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Emission inventory of crop residue open burning and its high-resolution spatial distribution in 2014 for Shandong province, China

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ABSTRACT

Crop residue open burning represents an important source of biomass burning. Shandong is a large agricultural province in China. An emission inventory of crop residue open burning was established in Shandong Province for 2014 based on emission factors (EFs) and activity data (include: crop production, residue-to-production ratios, fraction of straw burned in the field, fraction of dry matter in straw and combustion efficiency for the various crops). The latest local residue-to-production ratios, fraction of straw burned in the field and EFs of each crop were used in this paper to improve the accuracy of our inventory. The results showed that the total emissions of sulfur dioxide (SO₂), nitrogen oxide (NO_x), ammonia (NH₃), methane (CH₄), non-methane volatile organic compounds (NMVOCs), fine particulate matter (PM_{2.5}), elemental carbon (EC) and organic carbon (OC) in Shandong Province for 2014 were 3.48, 25.37, 5.41, 25.9, 60.37, 379.19, 62.3, 2.86 and 21.09 kt, respectively; corn stalks represented the largest contribution of different pollutants. The emissions were spatially allocated into grid cells with a resolution of 0.1° × 0.1° by Geographic Information System (GIS) using both cultivated land use area data and fire counts as spatial surrogates. The cities with the highest emissions were Linyi, Qingdao, Binzhou and Dezhou, which have a high crop yield and densely rural population. The emissions were temporally concentrated from April to July. The uncertainties in the estimated emissions from crop residue burned in the open field were quantified by a Monte Carlo simulation.

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1. Introduction

Crop residue open burning represents an important source of biomass burning. Yevich and Logan (2003) estimated that 400 Tg of crop residues were burned in developing world in the 1990s and accounted for 33% of total mass of biomass. Streets et al. (2003) estimated that the burning of crop residues in the field comprises 34% of the total biomass burning in Asia. The emission from crop residue burned in field could be an important source of atmospheric trace gas and aerosol pollutants in local, regional and global atmospheric environments (Zhang et al., 2010, 2013). The emissions from crop residue open burning have significant effects on atmospheric chemistry, the ecological system and climate change

(Crutzen and Andreae, 1990). Some hazardous trace gases and carbonaceous aerosol emissions from crop residue burning have adverse effects on human health (Lelieveld et al., 2015). In addition, active trace gases emitted from crop residue burning, such as CO, VOCs and NO_x, are precursors of ozone (O₃) in the troposphere. The increase of O₃ in the troposphere near the ground level may raise the risk of photochemical smog, which possesses a greater threat to human health and visibility.

Domestic biofuel combustion, field burning of crop residues, forest fires and grassland fires are the main components of biomass burning (Yan et al., 2006). In some European countries, such as Finland, Sweden and Portugal, emissions from residential wood combustion are the largest single source of black carbon and fine particle emissions (Gonçalves et al., 2012; Savolahti et al., 2016). However, in agricultural countries, such as China, emissions from crop residue burning could play an important role on total biomass burning emissions (Yan et al., 2006). Emissions from crop residue open burning were estimated to account for 12.8%–57.7% of various

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pollution which emission from biomass burning in China in 2007 (Lu et al., 2011). Crop residues are one of the oldest biomass fuels in China (Cao et al., 2008). However, with the rapid increase of the harvest yield in agriculture due to better agricultural management and technology in China since the 1980s, the proportion of crop residues that were used as energy and fodder dropped substantially (Cao et al., 2007). For instance, the ratio of crop residues used as energy dropped from 60% to 70% in 1980s to 32.26% in China, in 2008 (Ma, 1980). Therefore, the open burning of crop residue has become increasingly common to eliminate waste after harvesting. In the past few years, with the reduction of emissions from the industrial and transportation sectors following stringent control measures, the effect of crop residue burning on atmospheric pollution is gradually being recognized by the government, scholars and public (Permadi and Kim Oanh, 2013; Udeigwe et al., 2015; Zhang et al., 2008). With the strict prohibition of such burning implemented in China, agricultural fires in croplands have decreased significantly in China. However, there are still a few fires that can be detected by satellites (Liu et al., 2015; Wang et al., 2014). Furthermore, due to emissions from crop residue open burning concentrated in the harvest season, a large amount of pollution that enters the atmosphere in this period resulted in a rapid increase of the concentration of pollutants in the atmosphere. Therefore, emissions from crop residue open burning still play a significant role in the air pollution of China (Niu et al., 2016).

Shandong is a large agricultural province and one of the major production bases of commodity grain, cotton and peanuts in China (Li, 2014). There is a large amount of crop residue produced in Shandong each year. Therefore, the impact of crop residue open burning in Shandong should not be ignored. Previous studies have shown that Shandong is one of the main contributors to the total crop residue open burning emissions in China. Li et al. (2016) estimated the emissions from crop residue burned in the field for 2012 in China and observed that Shandong has a high emissions level. Sun et al. (2016) calculated the CO₂ emissions via agricultural crop residue open burning in China and discovered that Shandong contributed the largest portion of CO₂ emissions. However, specific and high resolution regional crop residue open burning emission inventories in Shandong Province are still limited (Wang et al., 2014).

Globally, numerous efforts have been made to improve crop residue open burning emissions estimates since 1979 (Crutzen et al., 1979). Most of the emission inventories of crop residue open burning were either developed using a top-down approach or bottom-up approach (Pereira et al., 2011). A top-down approach to estimate crop residue open burning emissions is based on the burned area detected by satellite (Chang and Song, 2010; Song et al., 2010). It is typically used for quantifying biomass burning over large areas, such as forest and savanna fires. However, the burning of agricultural fields is often missed by satellites because of the small size and temporal impermanency (Roy et al., 2008). Thus, the emissions from crop residue field burning may be underestimated if they are based on the top-down approach. Another approach to estimate crop residue open burning emissions is the bottom-up approach, which is based on the equation established by Seiler and Crutzen (1980). Emissions are estimated by the mass of the crop residue burned in the field multiplied by the corresponding EFs. One advantage of this method is that it does not require detailed information from a large-scale survey, but the uncertainties from the numerous parameters and EFs, especially foreign parameters and EFs, could result in significant uncertainties in the total estimation. There are numerous emission inventories of crop residue open burning that have been developed using the bottom-up approach (Streets et al., 2003; Tian et al., 2011; Yan et al., 2006). Therefore, a bottom-up approach was selected in this study,

and to reduce the uncertainties in the estimation, the latest local EFs and parameters were used to develop the emission inventory.

There are two major challenges in developing an accurate emission inventory of the crop residue burned in the field based on the bottom-up method. One is the estimation of the straw resources, and another is establishing a spatial distribution with a high resolution (Yin et al., 2015). In regard to the first aspect, the types of straw considered in previous studies often only include the three major crops (rice, wheat and corn). In addition, the residue-to-production ratios used in previous studies are outdated, and the differences between these parameters reported in previous studies and recent studies in China are vast. In addition, it is difficult to obtain an accurate proportion of crop residues burned in fields, and therefore, many previous studies used the value estimated by Hao and Liu (1994) rather than the value obtained from an investigation, and the same value is often used for various crops. For the dry matter content in crop residue and burning efficiency, these parameters are often neglected in the calculation of previous studies (Wang and Zhang, 2008; Zhu et al., 2012). Therefore, we combined numerous recent measurements and considered almost all of the factors to estimate the emissions of crop residue open burning. For crop residue open burning, as an area of source pollution, the spatial distribution is more difficult compared to point or line sources. Most of the previous studies used a single spatial surrogate for these area sources (Zheng et al., 2009; Zhou et al., 2015). In this study, a modified approach was used to improve the resolution of the spatial distribution.

In this study, SO₂, NO_x, NH₃, CH₄, NMVOC, CO, PM_{2.5}, EC and OC emissions from crop residue burning in fields in Shandong were estimated in 2014. We used the statistical data collected by the Shandong government offices and the EFs that combined numerous recent and local measurements to initially develop this emission inventory. Next, the total emissions were allocated at the county-level, and then, they were spatially allocated to 0.1° × 0.1° grids based on fire counts and cultivated land use area data; the emissions were further temporally disaggregated to better understand the temporal characteristics. Finally, a comparison with previous studies and an uncertainty assessment were provided.

2. Data and methods

2.1. Study domain

Shandong Province, located on the eastern coast of China, is set between latitudes 34°22.9'N and 38°24.01'N and longitude 114°47.5'E and 122°42.3'E. The study domain covers 17 cities at the prefectural level and 137 counties or districts at the county level (see Fig. 1). This region covers 157,126 km², which is only approximately 1.64% of China. However, it represented 7.16% of the population, 9.34% of the Gross Domestic Product (GDP) and 7.57% of the crop yields in 2014 (NBSC, 2015).

2.2. Methods for estimating crop residue open burning emissions

The bottom-up approach was adopted to estimate the annual emissions from crop residue open burning by using the amount of crop-species residue burned in the field and the corresponding EF of each straws species, as shown in Eq. (1):

$$E_p = \sum_j 10^{-6} \times M_j \times EF_{p,j} \quad (1)$$

where p is the different pollutant type; j is the different crop species; E is the total emissions of each species pollutant (kiloton/year); M is the amount of straw resource burned in the field (ton/

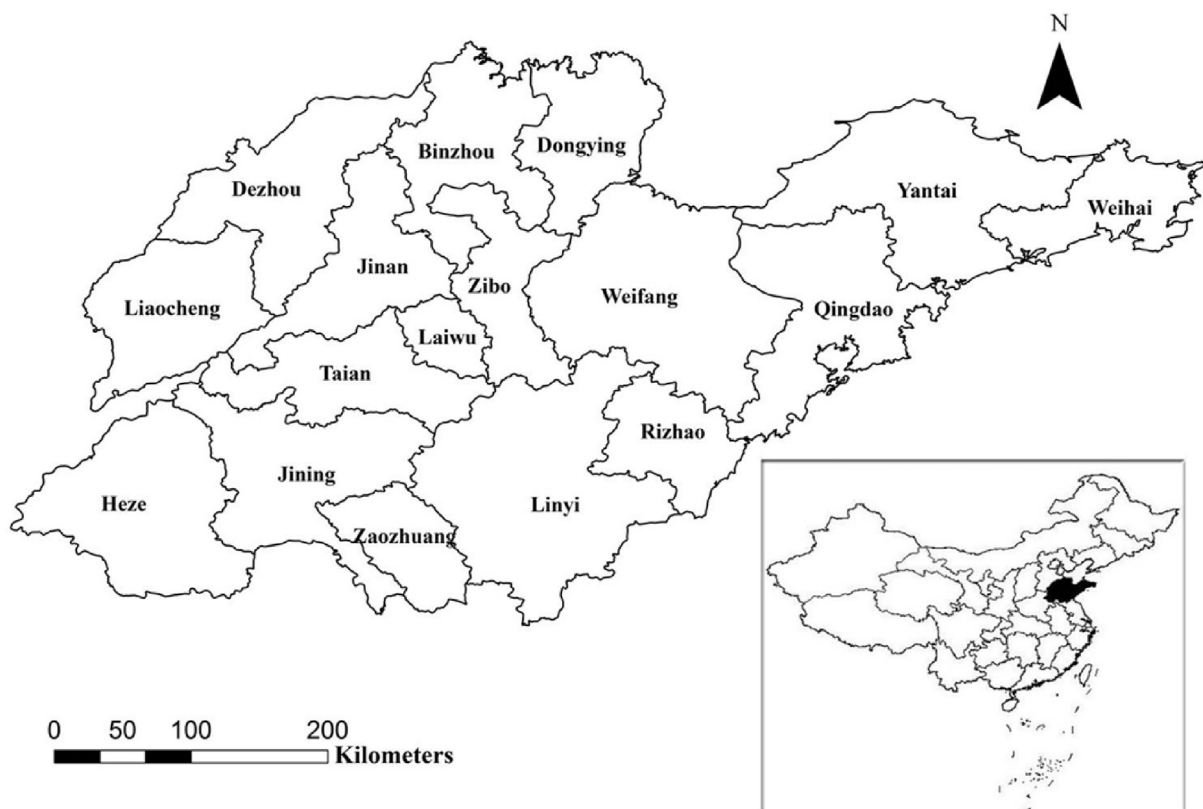


Fig. 1. Study domain and location.

year); and EF denotes the emission factor (g/kg).

2.2.1. Estimation of straw resources

The crops considered in our paper include wheat, rice, corn, beans, cotton, peanuts, rapeseed, tubers and tobacco leaves, which account for more than 95% of the total crop production in Shandong and cover the vast majority of crop residue burning (Cao et al., 2008). The mass of straw resources burned in the field was calculated using Eq. (2):

$$M_j = \sum_j P_j \times C_j \times D_j \times B_j \times F_j \quad (2)$$

where j represents the crop type; M is the amount of straw resources burned in the open field (ton/year); P is the city level production for different crops (ton/year); C is the ratio of residue-to-production of different crops; D is the fraction of dry matter in the different types of straws; B is the percentage of different crop straws burned in the open field; and F is the combustion efficiency for the various crops.

City level crop production data were taken directly from the Shandong Statistical Yearbook 2015 (SDSB, 2015). The fraction of dry matter in the various crop residues was taken from He et al. (2015). The values for the percentage of crop residues that were burned in the fields were derived from the survey by Zhang (2013). The burning efficiencies of each crop were compiled from Zhang (2009).

In previous studies (Cao et al., 2006; Chen et al., 2015; Tian et al., 2011), residue-to-production ratios were generally obtained from the China Association of Rural Energy Industry, whose values are usually based on experimental measurements from 1980 to 1984. These outdated data cannot reflect the current situation and have

led to an overestimation or underestimation of crop residue in many studies. Bi et al. (2009) determined that the annual amount of corn straw resources in China for 2005 was approximately 139 million tons based on the residue-to-production of corn, which was 1.2. However, if the ratio of corn was 2, there would be an approximate 112 million ton difference between these results. Therefore, the residue-to-production ratio used in this study refers to the latest and localized experimental measurement by Bi (2010). The parameters used in the calculation are provided in Table 1.

2.2.2. Determination of the emission factors

Table 2 summarizes the EFs applied in this study. Many early studies used EFs measured in foreign experiments or applied the same value to establish the emissions. Considerable efforts, such as conducting laboratory or field measurements, were made to obtain the real emission features for various pollutants from different crops in China. However, the measured EFs often varied widely in different experiments and sometimes even varied over orders of magnitude. Therefore, in recent studies, statistics, such as the medians or means of EFs from the literature, were usually adopted to estimate the emissions. In this study, EFs from recent or local measurements conducted in the field were preferentially selected. For instance, EFs of wheat straw and corn straw were chosen from Li et al. (2007), whose experiments were conducted at Shandong province. For some crops, very few studies have measured their EFs, and therefore, the EFs of these crops used in this study are the same as those of cotton or peanuts. In China, the main crop residue species that are subjected to farmers' open burning activity are wheat, rice, corn and cotton (Cao et al., 2008). Therefore, most of the latest research results are based on local measurements specific to these four crop straws, and studies on the EFs from other crop residue burned in the field are limited. Fortunately, the straws of

Table 1

Parameters used in the calculation of straw resources burned in the open field.

Species of crops	Residue-to-production ratio ^a	Fraction of Dry matter in straw ^d	Fraction of straw burned in the field ^g	Burn efficiency ^h
Wheat	1.2	0.89	0.07	0.917
Rice	0.9 ^b	0.89	0.12	0.925
Corn	1.34	0.87	0.14	0.917
Beans	1.6	0.91	0.17	0.68
Cotton	4.44	0.83 ^f	0.17	0.9
Peanut	1	0.94	0.17	0.9
Rapeseed	2.57 ^c	0.83 ^e	0.17	0.9197
Tubers	0.5 ^b	0.45 ^e	0.17	0.68
Tobacco leaf	1.6 ^b	0.83 ^e	0.17	0.804

^a Unless noted otherwise, all parameters are from Bi (2010).^b Bi et al. (2009).^c Wang et al. (2012).^d Unless noted otherwise, all parameters are from the IPCC (2007).^e HG-JWGSDEP (2005).^f He et al. (2015).^g Zhang (2013).^h Zhang (2009).**Table 2**

Emission factors for crop residue open burning (unit: g/kg).

Species of crops	SO ₂	NO _x	NH ₃	CH ₄	NMVOC	CO	PM _{2.5}	EC	OC
Wheat ^a	0.85	3.3	0.37	3.4	7.48	60	7.6	0.49	2.7
Rice ^b	0.18 ^c	3.83	2.2 ^d	2.19 ^e	6.05	53.2	8.5	0.21	3.3
Corn ^a	0.44	4.3	0.68	4.4	10.4	53	11.7	0.35	3.9
Beans ^f	0.48 ^c	1.08	2.2 ^d	2.893 ^e	9.42 ^h	32.3	1.84	0.13	1.05
Cotton ^c	0.4	2.49	2.2 ^d	1.82 ^e	9.42 ^h	67.4 ^g	6.3 ^d	0.82	1.83
Peanuts ^d	0.48 ^c	5.3	2.2	5.82	9.42 ^h	102	6.3	0.8	2.3
Rapeseed ^f	0.48 ^c	1.12	2.2 ^d	3.4 ^e	9.42 ^h	34.3	3.32	0.23	1.08
Tubers ^d	0.48 ^c	5.3	2.2	5.82	9.42 ^h	102	6.3	0.8	2.3
Tobacco leaves ^d	0.48 ^c	5.3	2.2	5.82	9.42 ^h	102	6.3	0.8	2.3

^a All parameters are from Li et al. (2007) unless otherwise noted.^b All parameters are from Zhang (2013) unless otherwise noted.^c All parameters are from Cao et al. (2008) unless otherwise noted.^d All parameters are from Akagi et al. (2011) unless otherwise noted.^e Liu et al. (2011).^f All parameters are from Tang (2014), unless otherwise noted.^g Wei et al. (2012).^h Kudo et al. (2014).

wheat, rice, corn, and cotton account for approximately 93.6% of the total straw in Shandong in 2014. Therefore, the error caused by the inaccurate EFs' values for the other crops would be small.

2.3. Methods for spatial allocation

To further grid the distribution, emissions from straw burned in the field at the city level were allocated to the county level first. For the purpose of obtaining accurate county-level emissions, the mass of the grain output and fire counts of each county were both used in this study. The mass of the grain output of each county was determined from the government statistics report (SDSB, 2015). The fire counts were determined from the website on the Ministry of Environmental Protection of the People's Republic of China (<http://www.zhb.gov.cn/hjzl/dqbj/jgjsjcbg/>), which provided data from all of the Terra, Aqua and FY-3 meteorological satellites. The fire events detected in croplands were identified and extracted by the Ministry of the Environment. This procedure removed fires from grasslands, forests and cities, which could also be detected by satellite. The mass of the grain output of each county cannot reflect the different percentages of residues burned in the field in various counties. Therefore, the fire counts of each county were used to modify the spatial allocation of the emissions. City emissions were allocated at the county level, using fire counts and grain output as surrogates, as shown in Eq. (3):

$$E_{n,j} = 50\% \times \frac{FC_n}{FC_m} \times E_{m,j} + 50\% \times \frac{CY_{n,j}}{CY_{m,j}} \times E_{m,j} \quad (3)$$

where n is the different counties; j is the different crop species; m is the different cities; E is the emissions from different crops in each city or county (ton/year); FC is the number of fire counts in each city or county; and CY is the crop yield for species j in each city or county. The value of the distribution coefficient is 50%, which was compiled from Li et al. (2016).

For use in the air quality model and to explore the spatial variation characteristics of the total emissions, the county level emissions were allocated to the grid with 0.1° × 0.1° resolution based on the use of a GIS software tool using Eq. (4):

$$E_{g,p} = 50\% \times \frac{FC_g}{FC_m} \times E_{m,p} + 50\% \times \frac{CA_g}{CA_m} \times E_{m,p} \quad (4)$$

where g is the different grids; m is the different cities; p is the different pollutant type; E is the emissions of each species pollutant in each city or grid (ton/year); FC is the number of fire counts in each city or grid; and CA is the cultivated land area in each city or grid.

The spatial surrogates include cultivated land use area data and fire counts. The cultivated land use area data in this study were derived from the Geographical Information Monitoring Cloud

Platform. In previous years, most of the studies used land use area data for spatial allocation (Morán et al., 2016), but some studies used the fire counts to show the spatial variation of the crop residue open burning emissions. However, the cultivated land use area simply reflects the amount of cultivated land in this grid, but cannot reflect the amount of crop residue burned in this grid. Therefore, the spatial variation allocated by the land use area data may not reflect the real situation. For example, a region with a strict prohibition regarding crop residue burning or with a high utilization of crop residue may just burn a small amount of crop residue in the field, but the cultivated land area in this region is vast. Agricultural field burning is often missed by satellite because of the small size and temporal impermanency, and therefore, spatial variation that simply uses fire counts is not comprehensive. Therefore, the land use area data and fire counts are both used in this study as spatial surrogates.

2.4. Methods for monthly allocation

To understand the monthly variation patterns of crop residue open burning emissions throughout the year and considering that emissions from crop residue open burning are affected by both natural and anthropogenic factors (He et al., 2011), PM_{2.5} emissions and fire counts were used as illustrative examples in this paper. The fire count data and mature times of various crops in Shandong were collected to describe the monthly characteristics of crop residue open burning emissions. The distribution of monthly fire counts was derived from the survey in China conducted by Li et al. (2016), and the crops' mature periods were obtained from Wei et al. (2012). In Shandong, wheat is harvested in June, rapeseed is harvested from mid-May to mid-June and most of other crops considered are harvested in the second half of the year. Considering the fire counts occurred in July may also be affected by wheat harvest, and residues of other crops are always not burned immediately after harvesting time, because farmland is vacant during the winter. Therefore, the emissions of wheat and rapeseed are distributed from May to July, and the emissions of other crops are distributed to all through the year.

3. Results and discussion

3.1. Emissions from crop residue open burning

The total emissions of SO₂, NO_x, NH₃, CH₄, NMVOCs, PM_{2.5}, EC and OC from each city in Shandong for 2014 are summarized in

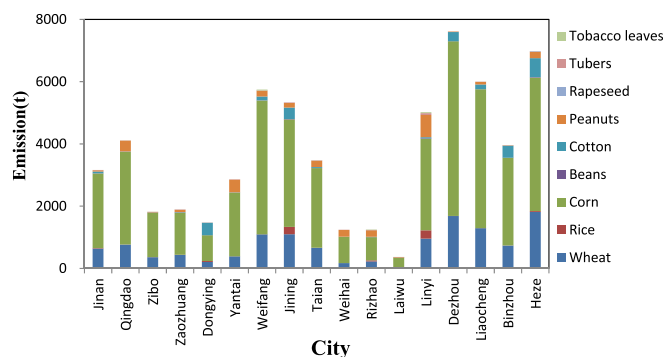


Fig. 2. City-level crop-specific PM_{2.5} emissions in Shandong for 2014.

Table 3, and county-level emissions are shown in Table S1. The total emissions of SO₂, NO_x, NH₃, CH₄, NMVOCs, PM_{2.5}, EC and OC were 3.48, 25.37, 5.41, 25.9, 60.37, 379.19, 62.3, 2.86 and 21.09 kt, respectively. The PM_{2.5} emissions from various crops in each city are shown in Fig. 2. The emissions are highly concentrated in Heze, Dezhou, Linyi, Liaocheng and Weifang, where there are large amounts of cultivated land areas and crop residues. The emissions are low in Laiwu, Weihai and Rizhao. The total yield of crop residue was estimated to be 70.9 million tons in Shandong province during 2014 based on the Shandong Statistics Yearbook.

Fig. 3 shows contributions by different crop categories to the total emissions in Shandong for 2014. For all of the types of pollutants, emissions from wheat, corn, cotton and peanut residues contributed to the majority of the emissions. Emissions from residue burning of these four crops accounted for an average of approximately 97.4% of the contribution of SO₂, NO_x, NH₃, CH₄, NMVOCs, PM_{2.5}, EC and OC emissions. Corn stalks share the largest contribution of different pollutants, with the contribution ratio ranging from 45.4% to 69.6%. This is because corn stalks have the largest production of crop residue in Shandong, and previous investigations regarding the utilization of crop straw in Shandong indicated that the proportions of crop residues burned in the field of corn stalks is higher than the proportions of other crops (Zhang, 2013). Wheat stalks are the second largest emissions (with the exception of NH₃ emissions) in Shandong, with the contribution ratio ranging from 11% to 40%. For NH₃ emissions, wheat stalks represent the fourth largest emissions. One reason is because the value of NH₃ emissions of

Table 3
Emission inventory of crop residue open burning in Shandong for 2014 (unit: kt).

City	SO ₂	NO _x	NH ₃	CH ₄	NMVOCs	CO	PM _{2.5}	EC	OC
Jinan	0.17	1.23	0.22	1.26	2.96	17.50	3.16	0.13	1.07
Qingdao	0.23	1.73	0.34	1.80	3.97	25.46	4.11	0.18	1.40
Zibo	0.10	0.70	0.12	0.72	1.68	9.83	1.82	0.07	0.61
Zaozhuang	0.11	0.77	0.15	0.80	1.83	11.33	1.90	0.08	0.65
Dongying	0.08	0.57	0.21	0.53	1.57	9.92	1.47	0.09	0.48
Yantai	0.15	1.28	0.29	1.34	2.87	19.26	2.86	0.14	0.98
Weifang	0.31	2.30	0.43	2.36	5.44	33.14	5.75	0.24	1.94
Jining	0.30	2.14	0.52	2.13	5.18	32.60	5.33	0.25	1.81
Tai'an	0.19	1.42	0.27	1.47	3.32	20.68	3.47	0.15	1.18
Weihai	0.07	0.58	0.14	0.61	1.28	8.89	1.24	0.07	0.43
Rizhao	0.07	0.59	0.15	0.61	1.28	9.28	1.25	0.07	0.43
Laiwu	0.02	0.15	0.03	0.15	0.35	2.06	0.37	0.01	0.12
Linyi	0.29	2.32	0.60	2.38	5.09	36.24	5.02	0.26	1.74
Dezhou	0.42	2.93	0.52	2.97	7.12	42.17	7.61	0.32	2.56
Liaocheng	0.33	2.34	0.41	2.39	5.63	33.56	6.00	0.25	2.03
Binzhou	0.21	1.52	0.34	1.52	3.84	22.96	3.96	0.18	1.32
Heze	0.43	2.81	0.66	2.84	6.97	44.32	6.98	0.35	2.35
Total	3.48	25.37	5.41	25.90	60.37	379.19	62.30	2.86	21.09

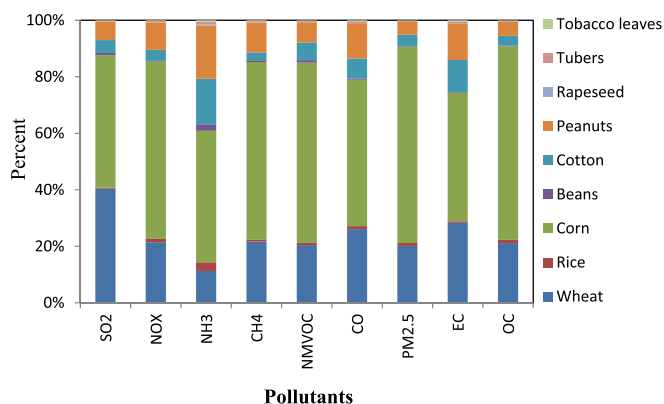


Fig. 3. Emission contributions by different crop categories in Shandong in 2014.

wheat stalks is much lower than that of corn and other crop residue (Li et al., 2007). In contrast to other provinces, peanut stalks represent the third largest emission in Shandong (however, for NH₃ emissions, peanut stalks represent the second largest emissions) and account for 5%–19% of the total emissions of various pollutants. Shandong is one of the major peanut-producing areas in China, and this large output caused by peanut residue burning contributes to high emissions for various pollutants. Cotton stalks represent the fourth largest source of emissions (for NH₃ emissions, cotton stalks represent the third largest emissions) and account for 3%–16% of the total emissions of various pollutants.

3.2. Spatial distribution emissions

The Shandong crop residue open burning emission inventory was spatially allocated using the methods described in Section 2.2. The spatial distribution of fire occurrences is shown in Fig. 4(a), and the resolution of a $0.1^\circ \times 0.1^\circ$ spatial distribution for various pollutants is shown in Fig. 4(b–h). Each point in Fig. 4(a) represents a fire count occurrence in Shandong. A large majority of the fire occurrences are concentrated south of Linyi, and some fire counts also occur south of Jining, north of Heze and west of Qingdao. A portion of the fire occurrences are band-shaped distributions along the northwest to southeast in Weifang and sparsely distributed in other cities, except Weihai, where no fires occur. Due to the increase of the weight of fire occurrences in the spatial distribution, the impact of fire occurrences on the spatial distribution can be clearly observed in these figures. The spatial distribution of different pollutants is generally similar. Heavy emissions are mostly distributed in Linyi, Qingdao, Binzhou and northeast of Dezhou, which have a large amount of cultivated land area and crop residue; and fire counts are more concentrated in these regions. However, fewer emissions are observed in central Shandong and the outlying areas of Shandong, including Weihai, Yantai, Dongying and a part of Weifang, where there are large areas of urban land use with a lower rural population and a smaller area for cultivated land. The amount of crop residue in Heze, Liaocheng, Jining and Weifang is higher than that in Linyi, but compared with Linyi, the emissions in these regions are not very heavy. The main reason for this discrepancy is that the proportion of crop residues burned in the field in these regions is lower than that in Linyi.

3.3. Monthly distribution emissions

Fig. 5 presents the monthly profiles of agricultural fire counts and crop residue open burning emissions in Shandong. As seen in

Fig. 5, the monthly distributions of both the fire counts and PM_{2.5} emissions have significant seasonal variations, and the maximum emission occurred in June. The monthly distribution of fire counts are predominantly from April to July, and the month in which most of the fires occurred is June (36.7%). The three peak values in the monthly distribution of fire counts occurred in June, April (12.8%) and October (5.3%). The peak of fire occurrences in April is because this period occurs during the first-round of seeding. In rural areas of Shandong, some crop residues are not burned immediately after harvest because farmland is vacant during the winter, except for some areas that are under cultivation of winter rapeseed and winter wheat. Therefore, local people need to clear the farmland to prepare for sowing in April. The peak in June is mainly caused by the widespread use of a wheat-corn rotation system in Shandong; therefore, corn seeds need to be sown as soon as possible after harvest wheat in June. It is worth noting that there is a large yield of crop residue in September and October because of the maturity of corn, but there are few fire counts in this period. This indicates that the prohibition of crop residue open burning has been a great success, especially for corn.

The monthly variation trend of the emissions of PM_{2.5} mass is similar to that of fire counts. The emissions are concentrated in April to July, and accounted for approximately 10.2%, 12.7%, 41.5% and 14.1% of the total emission, respectively. The month when the most PM_{2.5} emissions occurred was June, accorded with the peak of fire counts. The emissions from May to July are slightly above the fire counts, because crop residue open burning emissions during these months in Shandong are seriously influenced by the harvesting of wheat and rapeseed (Bi et al., 2009; Feng et al., 2011).

In the first half of the year, the emissions appear to be on the rise. It is related to the temperature rise and increasing agricultural activities. In the second half of the year, the emissions trend to decrease but have a little peak in October due to the harvesting of corn. It is clear that emissions estimated in the first half year are much more than the second half. The main reason is that compared with wheat straw, corn straw has more uses. For example, corn straw is always used as an important livestock fodder in winter or composted as fertilizer in the next spring.

3.4. Comparison with previous studies

There is generally a large difference between the emission inventories developed by different algorithms and data sources. The crop residue open burning emission inventories in Shandong from previous studies over the past several years were summarized and compared with our estimation. However, most of previous studies estimated such emissions on the national scales, and there are higher uncertainties in the activity data and parameters in this case.

As shown in Table 4, most of our estimation was lower than the value estimated by Li et al. (2016), but higher than the value estimated by Huang et al. (2012). The differences are most caused by the different values of the percentages of straw open burning. Huang et al. (2012) used the percentages of crop residues burned in fields with an average of 4.5% (ranging from 0.6% to 11%) for Shandong. Li et al. (2016) used the percentages of crop residues burned in fields with an average of 33% for Shandong. However, the average value of this parameter used in this study is 15% (ranging from 7% to 17%), which were based on the survey by Zhang (2013) and similar to the values used in the other studies listed in Table 4. Compared with other studies, our estimates are slightly lower. One significant reason for this result was the

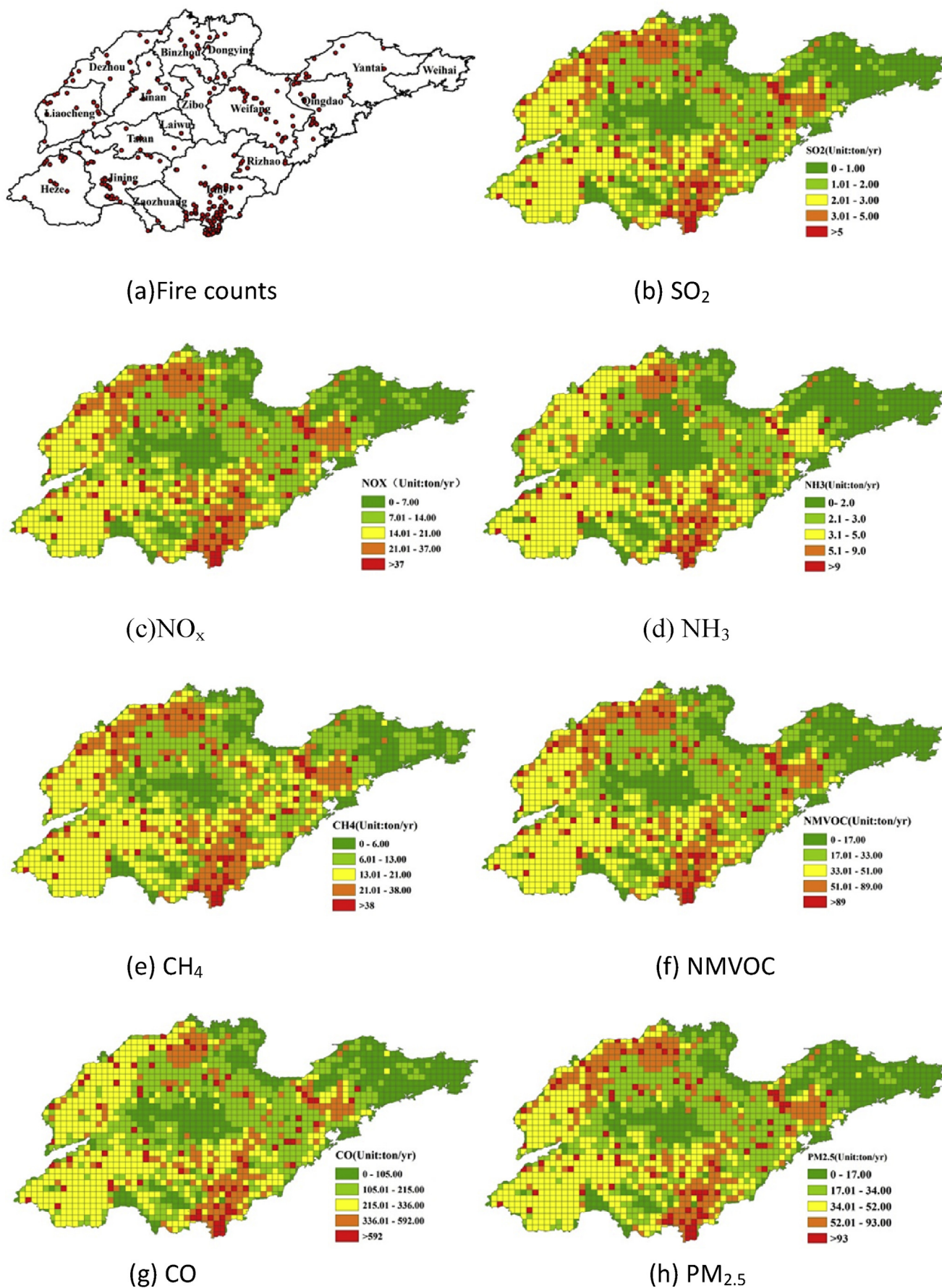


Fig. 4. The spatial distribution of fire counts and different pollutants in Shandong in 2014.

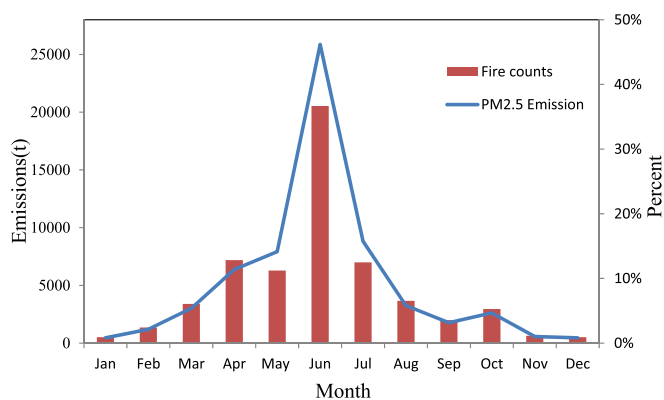


Fig. 5. Fire counts and Monthly variations of PM_{2.5} emissions in 2014.

difference in the value of EFs in the various studies. Some previous studies used EFs obtained from foreign studies, Wang and Zhang (2008) derived the EFs from 11 researches, but there are just two local researches; Huang et al. (2012) used EFs obtained from Akagi et al. (2011) and Andreae and Merlet (2001). Some studies used a same EF for various crops (Tian et al., 2011). Therefore, there are some difference between our study and previous studies. There are large difference between our study and Cao et al. (2007). This might attribute to the increasing activity data from 2000 to 2014, and the latest domestic EFs used in our study may also result in the discrepancy.

3.5. Uncertainty assessment

The uncertainty assessment of an emission inventory included a quantitative and qualitative approach (Waked et al., 2012; Zheng et al., 2012). From the qualitative point of view, uncertainties in crop residue open burning emissions were mainly from EFs and the parameters used to estimate the mass of the straw resources. To better understand the uncertainties in our inventory, a Monte Carlo simulation was used to quantify the uncertainties. Monte Carlo, as a quantitative approach, is based on the use of numerical sampling techniques in a probabilistic modeling environment. It is a frequently used approach to quantify the uncertainties in emissions from crop residue open burning. Based on the probability density distribution of activity data and EFs (Table S2), the Monte Carlo simulation ran 100,000 times to calculate the range of uncertainties with a 95% confidence interval. Table 5 lists the uncertainty ranges of each emission. Because the uncertainties from the estimation of straw resources were the same for all of the pollutants, the difference between the uncertainties of the various pollutants was mainly from the EFs. As shown in Table 5, there is a higher uncertainty in NH₃ and CO emissions compared to that of the other pollutants, which is approximately within –113.0% to 168.5% and –81.20% to 130.8%, respectively. The main reason is that the EFs of NH₃ and

Table 5

Uncertainties in emissions estimates.

Species	Uncertainty range
SO ₂	(–66.32%, 114.4%)
NO _x	(–77.56%, 126.6%)
NH ₃	(–113.0%, 168.5%)
CH ₄	(–78.08%, 126.6%)
NMVOCs	(–66.66%, 114.9%)
CO	(–81.20%, 130.8%)
PM _{2.5}	(–66.52%, 120.1%)
BC	(–66.56%, 115.0%)
OC	(–67.85%, 127.5%)

Note: the confidence interval is 95%.

CO were mainly obtained from foreign research. The estimated emissions of SO₂ and NMVOC were considered to have the least uncertainty (–66.32%–114.4% for SO₂ and –66.66% to 114.9% for NMVOC).

4. Conclusions

In this paper, a high resolution emission inventory of crop residue open burning for 2014 was established for the first time in Shandong. The latest published local residue-to-production ratios, fraction of straw burned in the field and EFs of each crop were used to improve the accuracy of the emission inventory. The emissions were distributed spatially based on the cultivated land use area data and fire counts, and the temporal distribution was established based on the fire counts and maturation times of the various crops in Shandong.

The total emissions of SO₂, NO_x, NH₃, CH₄, NMVOCs, PM_{2.5}, EC and OC from crop residue open burning were 3.48, 25.37, 5.41, 25.9, 60.37, 379.19, 62.3, 2.86 and 21.09 kt, respectively, for 2014 in Shandong. Wheat, corn, cotton and peanut residues were the main contributors of the emissions and accounted for 97.2% of the total mass of the straw resources. Corn stalks represented the largest contribution to the different pollutants and accounted for approximately 56% of the total emissions. At the country level, Dezhou, Heze, Liaocheng, Weifang and Jining were the biggest contributors and accounted for 51.7% of the total emissions. The spatial distributions of the emissions indicated that conspicuous high emissions are mainly concentrated in regions that have a high yield of crops and a densely rural population, such as Linyi, Qingdao, Binzhou and Dezhou. Analysis of the temporal variations showed that emissions from crop residue open burning have significant seasonal variations, are the highest in June, and have higher emissions from April to July. A comparison with previous studies indicated that the emissions calculated in this study were slightly lower because the value of the fraction of straw burned in the field is lower due to consideration of the strict prohibition of this type of open burning, and the difference in the value of EFs in the various studies.

Table 4

Comparison of the emissions calculated in our study with previous studies (unit: kt).

Reference	Year	SO ₂	NO _x	NH ₃	CH ₄	NMVOC	CO	PM _{2.5}	EC	OC
This study	2014	3.48	25.37	5.41	25.9	60.37	379.19	62.3	2.86	21.09
Li et al., 2016	2012	10	47	7	48	107	788	111	6	35
Tian et al., 2011	2007	1.88	35.2	–	37.7	57.1	763.1	–	6.8	–
Wang and Zhang, 2008	2006	5.1	30.4	7.1	31.6	73.8	618	183	4.2	40.6
Huang et al., 2012	2006	1	15.7	6.4	17.3	152.4	303	18.6	2.2	6.8
Cao et al., 2007	2000	0.8	46.9	23.9	34.7	–	1121.4	–	14.7	46

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <http://dx.doi.org/10.1016/j.apr.2016.12.009>.

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