Impact of crop field burning and mountains on heavy haze in the North China Plain: A case study

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Abstract. With the provincial statistical data and CFB activities captured by MODIS, we extracted a detailed CFB emission inventory in the North China Plain (NCP). The WRF-CHEM model is applied to investigate the impact of CFB on air pollution during the period from October 6 to 12, 2014, corresponding to a heavy haze incident with high concentrations of PM$_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 µm). The WRF-CHEM model generally performs well in simulating the surface species concentrations of PM$_{2.5}$, O$_3$ and NO$_2$ compared to the observations. And reasonably reproduced the observed temporal variations of wind speed, wind direction and planetary boundary layer height (PBLH). It is found that the CFB occurred in southern NCP (SNCP) have significant effects on PM$_{2.5}$ concentrations locally, causing a maximum of 34% PM$_{2.5}$ increase. Under the continuously southerly wind condition, the CFB pollution plume go through a long-range transport to northern NCP (NNCP-with several mega cities, including Beijing, the capital city of China), where few CFB occurred, resulting in a maximum of 32% PM$_{2.5}$ increase. As a result, the heavy haze in Beijing is enhanced by the CFB occurred in SNCP. Mountains also play significant roles in enhancing the PM$_{2.5}$ pollution in NNCP through the blocking effect. The mountains block and redirect the airflows, causing the pollutant accumulations along the foothill of mountains. This study suggests that the prohibition of CFB should be strict not just in or around Beijing, but also on the ulterior crop growth areas of SNCP. PM$_{2.5}$ emissions in SNCP should be significantly limited in order to reduce the occurrences of heavy haze events in NNCP region.

Key words: crop field burning; mountain effect; PM$_{2.5}$; WRF-CHEM
Crop residue burning is important for global biomass burning (Yevich and Logan, 2003; Shon, 2015), especially in agricultural countries such as China. Crop residue resources in China rank the first in the world, accounting for 17.3% of the global production (Bi et al., 2010), and increasing with the average annual proportion of 4% (Hong et al., 2015; Zhao et al., 2010). Compared with other approaches, crop field burning (CFB) is the most effective and less expensive to remove residues. The national annual average proportion of CFB to total residues is about 11-25% (Cao et al., 2008; Hao and Liu, 1994; Streets et al., 2003; Wang and Zhang, 2008; Zhao et al., 2010). Large numbers of annual CFB occur in China (Zhang et al., 2015; Yan et al., 2006), especially during the post-harvest seasons (Zhang et al., 2016; Shi et al., 2014; Cao et al., 2008). And most of the CFB occur on crop growth areas, such as the North China Plain (NCP) (Huang et al., 2012; Li et al., 2008), where have been frequently suffering haze events in recent years (Yang et al., 2015; Jiang et al., 2015; Wang et al., 2013; Wang et al., 2012).

However, CFB have adverse impacts on traffic conditions and ecology environments (Shi et al., 2014; Zhang, 2009), and release plenty of pollutants, such as CO, SO\(_2\), VOC, NOx and PM\(_{2.5}\) (Koppmann et al., 2005; Li et al., 2008). According to Guan et al. (2014) and Lu et al. (2011), annual CFB contribute about 13% of the total particulate matter (PM) emissions in China (Zhang et al., 2016). And it is more prominent during the harvest periods due to its strong seasonal dependence. Numerous studies have quantified the contribution of biomass burning and CFB to
PM pollution in China. According to Yao et al. (2016), Cheng et al. (2013), Wang et al. (2009; 2007) and Song et al. (2007), biomass burning has important impacts on the ambient PM$_{2.5}$ concentrations (15-24% in Beijing and 4-19% in Guangzhou). Yan et al. (2010) captured a heavy pollution with PM$_{10}$ concentrations higher than 350 µg m$^{-3}$ in some CFB locations. It is reported that CFB may contribute more than 30% of the PM$_{10}$ increase during CFB incidents (Zhu et al., 2012; Zha et al. 2013; Su et al., 2012). Cheng et al. (2014) report a summer case that CFB contributed 37% of PM$_{2.5}$ concentrations in the Yangtze River delta.

The impact of CFB to air quality is continental and regional. Air quality in China is influenced by the CFB occurred in Southeast Asia and on the Indian Peninsula (Qin et al., 2006). Mukai et al. (2014) have reported that CFB emissions in Southeast Asia contribute the carbonaceous aerosols in Beijing. Within China, the inter-province transported air pollutants emitted from CFB significantly affect regional PM levels and air quality (Cheng et al., 2014; Zhu et al., 2012). For Beijing, the smoke particles from CFB are expected to be one of the major components (Wang et al., 2014; Cheng et al., 2013), though the percentage of transported sources are seldom specified (Zhang et al., 2016). A recent study reports that CFB and regional transport illustrate two of the key processes of haze formation in October 2014, especially on Oct. 6$^{th}$, but without quantitative estimation in this work (Yang et al., 2015). Related quantification studies are of great importance for the control strategies of CFB in Beijing.
Yanshan and Taihang Mountains surround the NCP in the north and west (Fig. 1c). Such topography affects air pollution through PBL in complex ways (Miao et al., 2015b; Sun et al., 2013; Liu et al., 2009). Hu et al. (2014) have reported that the Loess Plateau and NCP result in a mountain-plains solenoid circulation, exacerbating air pollution over NCP. Chen et al. (2009) have found that a mountain chimney effect is dominated by mountain-valley breeze, enhancing the surface air pollution in Beijing. The mountain-plain breeze develops frequently in Beijing and may play important roles in modulating the local air quality (Miao et al., 2015b; Hu et al., 2014; Chen et al., 2009). Miao et al. (2015a) found that the mountains played a significant role in the sea-land aerosol circulation and the pollutants could be transported and accumulated in the NCP areas along the mountains, which is treated as the blocking effect (Zhao et al., 2015).

In this study, we analyzed a heavy haze episode occurred in NCP region from "LT" 12:00 6th to 00:00 12th October in 2014. During the period, the average PM$_{2.5}$ concentrations are much higher than class II standard in both SNCP and NNCP. The characteristics of the air pollution were analyzed based on PM$_{2.5}$ concentration. Depending on the satellite-based observations of Moderate Resolution Imaging Spectroradiometer (MODIS), a large number of CFB occurred in SNCP, whereas few CFB happened in NNCP. A more detailed CFB emission inventory was extracted. Thereafter we analyzed the regional transport of CFB emissions from SNCP to NNCP driven by prevailing southerly winds. Under the continuously southerly wind condition, the mountains play important roles for the northward transport, and cause
the accumulation of the aerosol pollutants at the foothill of the mountains. We also analyzed the impact of mountains (especially the Taihang Mountains and the Yanshan Mountains) on the air pollution transport.

2 Description of data

2.1 Geographical Location

In order to study the effect of CFB on local and regional air pollution, the research domain locates in eastern China, covering a large regional area (more than 10 provinces) (see Fig. 1a). The NCP region is in the middle of the research domain, with two mountains in the north and west. The Yanshan Mountains locate in the north of NCP with east-west directions, and the Taihang Mountains locate in the west of NCP with southwest-northeast directions (Fig. 1b). Figure 1c displays the distribution of online sampling sites and CFB captured by MODIS during the haze episodes. We defined two regions of interests according to CFB occurrences, topographic conditions, industrial and agricultural developments. One is the northern NCP (NNCP), including two mega cities (Beijing and Tianjin) and the north part of Hebei province, where only few CFB occurred. Another is the southern NCP (SNCP), where substantial CFB occurred during the haze episodes as shown in Fig. 1c. Because of the severe haze problem in the capital city of China (Beijing), one of the main focuses is to study the long-range transport of CFB pollution from SNCP to NNCP.
2.2 Meteorological conditions

The reanalysis meteorological data, including wind direction, wind speed and PBLH were obtained from the European Centre for Medium-range Weather Forecasts (ECMWF), with a spatial resolution of $0.125^\circ \times 0.125^\circ$. The data is available at: http://www.ecmwf.int/products/data/. The average wind direction and wind speed are displayed in Table 1. During the haze episode, the mean wind directions are $174.8^\circ$ in NNCP and $165.2^\circ$ in SNCP, and the average wind speeds are $2.4 \text{ m s}^{-1}$ in both NNCP and SNCP. The results suggest that the prevailing winds are continuously southerly winds with weak wind speeds, which is favorable to the pollution long-range transport from SNCP to NNCP, which has been reported as one of the major contributors to haze formation in the Beijing City (Tie et al., 2015).

2.3 PM$_{2.5}$ Measurements

The hourly PM$_{2.5}$ mass concentration were continuously monitored by the Ministry of Environmental Protection (MEP) of China (http://datacenter.mep.gov.cn), including 5 sites in NNCP and 7 sites in SNCP (indicated by green crosses in Fig. 1c). The data was updated from the website: http://www.pm25.in/. Table 1 summarizes the site information and the observed PM$_{2.5}$ concentrations. During the study period, the average PM$_{2.5}$ concentrations are 200.0 µg m$^{-3}$ in NNCP and 184.1 µg m$^{-3}$ in SNCP. The measured PM$_{2.5}$ concentrations are much higher than class II standard (daily mean of 75 µg m$^{-3}$), indicating an occurrence of heavy pollution event. We analyzed the characteristics of the air pollution based on PM$_{2.5}$ concentration simulated by
WRF-CHEM. Meanwhile, it is worth to note that the highest PM$_{2.5}$ concentrations occurred along the foothill sites of the Taihang Mountains. At the foothill sites of BJ, BD, SJZ and XT, PM$_{2.5}$ concentrations are 245.5, 287.7, 257.9, and 320.1 µg m$^{-3}$, respectively. The mean PM$_{2.5}$ concentration in these 4 sites is 277.8 µg m$^{-3}$, much higher than 147.2 µg m$^{-3}$ averaged from the other sites. Considering the continuously southerly winds and the topographic conditions, we studied the impact of the mountains on the air pollution transport.

3 Methods

3.1 Model description

We use Weather Research and Forecasting Chemical model (WRF-CHEM) (Grell et al., 2005) to simulate the spatial and temporal variability of PM$_{2.5}$ concentration. The specific version of WRF-CHEM model is developed by Li et al. (2010; 2011; 2012), with a new flexible gas phase chemical module and the CMAQ (version 4.6) aerosol module developed by US EPA (Binkowski and Roselle, 2003). The wet deposition follows the CMAQ method and the dry deposition is parameterized following Wesely (1989). The photolysis rates are calculated using the FTUV (Li et al., 2005; Tie et al., 2003), in which the impacts of aerosols and clouds on the photochemistry are considered (Li et al., 2011). The gas-phase chemistry was represented in the model by the modified RADM2 (Regional Acid Deposition Model, version 2) gas-phase chemical mechanism (Stockwell et al., 1990; Chang et al., 1987). Meanwhile, the ISORROPIA Version 1.7 (http://nenes.eas.gatech.edu/ISORROPIA/) is utilized to
simulate the inorganic aerosols, which is primarily used to predict the thermodynamic equilibrium between the ammonia-sulfate-nitrate-chloride-water aerosols and their gas phase precursors of H$_2$SO$_4$-HNO$_3$-NH$_3$-HCl-water vapor. The Yonsei University (YSU) PBL scheme (Hong et al., 2006), Lin microphysics scheme (Lin et al., 1983), Noah land-surface model (Chen and Dudhia, 2001) were utilized. The model has been successfully applied in several regional pollution studies (Tie et al., 2009; Tie et al., 2007; He et al., 2015).

The WRF-CHEM model is configured with resolution of 6 × 6 km (200 × 300 grid cells) centered in (117°E, 39°N). Vertical layers extend from the surface to 50 hPa, with 28 vertical layers, involving 7 layers in the bottom of 1 km. The meteorological initial and boundary conditions were gathered from NCEP FNL Operational Global Analysis data. The lateral chemical initial conditions were constrained by a global chemical transport model-MOZART4 (Model for Ozone and Related chemical Tracers, Version 4) 6-hour output (Emmons et al., 2010; Tie et al., 2005). For the episode simulations, the spin-up time of the WRF-CHEM model is 3 days.

The surface emission inventory used in this study was obtained from the Multi-resolution Emission Inventory for China (MEIC) (Zhang et al., 2009), which is an update and improvement for the year 2010 (http://www.meicmodel.org). The emission inventory estimated only anthropogenic emission such as non-residential sources (transportation, agriculture, industry and power) and residential sources related to fuel combustions. The biogenic emissions are calculated on-line with the WRF-CHEM model using the MEGAN model (Guenther, 2006). Additionally, we
added emission from CFB in the present study.

### 3.2 Crop field burning emissions

We analyzed the annual and monthly number of open crop fire events captured by MODIS in the research domain from 2008 to 2014. In the NCP region, the CFB activities gradually increase from the minimum fire events of 12,000 times in 2008 to 27,000 times in 2014 (Fig. 2a), suggesting that the CFB is not efficiently controlled in this region. This situation may be resulted from the limitation of local enforcement of regulation despite CFB have already been banned (Zhang and Cao, 2015; Shi et al., 2014). The CFB have a seasonal pattern due to the post-harvest activities with two distinct peaks in summer and autumn, especially in June (33-59%) and October (6-19%) (Fig. 2b). The strong seasonal dependence character suggests that the CFB emissions during October are much larger than annual averages. In order to have the detailed horizontal distribution of the pollutant emissions of CFB, we elaborated a method to generate emission inventory using the satellite data of “MODIS Thermal Anomalies/Fire product (MOD/MYD14DL)”. The MOD/MYD14DL product can detect small opening fires (<100 m²) (Giglio et al., 2003) and produce the geographic location of open fire activities (van der Werf et al., 2006). In this study, the CFB was defined as MOD/MYD14DL active fires occurred over the cropland, which is classified by the MODIS Combined Land Cover Type product (Friedl et al., 2010). Firstly, we estimated the CO emission of CFB, utilizing a widely used method (Streets et al., 2003; Cao et al., 2008; Zhang et al., 2008; Ni et al., 2015a) based on the
annual provincial statistical data. The provincial emission of crop residues burning can be calculated by Eq. (1):

\[ E_{i,CO} = \sum_{k=1}^{3} P_{i,k} \times F_i \times D_k \times R_k \times CE_k \times EF_{co} \]  

(1)

where \( i \) stands for each province and \( k \) for different crop species of rice, corn and wheat. \( E_{i,CO} \) stands for CO emission from CFB of \( i \)-th province in gigagrams [Gg]. \( P_{i,k} \) is the yield of crop in Gg. \( F_i \) is the proportion of residues burned in the field. \( D_k \) is the dry fraction of crop residue (dry matter). \( R_k \) is the residue-to-crop ratio (dry matter). \( CE_k \) is the combustion efficiency and \( EF_{co} \) is the emission factors of CFB. The \( P_{i,k} \) values were taken from an official statistical yearbook (NBS, 2015) (Table S1), and the \( F_i \) on a provincial basis were taken from Wang and Zhang (2008) and Zhang Yisheng (Unpublished doctor thesis-in Chinese) (Table S1). The parameters of \( D_k, R_k, \) and \( CE_k \) are listed in Table S2. The \( EF_{co} \) from CFB was summarized range from 52 to 141 g kg\(^{-1}\) in China (Table S3). In this study, we used 111 g kg\(^{-1}\) as the average \( EF_{co} \) of crop residue, which was used to estimate the emissions from open burning (Wiedinmyer et al., 2011).

The provincial CO emission was temporally and spatially allocated according to the CFB activities. The detailed daily CO emission of \( k \)-th grid \( (E_{k,co}) \) was calculated using Eq. (2):

\[ E_{k,CO} = \frac{FC_k}{FC_i} \times E_{i,CO} \]  

(2)

where \( FC_k \) and \( FC_i \) are the total CFB fire counts in \( k \)-th grid and \( i \)-th province, respectively (Table S1).
Thereafter, the emissions of various gaseous and particulate species \((E_{\text{spec1}})\) were calculated by the Eq. (3). And individual chemical compounds \((E_{\text{spec2}})\) were calculated by Eq. (4).

\[
E_{k,\text{spec1}} = \frac{E_{F_{\text{spec}}} \times E_{k,\text{CO}}}{E_{F_{\text{CO}}}}, \quad (3)
\]

\[
E_{k,\text{spec2}} = E_{k,\text{NMOC}} \times \text{scale}, \quad (4)
\]

where \(E_{k,\text{spec1}}\) and \(E_{k,\text{spec2}}\) are the \(k\)-th grid emission of the specify WRF-CHEM species; \(E_{\text{spec1}}\) and \(EFco\) are the emission factors of CFB; \(E_{k,\text{NMOC}}\) is NMOC emission in the \(k\)-th grid calculated by Eq. (3); \text{scale} is the value to convert NMOC emissions to WRF-CHEM chemical species. The emission factors for gaseous and particulate species and \text{scale} to convert NMOC emissions to WRF-CHEM chemical species from CFB were taken from available datasets (Wiedinmyer et al., 2011; Akagi et al., 2011; Andreae and Merlet, 2001), which were summarized by Wiedinmyer et al. (2011) (Table 2).

4 Results and discussions

4.1 Evaluate the Crop field burning emission

The provincial CO emissions of CFB were estimated based on Eq. (1), and there was 8.2 Tg CO emitted from CFB in 2014 (Table S1). This result is comparable to previous studies, which is 4.6-10.1 Tg yr\(^{-1}\) (Cao et al., 2008; Ni et al., 2015; Streets et al., 2003; Yan et al., 2006). According to the MODIS observations, a large number of CFB occurred in SNCP, including provinces of Henan with 61% and Shandong with 22%. Most of CFB occurred on Oct. 6\(^{th}\) and 7\(^{th}\), accounting for 75% (Table 3).
Table 4 shows the CFB emissions of gaseous and particulate species on Oct. 6th and 7th, including the mega cities of Beijing and Tianjin, and provinces of Hebei, Henan and Shandong in NCP. Figure 3 displays the CFB activities and related CO emission on Oct. 6th and 7th. Most of the pollutants are emitted from Henan in SNCP, accounting for 73% on Oct. 6th and 65% on Oct. 7th. Plenty of pollutants emitted from CFB on Oct. 6th, producing more than 5.1 Gg PM$_{2.5}$ and 98.0 Gg CO (1 Gg = 10$^9$ g).

4.2 Statistical characteristics of the evaluation

The characteristics of the haze pollution were defined by PM$_{2.5}$ concentration, which is significantly affected by the local wind fields and PBLH in the NCP region (Tie et al., 2015). In order to evaluate the model performance, the model simulations were compared with the measured results in both species concentrations (PM$_{2.5}$, O$_3$ and NO$_2$) and meteorological parameters (wind speed, wind direction and PBLH). The normalized mean bias (NMB) and correlation coefficient (R) were used to quantify the performance.

\[
NMB = \frac{\sum_{i=1}^{N} (P_i - O_i)}{\sum_{i=1}^{N} O_i},
\]

(5)

\[
R = \frac{\sum_{i=1}^{N} (P_i - \bar{P})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^{N} (P_i - \bar{P})^2 \sum_{i=1}^{N} (O_i - \bar{O})^2}},
\]

(6)

where $P_i$ is the predicted results and $O_i$ represents the related observations. N is the total number of the predictions used for comparisons. Meanwhile, $\bar{P}$ and $\bar{O}$ are the average prediction and related mean observation, respectively.

Figure 4 shows the measured and calculated temporal variations of regional average species concentrations, including PM$_{2.5}$, O$_3$ and NO$_2$. The WRF-CHEM model
reproduced the pollution episode well, with a good agreement with observations. The
correlation coefficients (R) of simulated and measured PM$_{2.5}$ concentrations are 0.88
in both NNCP and SNCP (Fig. 4a). The simulations are overall lower than the
observations with NMB of -12% in NNCP and -7% in SNCP. Considering the high
average PM$_{2.5}$ concentration with 200.0 µg m$^{-3}$ in NNCP and 184.1 µg m$^{-3}$ in SNCP,
obvious underestimates exist with the overall concentrations of 24.0 µg m$^{-3}$ in NNCP
and 12.9 µg m$^{-3}$ in SNPC. This may be related to the CMAQ (version 4.6) aerosol
module, which is likely to underestimated OM due to the uncertainty in secondary
organic aerosols mechanism (Baek et al., 2011). Meanwhile, the underestimates are
also related to the negative bias in S3, which may be related to cloud contamination
(Fig. S1). Whereas this has only a few impacts on the estimation of CFB contribution
since few CFB occurred during S3. The simulations of O$_3$ and NO$_2$ are also agree well
with observations, with R greater than 0.77 and absolute NMB lower than 17% (Fig.
4b and 4c). Figure 5 displays the measured and calculated temporal variations of
regional average meteorological parameters, including wind speed, wind direction,
and the PBLH in both NNCP and SNCP. The comparisons between simulated and
observed wind fields show good agreements (Fig. 5a and 5b), with all the R higher
than 0.64, and the absolute NMB are no more than 15%. Meanwhile, the R of PBLH
is larger than 0.88 and the absolute NMB is no more than 10% (Fig. 5c).

4.3 Characteristics of the heavy pollution events

According to the evolution of PM$_{2.5}$ concentration (Fig. 4a), the haze episode can be
divided into three stages: (I) pollution formation stage (S1, 12:00 6th - 00:00 8th), (II) pollution outbreak stage (S2, 00:00 8th - 00:00 10th) and (III) pollution clear stage (S3, 00:00 10th - 00:00 12th). The major characteristics of each stage are briefly summarized below. Related simulations in bracket follow the detailed observations.

- S1 (pollution formation): It is dominated by a continuously southerly wind, with mean wind speed of 2.5 (2.7) m s\(^{-1}\) in NNCP and 3.0 (3.6) m s\(^{-1}\) in SNCP. The backward trajectories, with the HYSPLIT model online version, of BJ, TJ and BD during S1 reflected how the CFB influenced the NNCP region (Fig. 6). The air mass mainly came from the south, originating from the SNCP region. The pollutants are continuously transported from SNCP to NNCP, leading to pollutants accumulation in NNCP, which is characterized by the steady rising of PM\(_{2.5}\) concentration in NNCP from 20.6 (41.0) µg m\(^{-3}\) (at 12:00 Oct. 6th) to 242.7 (217.5) µg m\(^{-3}\) (at 00:00 Oct. 8th) (Fig. 4 a1).

- S2 (pollution outbreak): During S2, the air pollution deteriorates. It is a relative stable period of heavy pollution with average PM\(_{2.5}\) concentration of 252.0 (241.2) µg m\(^{-3}\) in NNCP and 214.1 (235.0) µg m\(^{-3}\) in SNCP, which are higher than those in other stages. This phenomenon may be related to the relative lower wind speed and PBLH.

- S3 (pollution clear): During S3, the southerly winds gradually decrease, and turn to be northerly at the end of S3. Clean airs from the north region of China obviously improve the air quality. Compared with S2, the average PM\(_{2.5}\) concentrations are decreased in both NNCP and SNCP.
There were several important issues shown in the results, and should be addressed. (1) The PM$_{2.5}$ concentrations are extremely high during the S2 period, and the daily average concentrations are greater than the Chinese National Standard (75 $\mu$g m$^{-3}$) by 2-3 times. (2) The air pollutions are severe in a large region (occurred in both NNCP and SNCP). (3) During the S1 and S2 periods, there is a time lag between SNCP and NNCP for PM$_{2.5}$ concentrations. Because it is a continuously southerly wind condition, it shows the important impact of long-range transport of PM$_{2.5}$ particles from the SNCP to NNCP.

4.4 Contributions of crop field burning

Model sensitivity studies were conducted to separate the individual CFB contribution. Two model simulations were performed, i.e., one with both anthropogenic and CFB emissions while the other with only anthropogenic emission. We calculated PM$_{2.5}$ distributions by including CFB emissions (anthropologic and CFB) and excluding CFB emissions (only anthropologic). In this study, the CFB contributions were quantified by regional average contribution in mass concentration ($CPM_{2.5}$) and daily average contribution proportion ($PPM_{2.5}$).

$$CPM_{2.5} = TPM_{2.5} - APM_{2.5},$$  \hfill (7) 

$$PPM_{2.5} = \frac{CPM_{2.5}}{TPM_{2.5}}$$  \hfill (8) 

where $TPM_{2.5}$ represents the simulated PM$_{2.5}$ concentrations considering total emission; $APM_{2.5}$ denotes the simulated PM$_{2.5}$ concentrations only considering anthropologic emissions. $CPM_{2.5}$ and $TPM_{2.5}$ are daily average value for $CPM_{2.5}$
and $TPM_{2.5}$, respectively.

**Figure 7** displays the regional observed and simulated PM$_{2.5}$ concentrations considering total emissions (anthropologic and CFB) and only anthropologic emissions. It is clearly shown that the CFB had important contributions to PM$_{2.5}$ in both NNCP (**Fig. 7a**) and SNCP (**Fig. 7b**). This is also proved by the daily average contribution proportion ($PPM_{2.5}$) of CFB (**Table 5**). The high values of $PPM_{2.5}$ in SNCP appear on Oct 6$^{th}$ with 34% and on 7$^{th}$ with 17%, when plenty of CFB occurred. Simultaneously, the high values of $PPM_{2.5}$ in NNCP appear on Oct 7$^{th}$ with 32% and 8$^{th}$ with 10%, showing a later occurrence than that in SNCP. The time lag suggests that the plume with CFB may be transported from SNCP to NNCP.

The detailed hourly CFB contributions to PM$_{2.5}$ concentrations ($CPM_{2.5}$) are displayed in **Fig. 8**. The values of $CPM_{2.5}$ in NNCP are generally lag synchronized with that in SNCP, such as P$_{N1}$ versus P$_{S1}$ and P$_{N2}$ versus to P$_{S2}$ (**Fig. 8a and 8b**). Apparently, the lagged time is not constant and varied with the wind fields. The specific details perform relaxed lag synchronized, especially between the P$_{N2}$ and P$_{S2}$.

This phenomenon further indicates that the CFB contribution in SNCP is mainly due to local emission, whereas CFB contribution in NNCP is largely resulted from long-range transport from SNCP. Indeed, the CFB pollution plume go through a long-range transport to NNCP can cause an obvious increase to PM$_{2.5}$ concentration, with the maximum daily average contribution of 32% (**Table 5**). Such a high transported contribution indicates that the CFB is not only one of the significant local pollution sources, but also a considerable regional pollution source.
To clearly show the time evolution of the CFB effect on PM$_{2.5}$ concentration, four time-points were defined in Fig. 8c, such as T1 (23:00 6$^{th}$), T2 (05:00 7$^{th}$), T3 (20:00 7$^{th}$) and T4 (19:00 8$^{th}$). At T1, prominent CFB contribution occurred in SNCP with the highest value of 71.9 µg m$^{-3}$, but accompanied with unimportant CFB contribution in NNCP with a low value of 7.7 µg m$^{-3}$. At T2, the CFB contribution in SNCP decline with a relative high value of 44.2 µg m$^{-3}$, but rise in NNCP with 51.6 µg m$^{-3}$ (near the transition between P1 and P2). At T3, the CFB contribution rapidly decreases to a low value of 24.0 µg m$^{-3}$ in SNCP, but increase to the highest with 47.0 µg m$^{-3}$ in NNCP. At T4, the CFB contributions largely decrease, becoming lesser in both SCNP (9.1 µg m$^{-3}$) and NNCP (11.4 µg m$^{-3}$). Interestingly, the CFB contribution in SNCP drops faster than that in NNCP (P2 in Fig. 8c), resulting in stronger effects in NNCP than in SNCP, as well as longer effects in NNCP.

To further understand the evolution of CFB to heavy haze pollution, we analyzed the horizontal distributions of PM$_{2.5}$ concentration ($TPM_{2.5}$) and related CFB contribution ($CPM_{2.5}$) at T1, T2, T3 and T4 (Fig. 9). The pattern comparisons between simulated and observed near-surface PM$_{2.5}$ concentrations ($TPM_{2.5}$) perform well (Fig. 9 Left Panels). Meanwhile, the regional average CFB contributions are shown in Table 6, including mass concentration and related percentage as well as the related lag-time of NNCP corresponding to SNCP. At T1, massive local pollutants are emitted from CFB in SNCP and the CFB plume had not yet been largely transported to NNCP (see $CPM_{2.5}$ of Fig. 9 T1). The CFB contribution is high in SNCP with 72.6 µg m$^{-3}$, accounting for 71% of the total PM$_{2.5}$, whereas the CFB contribution is low with 8.1
µg m$^{-3}$ in NNCP, only accounting for 21%. At T2, high CFB contribution occurred in both SNCP and NNCP with 37 µg m$^{-3}$, suggesting that plenty of CFB pollutants emitted from SNCP and had been transported to NNCP (see $CPM_{2.5}$ of Fig. 9 T2). At T3, CFB contribution rapidly reduced in SNCP with 20.2 µg m$^{-3}$ (13%). It is worth to note that the high CFB contribution with 50.4 µg m$^{-3}$ (58%) is still remained in NNCP (see $CPM_{2.5}$ of Fig. 9 T3). At T4, the CFB contribution largely decreased in both SNCP and NNCP (no more than 6%) (see $CPM_{2.5}$ of Fig. 9 T4). The lag-time of NNCP to SNCP are 7-12 hours, and gradually increase from T1 to T4, implicating that the effect of CFB remains in longer time in NNCP than in SNCP. The highest PM$_{2.5}$ concentrations are along the foothill of the Taihang Mountains (Left panels of Fig. 9), which may be related to the mountain effects.

### 4.5 Impact of mountains

Sensitivity experiments were conducted to quantify the impacts of the Taihang Mountains (referred as R-T), the Yanshan Mountains (R-Y) and both of them (R-TY) on the heavy pollution. The mountains were removed from the model calculation, in which, the altitude of mountains were reduced to the average altitude of NCP (30 m). With the reduction of altitudes of the topography, the dynamical conditions calculated from WRF-CHEM changed, which affect pollutions transport, especially along the foothill of mountains. In this study, we utilized the differences between the simulations with or without mountains to represent the effect of the topography on PM$_{2.5}$ concentration, which were calculated based on Eq. (9). As an on-line dynamical
model, the topography changes in WRF-CHEM can lead to dynamical changes, such as the wind speeds at the foothill of the mountains. This is a useful and traditional sensitivity analysis method for numerical model to quantify the mountains effects, but with some shortcomings, which are to bring uncertainties to the sensitivity experiment. Firstly, the impact of topography is complicated to be completely quantified only by the altitude remove behavior. Secondly, the initial NCEP FNL data with mountains is treated as “real” in scenarios without mountains. The sensitive configuration and related enclosing scope are displayed in Fig. S2.

\[ \text{IPM}_{2.5} = \text{RPM}_{2.5} - \text{TPM}_{2.5}, \]  

(9)

where \( \text{IPM}_{2.5} \) is the net impacts of mountains on \( \text{PM}_{2.5} \); \( \text{RPM}_{2.5} \) denotes the simulated \( \text{PM}_{2.5} \) concentration with removal behaviors, involving R-TY, R-T, and R-Y; \( \text{TPM}_{2.5} \) represents the simulated \( \text{PM}_{2.5} \) concentration considering emission of anthropologic and CFB, which is correspond with the case of R0 (Fig. S2a).

The sensitivity study period was selected from 12:00 7\(^{th}\) to 00:00 10\(^{th}\). Fig. 10 displays the elevation contours and the horizontal distributions of \( \text{PM}_{2.5} \) concentration with the effect of mountains, exhibiting a good performance of the pattern comparisons between simulated and observed near-surface \( \text{PM}_{2.5} \) concentrations. The results illustrate that the mountains had important impacts on regional \( \text{PM}_{2.5} \) concentration, especially for the region along the foothill of mountains with a heavy pollution area, covering sampling sites of BJ, BD, SJZ and XT. Here, it is attributed to the mountain blocking effect, which has two categories of influences. Firstly, the mountains block the airflows, causing pollutant accumulation and resulting in high
PM$_{2.5}$ loading at the foothill of mountains (Influence-1, block). Secondly, the mountains redirect the airflows, causing the pollutants move toward the downwind foothill areas (Influence-2, redirect). Both influences act to prevent the pollutant plume to disperse toward western mountains, causing accumulations of the air pollutants along the foothill of mountains. These two influences of mountain blocking effect are illustrated as the schematic pictures in Fig. S3.

Fig. 11 displays the simulated PM$_{2.5}$ concentration due to the mountain effects (RPM$_{2.5}$), with the three cases (R-TY, R-T, and R-Y). The heavy pollution accumulation (Fig. 10) along the foothill of mountains is significantly reduced, especially with the removal of Taihang Mountains (R-T, and R-TY) (Fig. 11 a1 and a2). In these two cases, the pollution plumes dispersed westerly (Fig. 11 b1 and b2).

The PM$_{2.5}$ concentrations increase 40-120 µg m$^{-3}$ in the western part of Taihang Mountains, and reduce 20-60 µg m$^{-3}$ in NCP. The distribution of the reduced pollution plume shows a northeast band plume, indicating the mountain blocking effect. With the removal of the Yanshan Mountains (R-Y), the high PM$_{2.5}$ concentrations are still remained along the foothill of the Taihang Mountains (Fig. 11 a3), but more pollutants are pushed forward along the foothill, toward the northeastern NCP.

Without the blocking effect of the Yanshan Mountains, the PM$_{2.5}$ concentrations increased 20-80 µg m$^{-3}$ in the northern part of the Yanshan Mountains, and decreased 10-60 µg m$^{-3}$ in the southern part of the Yanshan Mountains (Fig. 11 b3).

In the foothill sampling sites (BJ, BD, SJZ and XT), the average PM$_{2.5}$ concentrations are reduced 54.2 µg m$^{-3}$ for the case of R-T, which is much higher than the case of
R-Y (28.4 µg m$^{-3}$). For the other non-foothill sites, the average reduction is 34.7 µg m$^{-3}$ for the case of R-T, which is also much higher than the case of R-Y (2.4 µg m$^{-3}$), suggesting that the Taihang Mountains have stronger effects than the Yanshan Mountains. Meanwhile, the higher impacts in the foothill sampling sites than non-foothill sites are further demonstrated.

5 Conclusions

In recent years, the NCP region, including the capital city of Beijing, has been suffering serious haze pollution problem, especially in winter and summer. Most studies concerned about the intense secondary formation, huge regional transport of pollutants, stationary meteorological conditions and large local emission. In autumn, CFB and movement of wind based on large scale topography are important in NCP, whereas the percentage of transported CFB emission sources are seldom specified. This is probably resulted from the contingency of CFB activities during harvest period and the limitation of temporal resolution of CFB emission inventories. In this study, we extracted a more detailed CFB emission inventory based on the provincial statistical data and CFB activities captured by MODIS. The WRF-CHEM mode was applied to study the effect of CFB on the PM$_{2.5}$ concentrations in NCP, especially the evaluation of CFB plums pollution, such as local influence and long-range transportation. We get some insights of how could CFB affect the air quality in NNCP and Beijing under heavy haze condition, though more and longer studies are needed to get more representative conclusions. The results are summarized:
A more detailed CFB emission inventory was generated in NCP. The daily CFB emissions were estimated depending on CFB activities captured by MODIS. Plenty of pollutants emitted from SNCP on Oct. 6th and 7th, producing plenty of PM$_{2.5}$ pollution, but few in NNCP during the entire haze period.

The WRF-CHEM model reproduced the pollution episode with a good agreement with observations. The correlation coefficients (R) of simulated and measured PM$_{2.5}$ concentration are 0.88 in both NNCP and SNCP, and the related NMB are -12% in NNCP and -7% in SNCP. The simulated winds and PBLH are also in good agreement with observations in both NNCP and SNCP.

The WRF-CHEM model was used to investigate the impacts of CFB contribution and its evaluation on PM$_{2.5}$ concentration. The SNCP region is mainly influenced by the local CFB emissions, causing a maximum of 34% PM$_{2.5}$ increase. Whereas the NNCP region is mainly affected by the long-range transport of pollution plume emitted from CFB in SNCP, causing a maximum of 32% PM$_{2.5}$ increase in NNCP.

The research domain includes two regions of interests. One is the NNCP, including two mega cities (Beijing and Tianjin), where few CFB occurred. Another is the SNCP, where substantial CFB occurred. This study shows that there are substantially long-transport of CFB plume from SNCP to NNCP. More importantly, the effect of CFB remains in longer time in NNCP than in SNCP along the foothill areas of the Taihang Mountains, causing significant enhancement in Beijing in both time and magnitude.
Another major finding is that the mountains, surrounding the NCP in the north and west, play significant roles in enhancing the PM$_{2.5}$ pollution in NNCP through the blocking effect. Mountains block and redirect the airflows, causing the pollution accumulation along the foothill of mountains. The Taihang Mountains had greater impacts on PM$_{2.5}$ concentration than the Yanshan Mountains. On account of various factors, such as pollutant long-range transport and pollutant accumulation caused by mountain effects, the prohibition of CFB should be strict not just in or around Beijing, but also on the ulterior crop growth areas of SNCP. Other PM$_{2.5}$ emissions in the SNCP should be significantly limited in order to reduce the occurrences of heavy haze events in NNCP region, including the Beijing City.

**Acknowledgement**

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Figure Captions

Figure 1 The study area, sampling sites and crop fires. (a) The research domain and related provinces in China. (b) Topographical conditions of North China Plain. (c) Location of sampling sites and crop field burning captured by MODIS during the haze episodes. Green crosses indicate the measurement sites, and the CFB are shown by the pink dots.

Figure 2 The (a) yearly and (b) monthly crop field burning observed by MODIS in the research domain during the year of 2008 to 2014.

Figure 3 Crop field burning captured by MODIS with the background of MODIS real-time true color map (Left) and related CO emission (Right) on Oct. 6th and 7th.

Figure 4 Regional averaged temporal variations in simulated (in red) and observed (in blue) results of species concentrations of (a) PM$_{2.5}$ (b) O$_3$ and (c) NO$_2$ over the regions of NNCP and SNCP.

Figure 5 Regional averaged temporal variations in simulated (in red) and observed (in blue) results of meteorological parameters of (a) wind speed (b) wind direction and (c) PBLH over the regions of NNCP and SNCP.

Figure 6 Backward trajectories of NNCP (Beijing, Tianjin and Baoding) during S1 (LST, 12:00 6th - 00:00 8th) in different height of 100m, 500m and 1000m.

Figure 7 Hourly PM$_{2.5}$ concentration of observations (obs) and simulations (sim-total and sim-anthro) in (a) NNCP and (b) SNCP. Sim-total represents the simulations considering total emissions (anthropologic and crop field burning), whereas sim-anthro is the simulations only considering anthropologic emissions.

Figure 8 CFB contribution to PM$_{2.5}$ concentration ($CPM_{2.5}$) (a) in SNCP, (b) in NNCP and (c) their comparison. The key point-in-local-times of T1 (23:00 6th), T2 (05:00 7th), T3 (20:00 7th) and T4 (19:00 8th) are signed with blue arrow.
Figure 9 The distributions of $TPM_{2.5}$ and $CPM_{2.5}$ of the key point-in-local-times of T1, T2, T3 and T4, which represent different pollution phase of emission from crop field burning to PM$_{2.5}$. Left panels also show the pattern comparisons of simulated vs. observed near-surface PM$_{2.5}$ concentrations ($TPM_{2.5}$), with PM$_{2.5}$ observations of colored circles. Black arrows denote simulated surface winds.

Figure 10 The elevation contours and the pattern comparisons of simulated vs. observed near-surface PM$_{2.5}$ concentrations from 12:00 7th to 00:00 10th. Colored circles: PM$_{2.5}$ observations of foothill sites; Colored squares: PM$_{2.5}$ observations of non-foothill sites; Black arrows: simulated surface winds. The 200-meter contour was highlighted with bold black line.

Figure 11 The averaged spatial distribution of PM$_{2.5}$ concentration and horizontal winds during 12:00 7th to 00:00 10th. (a) Simulated PM$_{2.5}$ loading with erase behavior $RPM_{2.5}$, involving R-TY, R-T, and R-Y. (b) The related impacts of mountains to PM$_{2.5}$ ($IPM_{2.5}$), which represents the net effect of related mountains. The bold black lines were used to stress enclosing scope of each erased behavior.
Table 1. The average PM2.5 concentration, wind direction and wind speed of the observations from 12:00 6th to 00:00 12th. The sampling sites located at the foot of mountains were emphasized with bold style.

<table>
<thead>
<tr>
<th>Region</th>
<th>Site</th>
<th>Longitude (°E)</th>
<th>Latitude (°N)</th>
<th>PM$_{2.5}$ (µg/m$^3$)</th>
<th>Wind-dir (°)</th>
<th>Wind-spd (m/s)</th>
</tr>
</thead>
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<tr>
<td>NNCP</td>
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<td>40.04</td>
<td>245.5</td>
<td>185.8</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td>Langfang (LF)</td>
<td>116.73</td>
<td>39.56</td>
<td>214.7</td>
<td>177.0</td>
<td>2.4</td>
</tr>
<tr>
<td></td>
<td>Tianjin (TJ)</td>
<td>117.31</td>
<td>39.09</td>
<td>134.7</td>
<td>173.5</td>
<td>2.4</td>
</tr>
<tr>
<td></td>
<td>Baoding (BD)</td>
<td>115.49</td>
<td>38.87</td>
<td>287.7</td>
<td>171.2</td>
<td>2.2</td>
</tr>
<tr>
<td></td>
<td>Cangzhou (CZ)</td>
<td>116.87</td>
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<td>117.3</td>
<td>166.6</td>
<td>2.5</td>
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<td></td>
<td>Shijiazhuang (SJZ)</td>
<td>114.49</td>
<td>38.04</td>
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<td>175.2</td>
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<td></td>
<td>Hengshui (HS)</td>
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<td>166.7</td>
<td>163.7</td>
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<td>Dezhou (DZ)</td>
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<td>37.47</td>
<td>152.4</td>
<td>162.7</td>
<td>2.6</td>
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<tr>
<td></td>
<td>Xingtai (XT)</td>
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<td>37.09</td>
<td>320.1</td>
<td>198.1</td>
<td>2.3</td>
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<td>Liaocheng (LC)</td>
<td>116.00</td>
<td>36.46</td>
<td>139.7</td>
<td>158.4</td>
<td>2.6</td>
</tr>
<tr>
<td></td>
<td>Hezhe (HZ)</td>
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<td>35.26</td>
<td>105.0</td>
<td>138.9</td>
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</tr>
<tr>
<td></td>
<td>Zhengzhou (ZZ)</td>
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<td>34.79</td>
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<td>2.4</td>
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<td>Shijiazhuang (SJZ)</td>
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Table 2. The gaseous and particulate species emission factors (g/kg) and scales to convert NMOC emissions (kg day\(^{-1}\)) to WRF/Chem chemical species (moles-species day\(^{-1}\)) from crop field burning. The detailed chemical species are described by Stockwell et al. (1990).

<table>
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<tr>
<th>Gaseous species</th>
<th>Particulate species</th>
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<td>CO (^1)</td>
<td>OC (^3)</td>
</tr>
<tr>
<td>NOx (^1)</td>
<td>BC (^3)</td>
</tr>
<tr>
<td>NO (^1)</td>
<td>PM(_{2.5}) (^1)</td>
</tr>
<tr>
<td>NO(_2) (^2)</td>
<td></td>
</tr>
<tr>
<td>SO(_2) (^3)</td>
<td></td>
</tr>
<tr>
<td>NH(_3) (^4)</td>
<td></td>
</tr>
<tr>
<td>NMOC (^1)</td>
<td></td>
</tr>
<tr>
<td>111</td>
<td>3.3</td>
</tr>
<tr>
<td>3.5</td>
<td>0.69</td>
</tr>
<tr>
<td>1.7</td>
<td>5.8</td>
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Chemical-compounds-to-NMOC scales\(^{1,2}\)

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<th>ETH</th>
<th>HC3</th>
<th>HC5</th>
<th>OL2</th>
<th>OLT</th>
<th>OLI</th>
<th>TOL</th>
<th>CSL</th>
<th>HCHO</th>
<th>ALD</th>
<th>KET</th>
<th>ORA2</th>
<th>ISO</th>
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<td>0.43</td>
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<td>1.09</td>
<td>0.27</td>
<td>0.20</td>
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<td>1.84</td>
<td>3.05</td>
<td>0.83</td>
<td>2.19</td>
<td>0.60</td>
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</table>

a. The values were taken from Andreae and Merlet (2001); b. The values were taken from Wiedinmyer et al., (2011); c. The values were taken from Akagi et al., (2011)
Table 3. The fire counts of crop field burning detected by the MODIS in the provinces over NCP during the haze episode (from Oct. 6th to 11th, 2014).

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<tr>
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<tbody>
<tr>
<td>Beijing</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0%</td>
</tr>
<tr>
<td>Tianjin</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0%</td>
</tr>
<tr>
<td>Hebei</td>
<td>60</td>
<td>11</td>
<td>14</td>
<td>1</td>
<td>5</td>
<td>6</td>
<td>10%</td>
</tr>
<tr>
<td>Henan</td>
<td>370</td>
<td>104</td>
<td>59</td>
<td>18</td>
<td>19</td>
<td>23</td>
<td>61%</td>
</tr>
<tr>
<td>Shandong</td>
<td>100</td>
<td>54</td>
<td>9</td>
<td>9</td>
<td>32</td>
<td>7</td>
<td>22%</td>
</tr>
<tr>
<td>Anhui</td>
<td>6</td>
<td>6</td>
<td>20</td>
<td>0</td>
<td>10</td>
<td>3</td>
<td>5%</td>
</tr>
<tr>
<td>Shanxi</td>
<td>3</td>
<td>0</td>
<td>0</td>
<td>3</td>
<td>4</td>
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<td>1%</td>
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<tr>
<td>Jiangsu</td>
<td>4</td>
<td>3</td>
<td>5</td>
<td>0</td>
<td>3</td>
<td>1</td>
<td>2%</td>
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<tr>
<td>Percentage</td>
<td>56%</td>
<td>18%</td>
<td>11%</td>
<td>3%</td>
<td>8%</td>
<td>4%</td>
<td>100%</td>
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Table 4. The emissions (Gg/day) of gaseous and particulate species from crop field burning on Oct. 6\textsuperscript{th} and Oct. 7\textsuperscript{th} in NCP region, including the provinces of Beijing, Tianjin, Hebei, Henan, Shandong.

<table>
<thead>
<tr>
<th>Time</th>
<th>Province</th>
<th>CO</th>
<th>NOx</th>
<th>NO</th>
<th>NO2</th>
<th>NMOC</th>
<th>SO2</th>
<th>NH3</th>
<th>PM2.5</th>
<th>OC</th>
<th>BC</th>
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<tr>
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<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
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<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>Tianjin</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
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</tr>
<tr>
<td></td>
<td>Hebei</td>
<td>10.58</td>
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<td>0.37</td>
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<td>0.22</td>
<td>0.55</td>
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<td>0.07</td>
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<tr>
<td></td>
<td>Henan</td>
<td>71.17</td>
<td>2.24</td>
<td>1.09</td>
<td>2.50</td>
<td>36.55</td>
<td>0.26</td>
<td>1.47</td>
<td>3.72</td>
<td>2.12</td>
<td>0.44</td>
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<td></td>
<td>Shandong</td>
<td>16.27</td>
<td>0.51</td>
<td>0.25</td>
<td>0.57</td>
<td>8.35</td>
<td>0.06</td>
<td>0.34</td>
<td>0.85</td>
<td>0.48</td>
<td>0.10</td>
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<tr>
<td></td>
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Table 5. Average contribution proportion of crop field burning to PM$_{2.5}$ concentration.

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<tr>
<td>SNCP</td>
<td>34%</td>
<td>17%</td>
<td>6%</td>
<td>3%</td>
<td>1%</td>
<td>1%</td>
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Table 6. The regional average contribution of CFB in mass concentration and percentage, and the lag-time of NNCP to SNCP for the four time-points of T1 (23:00 6th), T2 (05:00 7th), T3 (20:00 7th) and T4 (19:00 8th).

<table>
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<tr>
<th>Time</th>
<th>Mass (µg/m³)</th>
<th>Percentage</th>
<th>Lag-time (hours)</th>
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<td>SNCP</td>
<td>NNCP</td>
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<td>T4</td>
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Figure 1
Figure 2
Figure 3
Figure 4
Figure 5
Figure 6
Figure 7
Figure 8
Figure 9
Figure 10
Figure 11