheric pollutants and unfavorable diffusion conditions are two factors for the occurrence of extreme haze (Wang and Christopher, 2003; Chen et al., 2015; Wang et al., 2015). General emissions from vehicles, road dust, and industry have led to increased

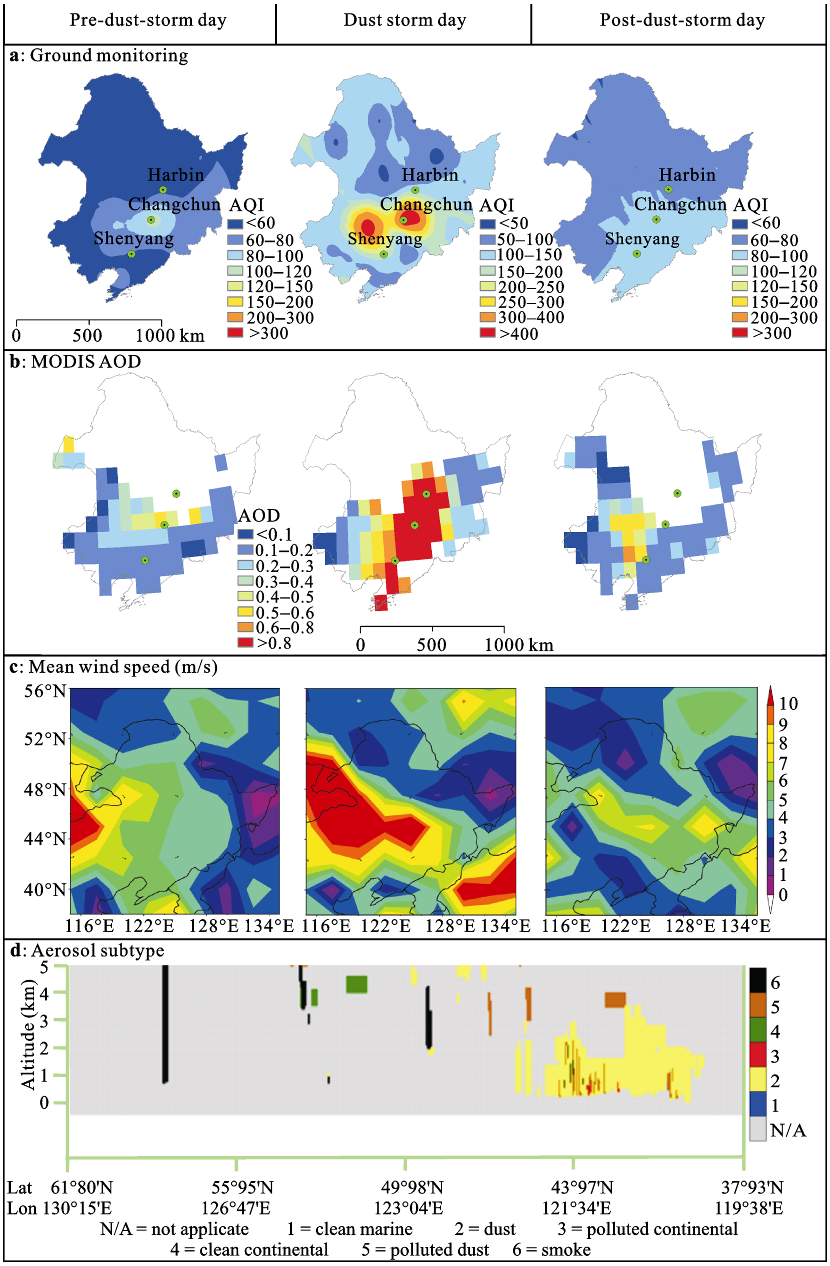

Fig. 7 Ground-monitored air quality index (AQI), satellite-based aerosol optical depth (AOD), mean wind speed and vertical profile of aerosol subtypes retrieved by CALIPSO during pre-dust storm (May 4), dust storm (May 5), and post-dust storm (May 6) periods in early spring in 2015. CALIPSO: Cloud-aerosol Lidar and Infrared Pathfinder Satellite Observation

background concentrations of atmospheric pollutants. At a regional scale, however, the primary emission sources in Northeast China are seasonal and are associated with crop residue burning in late-autumn, coal burning in winter, and spring dust in spring, all of which can lead to extreme haze events. Thus, a comprehensive set of pollution control policies, with restrictions on coal burning, crop residue burning, and dust emissions under certain meteorological conditions, are urgently needed to reduce the occurrence of extreme haze in Northeast China.

Direct combustion for thermal power, industrial production and heating, and residential consumption is the most common usage of coal in Northeast China. It occurs almost all year round, with coal burning for wintertime heating aggravating air pollution. In recent years, most families in urban areas have started to use gas stoves, but inefficient coal burning still occurs in thermal power plants, and industrial and civil heating boilers. Alongside the ongoing rapid economic development and industrial recovery, air pollution caused by coal burning has increased (Zhou et al., 2005). Thus, to improve the combustion efficiency and reduce the emission of pollutants from coal burning, which would solve the problem of the serious air pollution in Northeast China, the use of coal clean technology is inevitable. Clean coal technology, which is designed to reduce pollution and improve the efficiency of coal processing, combustion, conversion, and pollution control technologies, should be developed and used extensively, with coal processing technology also used to reduce the sulfur content prior to combustion (Zhu et al., 2016). Advanced combustion technology should be used to increase the combustion efficiency and desulfurization. In addition, gas purification technology can be used for desulfurization after combustion and will reduce pollutant emissions to control atmospheric pollution. Several comprehensive protection measures, such as local and sectional coal burning control planning and the readjustment of the energy structure, need to be implemented (He et al., 2002). Research and development needs to be undertaken to produce new sources of clean energy to replace coal. The development of clean and renewable energy, such as solar, biomass and wind energy, is crucial. With regard to winter heating, increasing the use of central heating systems and the extent of liquefaction and gasification in civilian fuel production would also reduce the emission of combustion pollutants (Zhao et al., 2017). To summarize, four aspects of technological improvement should be focused on: 1) further research and development of advanced methods of coal preparation to realize water conservation, clean combustion, economic processing, and efficiency; 2) increasing the amount of liquefaction and gasification in civilian fuel production and improving central heating systems; 3) promoting the efficiency, economics, and practicality of coal desulfurization and dust removal; and 4) boiler transformation to improve the thermal efficiency of combustion equipment.

For crop residue burning, complete prohibition is the most efficient way to reduce emissions in the long term. We estimated the production of crop straw from the provinces of Heilongjiang, Jilin, and Liaoning to be approximately 1.9×10^8 t, which accounted for 21% of national production in 2012. Planting in Northeast China follows a single cropping pattern, producing a large amount of crop straw at the national scale. Although agricultural open field burning was prohibited by the government in Northeast China, the effect of the ban effect is limited. Therefore, the key issue of complete prohibition is how to manage the large amounts of crop residue by substituting burning for alternative disposal methods or uses. Several solutions have been trialed in many regions, including electricity generation, animal feed supply, bio-ethanol production, returning straw to fields, and even prescribed burning. Regional straw comprehensive utilization technologies that produce fertilizers, fuels, and feeds using straw have been adopted in cities in Sichuan Province, with the rate of straw utilization reaching 94% (Shen et al., 2010). In the short term, it is necessary to time crop residue burning appropriately by considering air quality and meteorology model simulations at both regional and urban scales, and so avoid the simultaneous burning and accumulation of pollutant emissions. Regional transportation should also be taken into account by considering weather forecasts, so that the burning time in areas located windward of cities can be adjusted to reduce the potential for pollutants to be transported.

Spring dust in Northeast China mainly originates from long-range transportation from natural dust sources to the west and local anthropogenic dust generation. Natural windblown dust transport, caused by desertification of the grasslands in the Inner Mongolia Autonomous Region, is difficult to avoid in the short term and comprehensive measures are needed to prevent soil desertification and degradation. Road dust emissions and soil tilling-induced dust emissions are two of the most important anthropogenic sources. For local road dust, the frequent wetting of roads should be implemented and dust should be removed from roadways with rotary-brush vacuum wagons (Chalvatzaki et al., 2015). Additionally, the time that construction trucks operate should be limited and construction sites should be cleaned and covered (Yang et al., 2016). In the croplands around the city, conservation tillage technology is an effective and necessary method to decrease dust emissions, and has the advantages of increasing soil surface coverage, water conservation, soil/water erosion reduction, and maintaining soil productivity (Zhang et al., 2015). Rearranging the farming schedule and reducing farmland activity (e.g., soil plowing and planning) on strong wind days would reduce dust suspension into the atmosphere. Pollution control technology in cultivation machinery, such as soil dust removal bags or sprinklers, has also become available. In addition, other local dust sources (e.g., construction sites, barren land, and material storage yards) need to be cleaned up and pollution control management measures should be taken by local government (Zhu et al., 2004; Zhu et al., 2016).

4 Conclusions

In Northeast China, the 'Shenyang-Changchun-Harbin (SCH)' city-belt was found to be the most air polluted area on an annual scale. Three periods that typically experienced haze events were identified, i.e., from mid-October to mid-November (Period I), from late-December to February (Period II), and from April to mid-May (Period III). The first period experienced the most serious atmospheric pollution (AQI > 300) due to the strong $PM_{2.5}$ emissions from seasonal crop residue burning and coal burning for winter heating. The number of haze days in middle winter was larger than that in the other periods because of the increased $PM_{2.5}$ emissions from coal burning and vehicle fuel consumption, the lower atmospheric boundary layer, and stagnant atmospheric conditions. Spring dust-induced haze days were mainly affected by the regional transportation of windblown dust from degraded grassland in Inner Mongolia and bare soil in western Jilin Province, as well as dust releases from local agricultural tilling. Comprehensive control policies or restrictions on coal burning, fuel consumption, and crop residue burning in winter and autumn are urgently needed to resolve the heavy haze problem in Northeast China.

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