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Crop residual burning correlations with major air pollutants in mainland China

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Many studies have established the correlation between crop residual burning (CRB) and air pollutants such as particulate matter (PM) pollutants. However, few studies have compared CRB's correlations with all major air pollutants, including PM with aerodynamic diameters less than 10μm (PM_{10}) and 2.5 μm ($PM_{2.5}$), sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ozone (O_3), and carbon monoxide (CO), during open-field CRB seasons. This study monitored daily CRB spots from 2015 to 2016 in China, together with daily concentrations of $PM_{2.5}$, PM_{10}, CO, SO_2, NO_2, and 8 h average O_3 provided by the China National Environmental Monitoring Center. Temporal changes in air pollutant concentration at the provincial level and spatial contributions of CRB to air pollution at the monitoring-site level were analyzed. The results indicated that the decrease in CRB from 2015 to 2016 probably led to a decrease in the mean air pollutant concentration and maxima; it also probably led to more outliers at the provincial level in the CRB seasons, especially in Heilongjiang, Jilin, and Liaoning. Spatial contributions suggest that the Heilongjiang, Jilin, and Liaoning provinces experienced a regional increase in the concentrations of almost all major air pollutants during the CRB seasons. In Heilongjiang, Jilin, and Liaoning, 67.88%, 72.12%, 80.00%, 77.58%, and 69.09% of the monitoring sites recorded higher concentrations of $PM_{2.5}$, PM_{10}, NO_2, O_3 , and CO, respectively, in the winter CRB season of 2015 compared to other periods. In contrast, in other provinces with fewer and more widely distributed CRB spots, only the monitoring sites close to or on CRB spots experienced increases.

KEYWORDS
crop residue burning, MODIS, major air pollutant, China, spatiotemporal changes

1 Introduction

Owing to rapid economic development over the past three decades (Chen and Xie, 2014), China has experienced severe and persistent air pollution (Li et al., 2020). Air pollution is associated with severe health risks and has attracted considerable attention from the public and government (Shen et al., 2020). Both gaseous pollutant and particulate matter (PM) levels in the urban areas of China have significantly increased in the recent past (Ma et al., 2019; Wu et al., 2019). PM, including $PM_{2.5}$ and PM_{10} (aerodynamic diameter of the particle equal to or less than 2.5 and 10 μm, respectively), and super-dispersed particles and gaseous pollutants, such as sulfur dioxide (SO_2), nitrogen dioxide (NO_2), carbon monoxide (CO), and ozone (O_3), increase the risk of cardiovascular and respiratory diseases and, subsequently, mortality (Peng et al., 2009; Xu et al., 2016; Hwang et al., 2017; Filonchyk et al., 2018; Shen et al., 2020). In recent years, the Chinese central and local governments have introduced many laws and regulations to decrease primary emissions and improve urban air quality (Chen et al., 2013; Yin et al., 2017a; Tilt, 2019). Among them, the Five-year Air Pollution Prevention and Control Action Plan (known as the "Ten air pollution prevention and control measures") released in 2013 by the China State Council is the most popular setting target, specifically for the reduction of air pollution before the end of 2017 in most polluted cities across China (Wang et al., 2014).

Owing to these efforts, air quality in many urban centers in China has begun to show marked and sustained improvement (Silver et al., 2018; Li et al., 2019; Tilt, 2019; Zhai et al., 2019). A large body of literature has confirmed that annual average concentrations of $PM_{2.5}$ and some gaseous pollutants have significantly decreased, especially since 2013 (Lv et al., 2016; Geng et al., 2019; Ma et al., 2019; Shen et al., 2020). He et al. (2017) found that the annual average concentrations decreased by 5.3%, 4.9%, 11.4%, 12.0%, and 21.5% for CO, NO_2, PM_{10}, $PM_{2.5}$, and SO_2, respectively, but increased by 7.4% for O_3 in 2015 in major Chinese cities. Li et al. (2017) analyzed the spatial and temporal variations of $PM_{2.5}$, PM_{10}, SO_2, CO, NO_2, and O_3 in 187 Chinese cities from January 2014 to November 2016 and found that $PM_{2.5}$, PM_{10}, SO_2, CO, and NO_2 concentrations had decreased. Furthermore, an unprecedented decrease in the concentrations of five major pollutants, except O_3 in Beijing (Maji et al., 2020), has also been detected by researchers.

As a significant air pollutant, crop residue burning (CRB) in open fields is among the most important contributors to short-term and severe air pollution in China (Li et al., 2010; Tao et al., 2013; Ni et al., 2017); China accounts for 17.9% of the global crop straw production (Yin et al., 2017b). The country has suffered from intensive open CRB on a large scale for a long time (Chen et al., 2017). According to estimates, China produced 630 million tons of crop residue from 1995 to 2005; on average, one-third of it remained unused, most of which was burned in fields (Liu et al., 2008; Wang et al., 2013; Qiu et al., 2014; Hou et al., 2019). Studies have reported that despite the Chinese government's proposed regulations and laws to prohibit CRB, the total amount of CRB in China has continuously increased from 2001 to 2018 (Cao et al., 2006; Yan et al., 2006; Huang et al., 2012a; Zhuang et al., 2018b; Yin et al., 2021). Thus, it is essential to determine the impact of CRB on air pollution.

Many studies have demonstrated that periods of significant CRB or biomass burning lead to an immediate increase in $PM_{2.5}$ and PM_{10} concentrations (Li et al., 2010; Tao et al., 2013; Zhang et al., 2016; Yin et al., 2017b; Ni et al., 2017). For example, Li et al. (2010) found that agricultural biomass burning in northern China significantly contributed to the haze between June 12 and 20 in Beijing. Yin et al. (2017b) analyzed the correlations between CRB spots and concentrations of $PM_{2.5}$, and found that in summer, China's middle-east region experienced r values greater than 0.5. In autumn–winter, crop residue burning can induce an evident $PM_{2.5}$ increase in China's northeast region. Yin et al. (2017b) identified the impact of CRB on $PM_{2.5}$ concentrations in China. Similarly, Saxena et al. (2021) attempted to determine the impact of CRB on PM_{10}, $PM_{2.5}$, NO_2, and SO_2 air quality in New Delhi.

These studies have confirmed the high correlation between CRB and certain air pollutants and suggested that during periods of CRB, concentrations of a few air pollutants increase. However, few studies have fully examined changes in all major air pollutants, including PM_{10}, $PM_{2.5}$, SO_2, NO_2, O_3, and CO, during periods of substantial CRB, even after all these air pollutants have been proven to adversely affect human health and climate change (Tao et al., 2013).

However, a few scholars have used emission factors to calculate the emission inventory for CRB. In these studies, all major air pollutants were calculated and estimated (Yang et al., 2008; Granier et al., 2011). For example, Yang et al. (2008) used emission factors proposed by a few studies to estimate the amount of crop residues in Suqian, China, and the total amount of emissions from CRB during summer and autumn harvests. Specifically, the emissions included total suspended particulates (TSP), PM_{10}, SO_2, nitrous oxides (NO_x), ammonia gas (NH_3), methane (CH_4), ethyl cellulose (EC), oxidation catalyst (OC), volatile organic compounds (VOC), CO, and carbon dioxide (CO_2) in Suqian, China. Similarly, Sahu et al. (2021) estimated seasonal emissions of $PM_{2.5}$, PM_{10}, biochemical (BC), OC, CO, NO_x, SO_2, VOC, CH_4, and CO_2 from CRB in India.

These studies, based on emission factors, can provide general information regarding the spatial distribution of pollutant loads or emissions produced by CRB on a large scale. However, because pollutants were calculated using a coefficient method at the annual level, it is difficult to track specific and accurate variabilities of air pollutants during periods of intensive CRB (Yu et al., 2019). Within this context, it is necessary to determine the correlation or impact of CRB on the changes in all important air pollutants. Although many studies have focused on the impact of CRB on PM air pollutants or the emission inventories of CRB for many pollutants, few studies have used national site monitoring to clarify the influence of seasonal CRB in China on all major air pollutants. The impact of seasonal CRB in China on the spatial and temporal changes in all major air pollutants is still unclear. To fill the gaps in the literature, the present study aimed to estimate how all major air pollutant concentrations change during CRB seasons and their differences in different regions with varying amounts of CRB.

As there was an obvious change in CRB spots in China from 2015 to 2016 (Yu et al., 2019), the present study used the 2015 and 2016 data and extracted the daily spatial

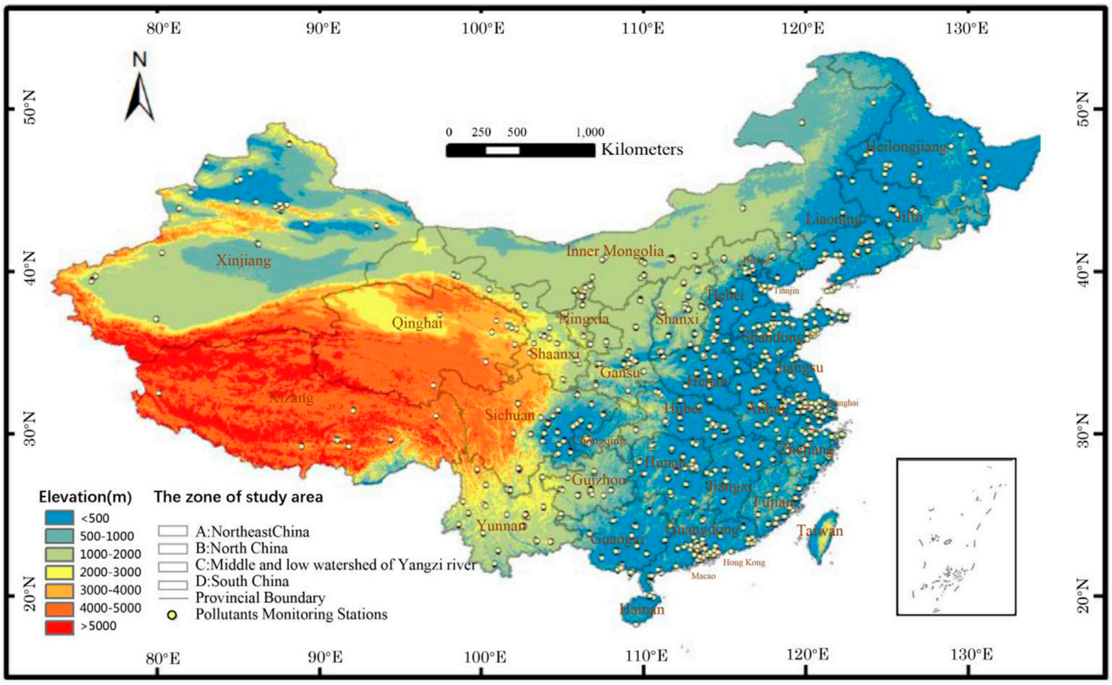

FIGURE 1
Spatial distribution of air quality stations in China and the study area and elevation.

distribution of CRB in mainland China from the Moderate Resolution Imaging Spectroradiometer (MODIS) images to obtain the hourly concentrations of six major pollutants, $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO , from national air quality monitoring stations. For temporal changes, the monthly changes in air pollutant concentrations and the number of CRB spots were compared between 2015 and 2016 at the provincial level. The differences in air pollutant concentrations during the same period with intensive CRB in both 2015 and 2016 were compared using box-plot analysis. For spatial changes, differences in air pollutant concentrations at the monitoring-site level were quantified from the non-CRB to CRB seasons.

2 Materials and methods

2.1 Data

2.1.1 MODIS fire product

We selected the MODIS Thermal Anomalies/Fire Daily L3 Global Product (MOD14A1) from 2015 to 2016 to detect fires (accessed at https://modis.gsfc.nasa.gov/data/dataprod/mod14.php). We downloaded 18 images covering all of China every day from January 2015 to December 2016 and then extracted the daily fire burning spots (Huang et al., 2012a; Yin et al., 2017b). The values were 7, 8, or 9 if there were fire spots, according to the product. Thereafter, the daily fire spots provided by MOD14A1 were overlaid with the spatial scale of agricultural land provided by the land-use datasets to identify the fire spots as CRB spots. It is assumed that the fire points located on agricultural lands are CRB spots.

2.1.2 Air quality monitoring data

Air quality monitoring data were obtained from the China National Environmental Monitoring Center (CNEMC) (accessed at http://www.cnemc.cn/), which has published hourly air quality data for the six criteria pollutants at individual monitoring sites since January 2013 (Wang et al., 2014). Data from 1 January 2015 to 31 December 2016 were used in the present study. Daily concentrations of six criteria air pollutants, including $PM_{2.5}$ ($\mu g/m^3$), PM_{10}($\mu g/m^3$), SO_2 ($\mu g/m^3$), NO_2 ($\mu g/m^3$), and CO(mg/m^3), were calculated based on hourly concentrations for sites, and the daily maximum 8 h averageO_3 ($\mu g/m^3$) concentrations were calculated based on the average hourly ozone concentrations for sites (Shen et al., 2020). The average provincial concentrations of air pollutants were calculated by averaging the concentrations at all sites in each province.

The monitoring sites, located in eastern and central China, had high economic development and a dense population, which included a mix of urban and background sites (Wang et al., 2013). The spatial distribution of monitoring sites along with the elevation and provincial boundaries is presented in Figure 1.

2.1.3 Land-use data and precipitation data

Land-use data were used to detect the spatial distribution of CRB. We downloaded the 2015 land-use dataset from the Data Center for Resources and Environmental Sciences, Chinese Academy of Sciences (accessed at http://www.resdc.cn). The land-use dataset was produced based on Landsat 8 images, including 25 primary land-use categories (http://www.resdc.cn/data.aspx?DATAID=184) with a resolution of 1 km × 1 km. In the present study, classes 11 and 12, which represent agricultural land, were used to overlay the fire spots and extract the CRB spots.

The daily precipitation data at each meteorological station from January 2015 to December 2016 were sourced from the China Meteorological Administration (http://www.cma.gov.cn/). Daily precipitation at meteorological stations was interpolated to present daily precipitation to a 1 km spatial resolution using an inverse distance weighting technique in ArcGIS software. The spatial precipitation resolution was the same as that of the MODIS fire production used in the present study. On the day when an air quality monitoring site was located in a place with precipitation greater than zero, the air quality monitoring site should be eliminated when analyzing the influence of CRB on air quality changes.

2.2 Methods

2.2.1 Temporal impact of CRB on air pollutants

There are several sources of air pollution. To clarify the temporal impact of CRB on air pollutants, we assumed that the other sources were generally unchanged in two adjacent months. Therefore, the months with abnormally high pollutant concentrations (Mc_m) compared with adjacent months at the provincial level were identified using the following equation:

$$Mc_{(i,j,m)} = \begin{cases} 1, & pc_{(i,j,m)} > max\left(pc_{(i,j-1,m)}, pc_{(i,j+1,m)}\right) \\ 0, & \text{other conditions} \end{cases}, \quad (1)$$

where $Mc_{(i,j,m)}$ is a binary variable with a value of zero or 1. A value of 1 indicates that the concentration of the mth pollutant experienced an abnormally high value in the jth month for the ith province. $pc_{(i,j,m)}$ is the monthly average concentration of the mth pollutant in the jth month for the ith province, and $max(pc_{(i,j-1,m)}, pc_{(i,j+1,m)})$ denotes the higher concentration of the mth pollutant in the ith province in both the (j-1)-th and (j+1)-th months.

Thereafter, Mc_m was analyzed along with the daily change in CRB spots, with the aim of determining whether months with intensive CRB would suffer severe concentrations of air pollution. Variations in air pollutants in periods with intensive CRB spots were compared using a box-plot analysis, where the maximum, minimum, median, mean, and outliers of pollutant concentrations in the periods were presented. Practically, the periods with intensive CRB spots for each province were selected in 2015 and 2016, and the daily mean concentrations ($PM_{2.5}$,PM_{10}, SO_2, CO, and NO_2) and 8 h concentrations (O_3) at monitoring sites in the periods with no precipitation were the statistics used for each province.

2.2.2 Spatial impact of CRB on air pollutants

The temporal analysis presented the overall influence of CRB on air pollution at the provincial level. To present the spatial impact of CRB on air quality, the rate of air pollutant change in periods with intensive CRB spots, denoted as RPR, was calculated for each monitoring site (see Eq. 2). Before calculating the RPR, the period with intensive CRB spots (Pd) is defined. CRB is usually concentrated in one or two seasons, which is consistent with the sowing or harvest season (Huang et al., 2012b; Chen et al., 2017). Therefore, it is necessary to focus on periods with intensive CRB to quantify the influence of CRB on air pollutants (Chen and Xie, 2014; Yu et al., 2019). In the present study, we selected Pds at the provincial level because the agricultural form was similar in one province. We assumed that the Pds in 2015 and 2016 were the same, which facilitated the following comparative analysis. There is no consensus on Pd selection because the studies have been performed at different spatial and temporal scales. To identify the Pd in the present study, we ruled that one province should have one or two Pds, which was determined by its CRB seasonal features. The Pd for a specific province should be a continuous period and contain the maximum daily CRB spots in the CRB season. Meanwhile, during Pd, the number of CRB spots should show an increasing trend in the beginning, then reach a peak, and finally, experience a decline. This can be considered one Pd for a specific province.

After identifying Pd, the RPR was calculated as the ratio of the average air pollutant concentrations in Pd at a site to the average air pollutant concentration in the periods before and after Pd at the site:

$$\mathbf{RPR}_m = \frac{MeanCon_{m-RP}}{MeanCon_{m-nonRP}}, \quad (2)$$

where $MeanCon_{m-RP}$ is the average daily concentration of the mth air pollutant at a specific monitoring site during the selected Pd and $MeanCon_{m-nonRP}$ is the average daily concentration at the monitoring site during the period 20 days before Pd and 20 days after Pd.

To avoid the influence of precipitation on air quality, monitoring sites located in places with precipitation greater than zero were eliminated when calculating $MeanCon_{m-RP}$ and $MeanCon_{m-nonRP}$. According to Eq. 2, an RPR_m larger than 1 indicates that the concentrations of the mth air

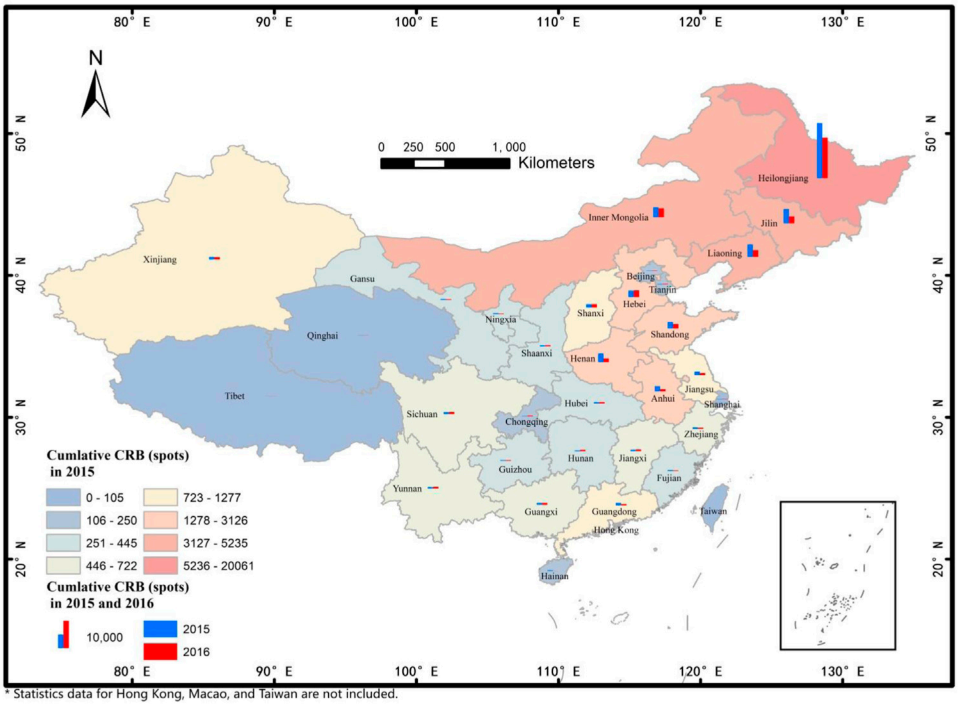

FIGURE 2
Spatial distribution of cumulative CRB spots in China (2015 and 2016) at the provincial level.

pollutant were higher in the CRB seasons than in other periods, suggesting a higher impact of CRB on the *m*th air pollutant at the site.

The spatial distribution of the CRB was also extracted for Pd. We applied the methodology described by Yu et al. (2019), who calculated the intensity of the CRB spots (ICP) at each spatial grid during the Pd. The spatial patterns of RPR and ICP, which reflect the changes in air pollutants during the CRB seasons and the density of CRB, respectively, were compared and analyzed to determine the spatial impact of CRB on air pollution.

3 Results

3.1 CRB counts and concentration of air pollutants at the provincial level

Quantitative statistics of cumulative CRB spots in 2015 and 2016 for provinces and their ratio to the total national number of CRB spots are presented in Figure 2. In 2015, there were 54,071 CRB spots in China, which decreased to 39,140 in 2016. Approximately 55% and 50% of the retrieved CRB spots were located in Heilongjiang, Liaoning, and Jilin in 2015 and 2016, respectively. In contrast, only approximately 1.40% of the retrieved CRB spots were located in the northwestern provinces of Gansu, Ningxia, Qinghai, and Tibet in 2015 and 2016, respectively. In summary, a significant decrease in CRB occurred between 2015 and 2016, and there was a considerable difference in CRB at the provincial level. The differences in quantitative CRB at the provincial level are probably due to the varying local climates and topological limitations on agricultural production in provinces. Therefore, it is sensible to analyze the amount of CRB and its influence on air pollution at the provincial level.

To simplify the analysis process, we selected provinces with high CRB to conduct the following analysis. Specifically, eight provinces with the largest number of CRB spots were selected: Heilongjiang, Jilin, Liaoning, Inner Mongolia, Henan, Shandong, Hebei, and Anhui. The total number of CRB spots in these eight provinces was 43,300 and 29,223, contributing approximately 80% and 75% of the total national CRB, in 2015 and 2016, respectively.

3.2 Temporal changes in air pollutants under CRB

3.2.1 Changes in monthly air pollutants

The monthly average concentrations of the six major air pollutants and the daily number of CRB spots at the provincial

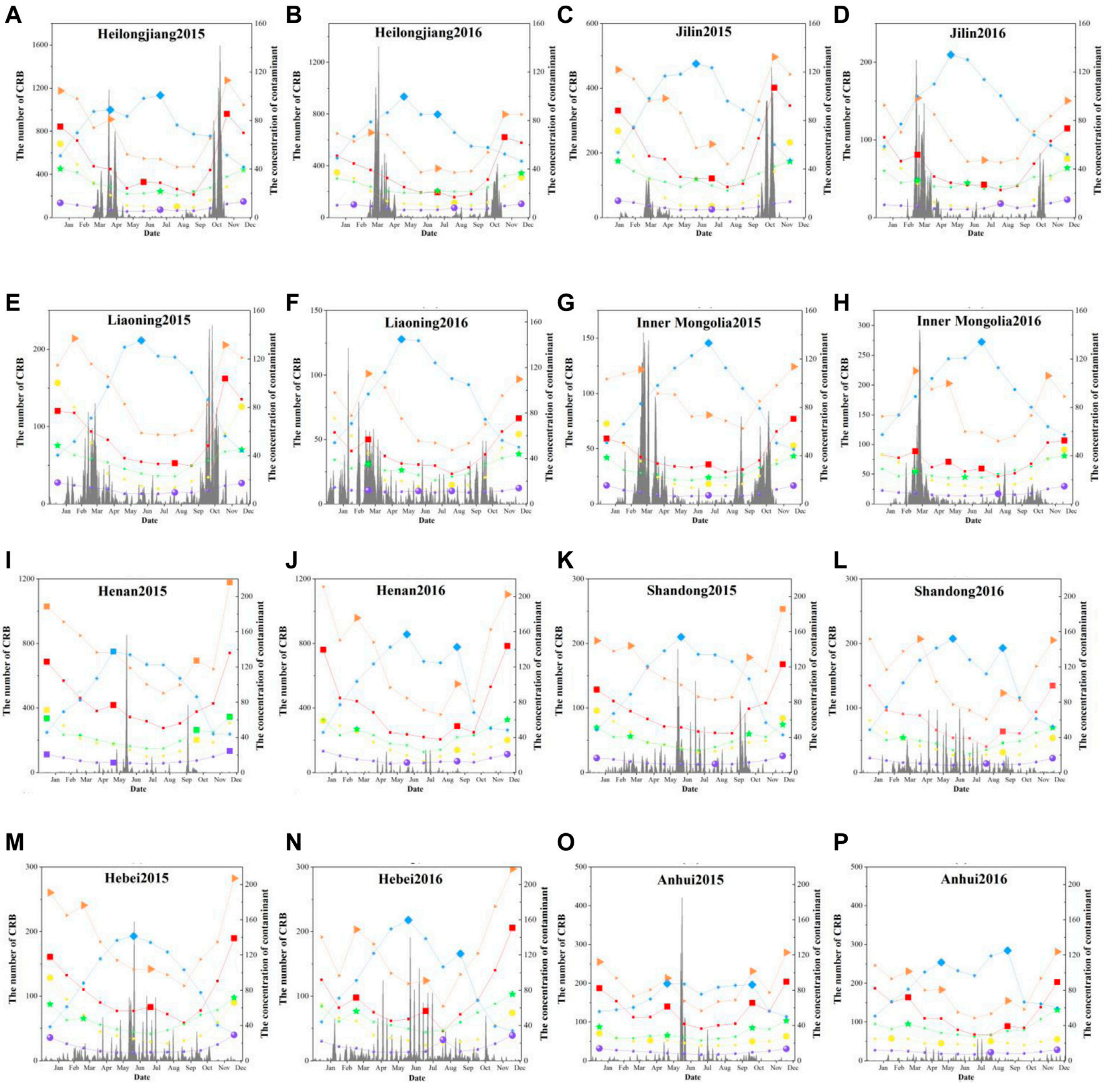

FIGURE 3
Temporal change in monthly average concentrations of six major air pollutants and the daily number of CRB spots from 2015 to 2016, **(A,B)** in Heilongjiang, **(C,D)** in Jilin, **(E,F)** in Liaoning, **(G,H)** in Inner Mongolia, **(I,J)** in Henan, **(K,L)** in Shandong, **(M,N)** in Hebei, and **(O,P)** in Anhui.

level in 2015 and 2016 were analyzed; Figure 3 presents the changes in air pollutant concentrations. The unit of each air pollutant in all figures and tables is consistent with the original data provided by the CNEMC. To make it readable in Figure 3, we multiplied the coefficient of 10 by the CO concentrations in Figure 3.

In general, the concentrations of most pollutants were higher in winter and lower in summer, including $PM_{2.5}$, PM_{10}, SO_2, NO_2, and CO (Figure 3). The lower concentrations in winter are probably associated with unfavorable meteorological conditions (low boundary layer height, temperature, wind speed, etc.) and human activities (coal combustion, biomass burning, residential heating, etc.) (Sun et al., 2019; Shen et al., 2020). The concentrations of O_3 exhibited an opposite trend, being lower in winter, which is attributed to lower vertical mixing due to lower planetary boundary heights, slowest chemical loss due to low temperature, and solar radiation in winter (Wen et al., 2018). These trends are consistent with the conclusions of previous

TABLE 1 Selected Pds and daily mean CRB spots in 2015 and 2016.

Province	Name	Period	Length (days)	Daily mean CRB in 2015 (spots)	Daily mean CRB in 2016 (spots)
Heilongjiang	HLJ-Pd1	Apr 1st–May 1st	31	230.74	121.35
	HLJ-Pd2	Oct 14th–November 10th	28	363.54	130.18
Jilin	JL-Pd1	Mar 17th–March 31st	15	50	58.6
	JL-Pd2	Oct 25th–November 4th	11	184.64	38.45
Liaoning	LN-Pd1	Mar 9th–March 27th	19	49.84	28.63
	LN-Pd2	Oct 12t–November 4th	24	79.5	10.67
Inner Mongolia	IM-Pd	Mar 5th–March 31s	27	50.26	68.78
Hebei	HB-Pd	Jun 8th–June 22nd	15	47.2	16.8
Shandong	SD-Pd	Jun 8th–July 11th	34	21.47	10.94
Henan	HN-Pd	Jun 5th–June 22nd	18	89.11	5.78
Anhui	AH-Pd	Jun 9th–June 22nd	14	76.07	3.07

studies (Wang et al., 2014; Shen et al., 2020). The concentrations of pollutants experienced distinct and unique seasonal features. Under their individual features, abnormal changes were detected according to McDonald's analysis.

In Heilongjiang, Jilin, and Liaoning, CRB was concentrated in spring (March and April) and autumn (October) (Figure 3), which is consistent with the results of previous studies (Zha et al., 2013; Wang et al., 2015). In months with concentrated CRB, the provinces experienced abnormally high levels of air pollution due to PM pollutants. For example, in 2015 in Heilongjiang (Figure 3A), the Mc of O_3 and PM_{10} was detected during the spring CRB season, and the Mc of $PM_{2.5}$ and PM_{10} was detected in the autumn CRB season. Similarly, the Mc of $PM_{2.5}$ and PM_{10} was detected in the autumn CRB season in 2015 in Jilin, the spring CRB season in 2016 in Jilin, the autumn CRB season in 2015 in Liaoning, and the spring CRB season in 2016 in Inner Mongolia. In provinces with many CRB spots, abnormally high concentrations of one or two major air pollutants (usually PM pollutants) were found during the CRB seasons.

Henan, Shandong, Hebei, and Anhui had limited CRB, which was usually concentrated during summer. This is probably because a small amount of CRB cannot create a regional or provincial impact on air pollution, and weather conditions in summer do not promote air pollution for most pollutants except O_3 (Bao et al., 2015; Yin et al., 2017a). Therefore, only the Mc of O_3 was detected in Shandong and Hebei.

The Mc analysis showed that in northeast China, a significant amount of CRB occurred (contributing to more than 50% of the total CRB in China, based on the data listed in Figure 3) during spring and summer, and it was vulnerable to abnormally high concentrations of one or two major air pollutants. However, for other provinces in China, CRB usually occurs during summer at a lower frequency. Although this does not usually lead to a provincial increase in major air pollutants, attention should be paid to O_3 control during the CRB seasons in these provinces.

3.2.2 Changes in air pollutants in 2015 and 2016

In addition to the monthly analysis, the changes in air pollutants in 2015 and 2016 were compared because there was a considerable drop from 54,071 CRB spots in 2015 to 39,140 in 2016 on the national scale. Table 1 presents the selected Pds in the provinces in 2015 and 2016. The box-plot analysis showed the ranges, means, medians, and outliers of all air pollutant concentrations in the selected Pds in 2015 and 2016.

Heilongjiang, Jilin, and Liaoning had the highest number of CRB spots, which decreased from 29,831 CRB spots in 2015 to 19,743 in 2016. HLJ-Pd1, HLJ-Pd2, JL-Pd2, LN-Pd1, and LN-Pd2 experienced a decrease in the mean concentrations of almost all the major air pollutants from 2015 to 2016 in the same Pds. For example, in HLJ-Pd1, the mean concentrations of $PM_{2.5}$ decreased from 39.22 to 31.37$\mu g/m^3$ from 2015 to 2016, which also decreased from 77.93 to 73.52 $\mu g/m^3$ in HLJ-Pd2 from 2015 to 2016 (see Figure 4A). The mean concentrations of other major air pollutants also decreased to different extents from 2015 to 2016 (see Figures 4B–F).

In addition to the decrease in the mean concentrations of air pollutants from 2015 to 2016, our findings also showed that the maximum air pollutant concentrations in the Pds decreased from 2015 to 2016. For example, in JL-Pd2, the mean concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO decreased from 107.61 to

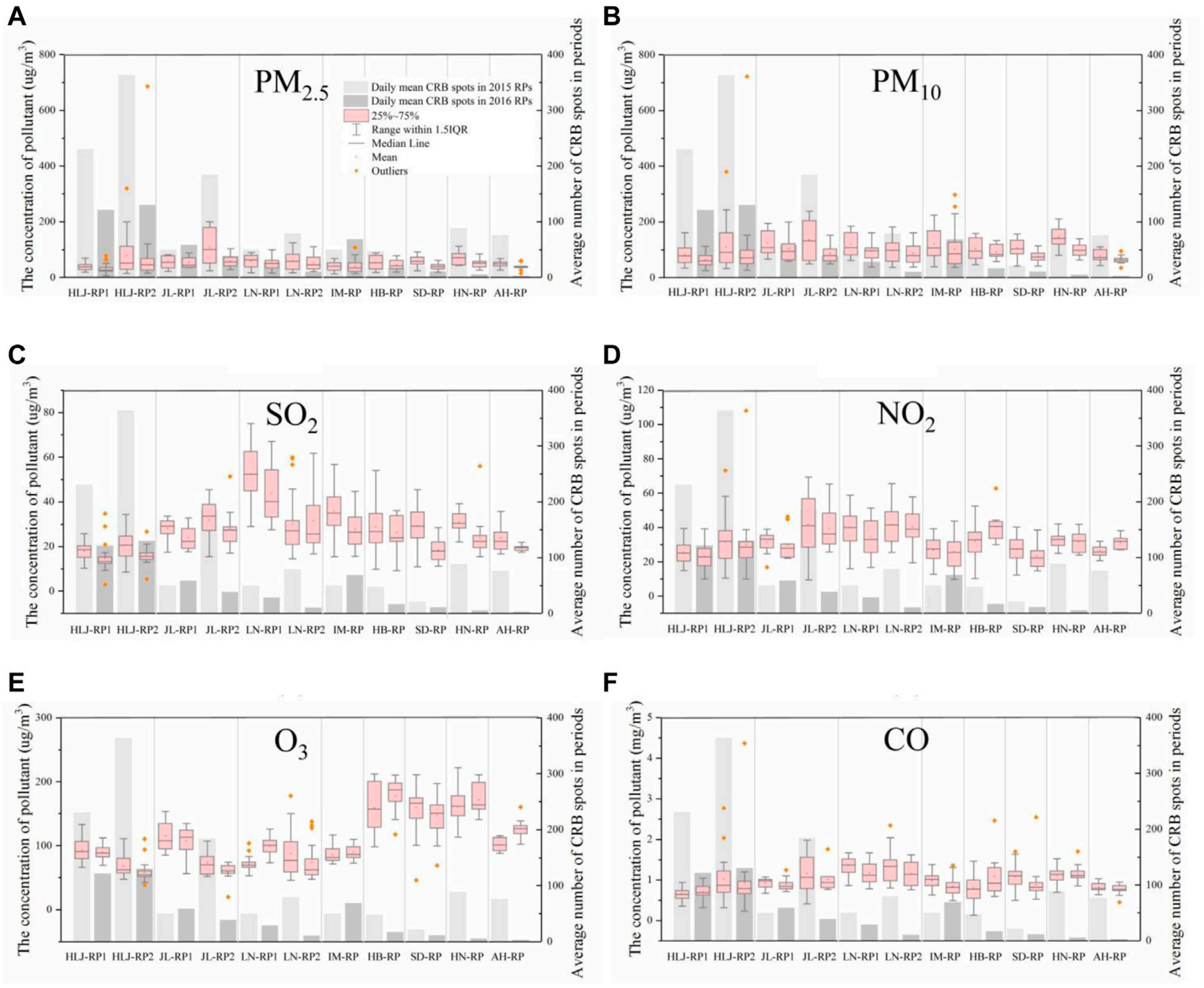

FIGURE 4
Boxplots of six air pollutants in Pds at the provincial level in 2015 and 2016 for **(A)** $PM_{2.5}$, **(B)** PM_{10}, **(C)** SO_2, **(D)** NO_2, **(E)** O_3, and **(F)** CO.

57.72 $\mu g/m^3$, 135.61 to 86.62 $\mu g/m^3$, 31.87 to 28.06 $\mu g/m^3$, 41.78 to 39.32$\mu g/m^3$, 73.08 to 58.31 $\mu g/m^3$, and 1.16 to 1.01 mg/m^3, respectively. The maximum concentrations for $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO decreased to 251.59$\mu g/m^3$, 251.00 $\mu g/m^3$, 17.55 $\mu g/m^3$, 23.93 $\mu g/m^3$, 37.28mg/m^3, and 1.21mg/m^3, respectively.

The results for northeast China suggest that a substantial decrease in CRB in a short time would lead to an obvious decrease in the mean concentrations and a decrease in the maximum concentrations of air pollutants. Even if CRB was lower in other provinces, changes in the concentrations of air pollutants and CRB spots were also detected. For IM-Pd, the daily mean number of CRB spots increased from 50.26 spots in 2015 to 68.78 in 2016. Changes in the mean concentrations of Pd from 2015 to 2016 were not significant. However, the maximum concentrations of $PM_{2.5}$, PM_{10}, and NO_2 were higher in 2016 than in 2015. The maximum concentration of $PM_{2.5}$ was 94.52 $\mu g/m^3$ in 2016 and decreased to 91.45 $\mu g/m^3$ in 2015. Outliers of $PM_{2.5}$ concentrations also occurred in IM-Pd in 2016, even though the mean concentrations of $PM_{2.5}$ in IM-Pd in 2015 and 2016 did not change significantly (40.96 $\mu g/m^3$ in 2015 and 39.05 $\mu g/m^3$ in 2016). Similar results were observed for PM_{10} and NO_2. Therefore, when CRB was similar in Pds, the effects on the mean concentrations may be negligible, whereas larger CRB spots promote maximum concentrations and lead to outliers.

For Hebei, Shandong, Henan, and Anhui, the daily mean CRB spots were lower compared to Heilongjiang, Jilin, Liaoning, and Inner Mongolia; however, a decreasing trend in CRB was also observed in the latter four provinces from 2015 to 2016. Along with the decrease in CRB in 2015, the mean concentrations of $PM_{2.5}$, PM_{10}, SO_2, NO_2, and CO generally decreased. Specifically, in Henan, the daily mean CRB was 89.11 spots in HN-Pd 2015, which decreased to

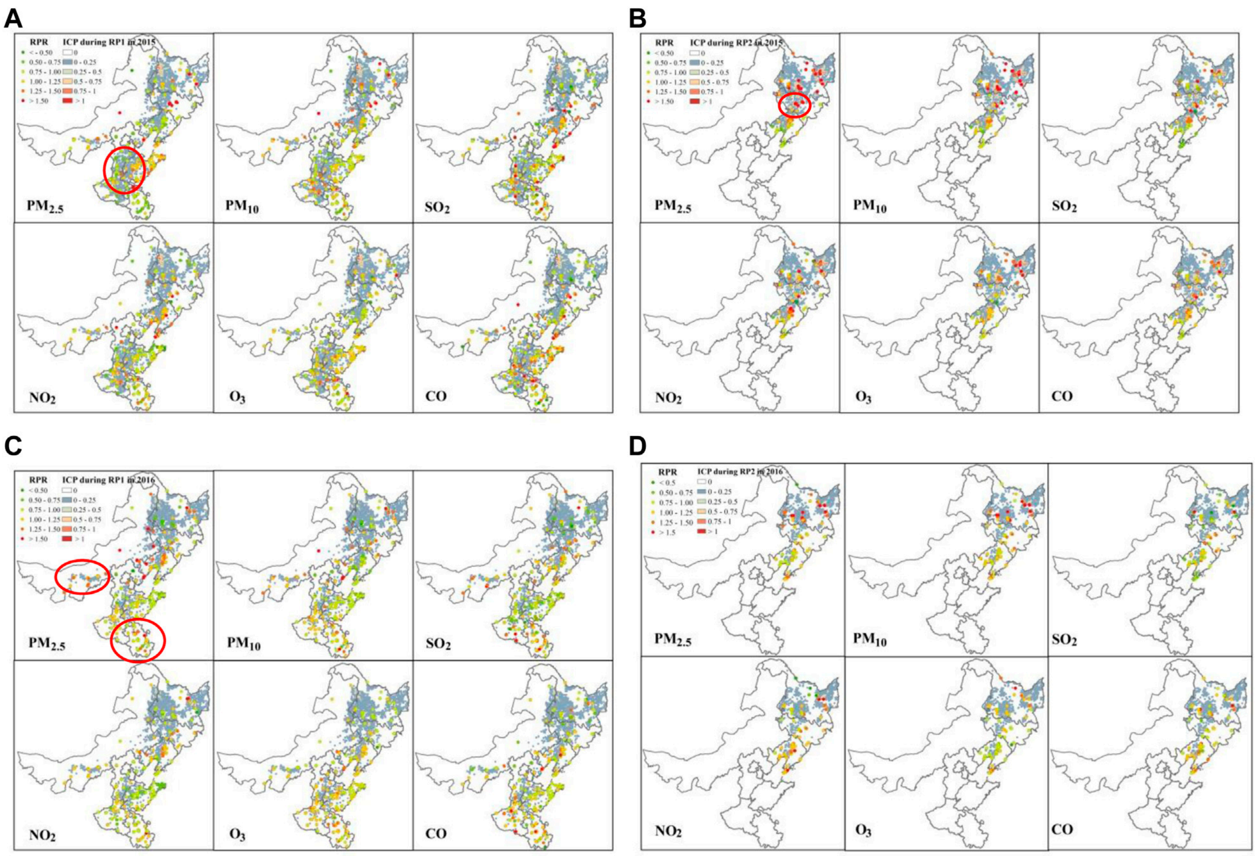

FIGURE 5
Spatial distribution of RPR for monitoring sites in different Pds: **(A)** HLJ-Pd1, JL-Pd1, LN-Pd1, IM-Pd, HB-Pd, SD-Pd, HN-Pd, and AH-Pd in 2015, **(B)** HLJ-Pd2, JL-Pd2, and LN-Pd2 in 2015, **(C)** HLJ-Pd1, JL-Pd1, LN-Pd1, IM-Pd, HB-Pd, SD-Pd, HN-Pd, and AH-Pd in 2016, and **(D)** HLJ-Pd2, JL-Pd2, and LN-Pd2 in 2016.

5.78 in HN-Pd 2016. Consequently, the mean concentrations of $PM_{2.5}$, PM_{10}, SO_2, and NO_2 decreased from 70.15 to 50.40μg/m^3, 145.22 to 96.78 μg/m^3, 30.85 to 23.92 μg/m^3, and 33.11 to 31.63μg/m^3, respectively. Therefore, in these four provinces, even if CRB was low, it still increased the mean air pollutant concentrations during the CRB seasons, especially for PM pollutants, SO_2, NO_2 , and CO. However, no influence of CRB on O_3 was observed.

In summary, according to the box-plot analysis of air pollutant concentrations during the CRB seasons, CRB decreased from 2015 to 2016, and most air pollutants decreased in the provinces. In most cases, the mean concentrations of air pollutants were higher in Pds with greater CRB. Furthermore, both the maximum concentration and the number of outliers increased during the period with greater CRB. Therefore, it is beneficial for residents to pay greater attention to controlling extremely high air pollutants during CRB seasons, rather than just controlling mean concentrations.

3.3 Spatial contributions of CRB to six major air pollutants in Pds

Additionally, the spatial contributions of CRB to the changes in air pollutants were calculated and presented. The RPR of each pollutant is presented at the monitoring-site level (Figure 5). The RPR is the increase in the ratio of one air pollutant concentration in the CRB season compared to the air pollutant concentrations before and after the CRB season. The RPR was calculated at the air quality monitoring-site level for the six major air pollutants, with a value larger than 1, indicating that CRB contributed to the increase in air pollutants in the CRB seasons (or Pd).

Heilongjiang, Jilin, and Liaoning had high RPR for almost all pollutants. Specifically, the RPR for Pd2 was the highest in 2015. In Heilongjiang, Jilin, and Liaoning, 67.88%, 72.12%, 48.48%, 80.00%, 77.58%, and 69.09% of the monitoring sites experienced an RPR greater than 1 during this period for $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3 , and CO, respectively. In 2016, because of the decrease in the number of CRB spots, the number of sites with an RPR larger than 1 decreased to 61.35%, 77.30%, 37.42%, 72.39%, 42.33%, and 58.90% for the six major air pollutants, respectively. The spatial contributions of CRB to $PM_{2.5}$, PM_{10}, and NO_2 were higher than those of the other pollutants (60–80% of monitoring sites with an RPR larger than 1 for these three air pollutants) in Heilongjiang, Jilin, and Liaoning. Spatially, the sites with a larger RPR in Heilongjiang, Jilin, and Liaoning were spread out in the region rather than being concentrated in a few hotspots.

For IM-Pd, in 2016, the sites close (see the marked area in Inner Mongolia in Figure 5C) to CRB experienced a higher RPR, especially for PM pollutants, SO_2, and NO_2. For Hebei, Shandong, Henan, and Anhui, because their Pds were all in summer (June and July), the RPR of these four provinces was analyzed together. In 2015, CRB spread in the provinces and places (see the marked places in Figure 5A), with intensive CRB spots being vulnerable to RPR greater than 1. In 2016, when the number of CRB spots decreased, sites close to the CRB did not experience a higher RPR. In contrast, in the southern part of Anhui (see the marked place in Anhui in Figure 5C), where there was no CRB spot, the sites still experienced a higher RPR. The higher RPR in this region might have been caused by social or economic factors other than CRB.

In general, the results of the spatial analysis were consistent with those of the box-plot analysis. A severe influence of CRB on air pollution has been detected in Heilongjiang, Jilin, and Liaoning. Therefore, substantial CRB leads to a large-scale impact on air pollution. When the CRB spots were limited, such as in Hebei, Shandong, Henan, and Anhui, the impact of air pollution occurred at places in or close to CRB spots and did not lead to an obvious impact on places further away from CRB.

4 Discussion

Given the immensity of crop residue in China and its adverse impact on air quality, regulations have been established to control emissions from open CRB since 1999 (Sun et al., 2016; Huang et al., 2019; Yang et al., 2020). However, despite these regulations, changes in practice have not been widely adopted by farmers (Yang et al., 2020) because of difficulties in the sustainable use of crop residues (Huang et al., 2019). In this context, understanding the changes in air pollution during periods of intensive or substantial CRB remains a significant public issue.

Although many studies have calculated the annual emissions of CRB in China for various air pollutants using emission factors from different aspects, short-term changes in air pollutants during the CRB seasons remain a concern for the public and government. Previous studies have confirmed a decrease in air quality during seasons with intensive CRB for PM pollutants on different scales (Chen et al., 2013; You et al., 2015; Shen et al., 2017; Zhuang et al., 2018a; Luo et al., 2020; Maji et al., 2020; Shen et al., 2020). The results of our study are consistent with those of previous studies regarding PM pollutants; seasons with intensive CRB led to higher PM pollutants.

Many studies have suggested that PM air quality had improved from 2015 to 2016 because of the introduction of relevant laws and regulations, such as the Five-year Air Pollution Prevention and Control Action Plan in China. Silver et al. (2018) found that in China, from 2015 to 2017, $PM_{2.5}$ and SO_2 concentrations decreased at 53% and 59% of ground-based monitoring stations, respectively. Similar conclusions were reported in other studies (Li et al., 2019; Zhai et al., 2019). According to the daily concentrations of air pollutants presented in our study, PM pollution had reduced from 2015 to 2016, which is consistent with previous studies (Song et al., 2017; Li et al., 2019).

There are multiple anthropogenic sources of air pollution, including industrial processes and fossil fuel combustion (Song et al., 2017), motor vehicles (Liao et al., 2015), and waste burning. However, these sources usually show annual rather than seasonal trends. As an important source of air pollution, seasonal changes in CRB may lead to seasonal changes in air pollution. The results showed a drop in CRB spots from 2015 to 2016, which could be a contributor to air quality improvement in China (Yamaji et al., 2010; Chen et al., 2015; Wen et al., 2018). Wen et al. (2018) analyzed the spatiotemporal variations of daily $PM_{2.5}$ concentrations in Jilin Province at the city level. It was found that the densest burning points were mainly distributed in Changchun city and Siping city, with the 2-year average $PM_{2.5}$ concentration being the highest. The authors suggested that straw burning was an important source of $PM_{2.5}$ in the plain and meadow areas of Jilin Province. This conclusion is consistent with our results. As the ICP represents in Figure 5, Changchun city and Siping city experienced higher RPR (see the read points in the circle of Figure 5B) values along with larger ICPs. Similarly, Chen et al. (2015) confirmed the contribution of CRB to air quality improvement in northeastern China. The air quality in China improved from 2015 to 2016, and the contribution of the drop in CRB has been confirmed by our study and other studies. For other pollutants, Yamaji et al. (2010) used a regional chemical transport model to investigate changes in O_3, CO, black carbon, and organic carbon concentrations during the Mount Tai Experiment in 2016. This study identified that high O_3, carbon monoxide, black carbon, and organic carbon concentrations occurred during periods of substantial CRB, which was consistent with our study. Our research focused on temporal and spatial aspects to reveal the changes in all air pollutants during the CRB seasons. It was found that at the provincial level, intensive CRB would lead to an obvious increase in PM pollutant concentrations, especially in northeast China during the CRB season, and the increase in the air pollutant concentration in space was consistent with the spatial distribution of CRB. The present study investigated the changes in six major pollutants,

TABLE 2 Ratio of stations exceeding health risk limitations in different periods in 2015.

Province	Periods	$PM_{2.5}$		PM_{10}	
		WHO	CAAQS grade II	WHO	CAAQS grade II
HLJ	Before HLJ-Pd1	0.72	0.13	0.69	0.14
	HLJ-Pd1	0.79	0.07	0.78	0.10
	Whole year	0.70	0.22	0.66	0.13
	Before HLJ-Pd2	0.44	0.08	0.48	0.04
	HLJ-Pd 2	0.78	0.36	0.76	0.25
	Whole year	0.70	0.22	0.66	0.13
JL	Before JL-Pd1	0.91	0.10	0.80	0.10
	JL-Pd1	0.94	0.20	0.92	0.17
	Whole year	0.83	0.29	0.83	0.18
	Before JL-Pd2	0.93	0.43	0.93	0.20
	JL-Pd 2	0.95	0.66	0.90	0.43
	Whole year	0.83	0.29	0.83	0.18
LN	Before LN-Pd1	0.82	0.27	0.81	0.21
	Pd1	0.88	0.26	0.94	0.26
	Whole year	0.79	0.26	0.81	0.17
	Before LN-Pd2	0.61	0.12	0.69	0.06
	LN-Pd 2	0.82	0.32	0.84	0.16
	Whole year	0.79	0.26	0.81	0.17

rather than PM pollutants, in the CRB seasons and changes in the associated health risks. The results suggest that large-scale and substantial CRB increased the mean and maximum concentrations during the periods for almost all major air pollutants, including $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO. This finding raises concerns about CRB control in the sowing and harvest seasons in rural areas, especially with dense populations, as CRB would lead to a seasonal increase in mean concentrations and maximum concentrations of multiple air pollutants. Spatially, the influence of CRB on air pollution occurs at a regional scale when CRB is relatively widespread. In contrast, the spatial impact of air pollution was evident at or near a location with CRB.

Our study detected the severe impact of CRB on PM pollutants in northeast China, and to fully understand this impact, two associated health risk standards of air pollution were used to discuss the impact degree of CRB on air quality. These two standards are grade II standards of the Chinese Ambient Air Quality (CAAQS) issued by the Chinese Ministry of Ecology and Environment (2012) (revised in 2018) and the standard released by the World Health Organization (2016) (https://www.who.int/airpollution/publications/aqg2005/en/). The ratio of monitoring sites exceeding health risk limitations in periods with intensive CRB was calculated for the provinces in northeast China for PM pollutants (see Table 2).

The results showed that the ratio of sites exceeding health risk limitations was higher for Pd than for 20 days before Pd or for the entire year. These results confirm our conclusions. The results of this study are useful for the Chinese government to formulate regulations on air quality promotion during CRB seasons from a holistic perspective. Recently, scholars have focused on air pollutant characteristics and health risks at national, regional, and provincial scales (Chen et al., 2013; Shen et al., 2017; Luo et al., 2020; Maji et al., 2020; Shen et al., 2020). However, most of these studies have been conducted at the annual change level. In this study, the associated health risks of air pollutants during a specific period with intensive CRB were discussed. As high health risks and increases in air pollutant concentration have been confirmed during CRB seasons, it is necessary to emphasize CRB management during specific periods, such as sowing and harvesting seasons.

5 Conclusion

This study reveals changes in air quality during periods of substantial CRB. The results suggest a spatially varying impact of

CRB in China. Severely poor air quality was found in northeast China, where CRB was concentrated. Using remote sensing images, higher concentrations of PM pollutants were detected during seasons with substantial CRB. Meanwhile, an increase in pollutant concentration occurred on a larger scale in northeast China compared to other provinces. The results also confirm a decrease in air pollutant concentration from 2015 to 2016, when CRB decreased. These results may contribute to CRB control in China by identifying hotspots.

However, this study has a few limitations. It used a statistical method to identify the spatial and temporal changes in air pollutant concentrations during CRB seasons. The mechanism by which CRB emits air pollutants and how these pollutants are processed in the atmosphere have not been discussed. The statistical results indicate a seasonal trend in CRB and a correlation between CRB spots and concentrations of air pollutants. This facilitates the overall control of CRB and air pollution to determine key seasons and air pollutant types in regions. In future studies, spatial statistics can be combined with simulation models, such as the Community Multiscale Air Quality Modeling System (CMAQ) and Weather Research and Forecasting (WRF) model coupled with Chemistry (WRF-Chem), to fully reflect the changes in air quality.

Data availability statement

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

Author contributions

All authors contributed to the study conception and design. TW: methodology, investigation, formal analysis, and writing the original draft; KM: data curation, software, visualization, and writing the original draft; PF: conceptualization and writing—editing; and WH: funding acquisition and writing—review and editing. All contributed to the manuscript and agree to its submission. All authors read and approved the final manuscript.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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