Chemosphere 260 (2020) 127517

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Impact of rural residential coal combustion on air pollution in Shandong, China

Ying Zhou [a], Teng Zi [a], Jianlei Lang [a, *], Dawei Huang [a], Peng Wei [b], Dongsheng Chen [a], Shuiyuan Cheng [a]

[a] Key Laboratory of Beijing on Regional Air Pollution Control, College of Environmental & Energy Engineering, Beijing University of Technology, Beijing, 100124, China

[b] Chinese Research Academy of Environmental Sciences, Beijing, 100012, China

HIGHLIGHTS

- Rural residential coal combustion for heating had considerable impact on air pollution.
- BC + ACS comprised 60.3%–68.8% of the total RRCC contribution to air pollution.
- Mitigation efficiency of BC + TCS was 5.1 times that of ABC + ACS.
- Obvious regional transport contributions of near-surface RRCC on air pollution were found.

ARTICLE INFO

Article history:
Received 1 March 2020
Received in revised form
5 June 2020
Accepted 19 June 2020
Available online 12 July 2020

Handling Editor: Chennai Guest Editor

Keywords:
Rural residential coal consumption
Emissions
Coal and stove type
Mitigation efficiency
Regional transport

ABSTRACT

Rural residential coal combustion (RRCC) for household heating is a potentially important source of air pollution. However, little research has been done on the environmental impacts of RRCC. This study therefore investigated the impacts of RRCC on air pollution based on detailed household heating data obtained from intensive face-to-face interviews in Shandong province, China. The total contributions and specific contributions of coal, stoves, and coal–stove combinations to air pollution were simulated using the WRF-CAMx-PSAT model. The RRCC for heating had a considerable impact on air pollution, contributing 36.1, 9.1, and 16.1% of atmospheric SO_2, NO_x, and $PM_{2.5}$ in winter, respectively. Different coal–stove combinations had different impacts on air pollution and mitigation efficiencies. The combination of bituminous coal and advanced coal stoves was the dominant contributor to air pollution, comprising 60.3–68.8% of the total RRCC contribution to different air pollutants. Sensitivity analyses indicated that bituminous coal burnt in a traditional stove had the highest mitigation efficiency (0.67 $\mu g \cdot m^{-3}$/10 kt) for atmospheric $PM_{2.5}$ pollution, 4.1 times higher than that of anthracite briquette coal burnt in advanced coal stoves. Moreover, although RRCC is a near-surface emission source, it contributed considerably to regional pollution. Non-local RRCC emissions accounted for 21.8–74.6, 15.5–72.3, and 35.3–79.9% of the total contribution to SO_2, NO_x, and $PM_{2.5}$ in different cities, respectively. The findings of this study improve understanding on the environmental impacts of rural emissions and can provide scientific support for the formulation of effective air pollution mitigation strategies.

1. Introduction

Coal combustion is widely used for household heating in developing countries, particularly in rural regions. The pollutants emitted from rural residential coal combustion (RRCC) have a major impact on air quality, climate, and human health (Lui et al., 2017; Ma et al., 2017; Guo et al., 2018; Huang et al., 2018; Li et al., 2018a). RRCC has been recognised as an important source of air pollution in China, especially in northern China, which has relatively low temperatures in winter (Liu et al., 2017a; Zhang et al., 2017). Compared with coal combustion in other sectors (e.g. industry), the impact of RRCC on air pollution warrants research attention because of the inefficiency of such coal combustion (Liu et al., 2018), varied coal

* Corresponding author. College of Environmental & Energy Engineering, Key Laboratory of Beijing on Regional Air Pollution Control, Beijing University of Technology, Beijing, 100124, China.
E-mail address: jllang@bjut.edu.cn (J. Lang).

https://doi.org/10.1016/j.chemosphere.2020.127517
0045-6535/

and stove types (Li et al., 2016b), low emission height (i.e. air pollutants are more likely to be emitted near ground-level), and lack of pollutant abatement facilities (e.g. desulfurisation and denitrogenation) (Li et al., 2017). Furthermore, the timeframe of RRCC for household heating is often accompanied by unfavourable atmospheric diffusion conditions (e.g. lower temperatures and planetary boundary layer height) (Zhang et al., 2015; Yang et al., 2020). Consequently, investigation of the impact of RRCC, specifically that used for household heating, on air pollution is crucial for improving understanding of the formation of air pollution and can help in the scientific formulation of effective control strategies.

The impacts of residential coal combustion on air pollution have been studied (Liu et al., 2016, 2017a; Xue et al., 2016; Zhang et al., 2017; Li et al., 2018b; Dai et al., 2019; Yan et al., 2020). For example, Dai et al. (2019) indicated that the $PM_{2.5}$ emissions from residential coal combustion were high, accounting for 26 and 32% of ambient $PM_{2.5}$ mass during the heating season in urban and rural sites in Xi'an, respectively. Yan et al. (2020) pointed out that residential coal combustion emissions in China accounted for 25.7% of ambient $PM_{2.5}$ and 7.1% of sulfate on average. These studies illustrated the important role of residential coal combustion in air pollution formation. However, they mainly evaluated the overall contributions to air pollution (i.e. the total impact of residential coal combustion in urban and rural areas). The environmental impacts of RRCC for heating specifically remain unclear. Rural coal combustion characteristics (e.g. decentralised usage in different households) differ from those in urban regions (which mainly have central heating with pollutant abatement facilities), and control strategies in rural areas should therefore be adapted accordingly (Zhang et al., 2008). Determining the influences of RRCC on air pollution will not only contribute to the understanding of environmental impacts of rural emissions but can also provide a scientific reference for the development of effective control policies for rural emission sources.

Different coal and stove types are used for RRCC. Pollutant composition (e.g. sulfur and ash) varies among coal types, such as bituminous coal (BIC) and anthracite briquette coal (ABC). Different stoves, such as traditional coal stoves (TCS) and advanced coal stoves (ACS), have distinct combustion rates and efficiencies (Li et al., 2016a). As a result, the combination of different coals and stoves (e.g. BIC + TCS or ABC + ACS) can cause variations in coal consumption and air pollutant emissions. For example, laboratory research has indicated that the $PM_{2.5}$ emission factor for BIC + ACS was 1.8 times higher than that for ABC + ACS (Li et al., 2016a). Therefore, the environmental impacts vary, and, when control measures are based on the same coal usage, the mitigation efficiency varies substantially among different coal–stove combinations. However, to the best of our knowledge, the specific contribution of coal–stove combinations to air pollution has not been studied. Moreover, regional transport is an important factor in air pollution formation. The air pollutants from RRCC are emitted near ground-level, and it is unclear whether this source contributes to regional air pollution.

Consequently, this study had the following objectives: (1) to determine the extent to which RRCC for heating impacts air pollution, (2) to determine the differences in the environmental impact and air pollution mitigation efficiency for various coal–stove combinations, and (3) to determine whether near-surface RRCC emissions contributed to regional air pollution. A case study was conducted in Shandong, a densely populated province in northern China with large rural areas, where household heating by coal burning and heavy air pollution are common (Shen et al., 2020). Detailed RRCC data were obtained via intensive face-to-face interviews. The total contributions as well as specific contributions of different coals, stoves, and coal–stove combinations to atmospheric $PM_{2.5}$, SO_2, and NO_x were simulated based on the Weather Research and Forecasting (WRF)–Comprehensive Air Quality Model with extensions (CAMx)–Particulate Source Apportionment Technology (PSAT) model. The sensitivity of the air quality impact of by different coals, stoves, and coal–stove combinations was assessed. In addition, the impact on air pollution of both local and non-local RRCC emissions in different cities in Shandong were analysed. The findings of this study can improve understanding of the environmental impacts of rural emissions and provide scientific support for generating effective air pollution mitigation strategies.

2. Materials and methods

2.1. Study area

Shandong is one of the largest agricultural provinces in northern China, covering 156 700 km^2 and containing 17 prefecture-level cities (Fig. S1). It had the third largest GDP in China in 2016, with over 4.1 million residents in rural areas (NBS, 2017). The 20 cities with the worst air quality and those with the worst air quality improvement were identified by the Chinese Ministry of Ecology and Environment (MEE, 2020a) based on the air quality ranking of 168 Chinese cities in 2019. In both rankings, five prefecture-level cities were located in Shandong. The average annual $PM_{2.5}$ concentration in Shandong cities ranged from 39.58 to 59.75 $\mu g/m^3$ in 2019 (MEE, 2020b). These values were far above the air quality standard recommended by the World Health Organization (10 $\mu g/m^3$) and the secondary air quality standard of China (35 $\mu g/m^3$). Shandong is a densely populated region with widespread household coal heating, and thus the air quality needs to be urgently improved (Shen et al., 2020).

2.2. Emission estimation

A bottom-up approach was used to develop an RRCC emission inventory for household heating based on coal- and stove-specific coal consumption and localised emission factors (EFs) to distinguish the combustion state (i.e. flaming or smoldering). The emissions were estimated using Eq. (1):

$$E_i = \sum_j \sum_k \sum_s (EF_{i,k,s} \times CC_{j,k,s} \times 10^{-3}) \tag{1}$$

where E_i is the RRCC total emissions (t) for pollutant i, including SO_2, NO_x, CO, volatile organic compounds (VOC), $PM_{2.5}$, elemental carbon (EC), organic carbon (OC), and NH_3; $CC_{j,k,s}$ is the coal consumption (t) of county j in Shandong for coal–stove combination k under combustion state s, i.e. flaming or smoldering states; and $EF_{i,k,s}$ is the emission factor (g/kg) of pollutant i for coal–stove combination k under combustion state s.

The coal types were BIC and ABC, and the stoves types were TCS, ACS, and home Arcola (HAC). Six coal–stove combinations were thus used in this study: BIC + TCS, BIC + ACS, BIC + HAC, ABC + TCS, ABC + ACS, and ABC + HAC.

2.2.1. Emission factors

The EFs of RRCC were dependent on coal and stove characteristics as well as the combustion state, which were selected based on the systematic evaluation of localised measurements. The detailed screening process and EFs used in this study are described in Text S1 and Table S1.

2.2.2. Coal consumption investigation and calculation

This study avoided using uniform per capita fuel consumption for total coal consumption estimation without distinguishing coal and stove types. Instead, regional differences in natural and social economic conditions were considered. Shandong was consequently divided into four clusters, namely southwestern Shandong (SD-1), southern Shandong (SD-2), peninsula area (SD-3), and northern Shandong (SD-4). Furthermore, 6–10 typical counties in each cluster were chosen as the objects of the investigation (Fig. S2) based on a multi-stage stratified randomised sampling method (Text S2). Excluding questionnaires with missing data and information errors, a total of 3716 valid households across 105 villages were surveyed. Detailed data (e.g. coal consumption data; coal, stove, and heating types (radiator (RA) and *kang* (KA)); heating days; daily heating hours; combustion states; and start and end heating dates) were collected through face-to-face investigations. Based on the detailed investigation data, RRCC fuel consumption distinguished by coal and stove type as well as combustion state were calculated. A detailed description of this process is given in Text S3.

2.3. Model simulation design

2.3.1. Model configuration

An integrated WRF–CAMx modelling system coupled with the PSAT module was used to investigate the contributions of RRCC to air quality in the 2016–2017 heating season. A two-level nested-grid was established for the modelling system (Fig. S1). Domain 1 covered most of eastern China, with a spatial resolution of 27 × 27 km, and Domain 2 covered Shandong, with a spatial resolution of 9 × 9 km. The simulation of Domain 1 provided time-varying boundary conditions for Domain 2, and the simulation period was January 2017. The emission data used in this model included (1) the RRCC emission inventory established in this study and (2) the emission inventory of other anthropogenic sources in 2016 from the Multiresolution Emission Inventory of China (http://www.meicmodel.org).

The meteorological fields required by CAMx were simulated by the WRF model (version 3.5). The meteorological initial and lateral boundary conditions were derived from the National Center for Environmental Prediction Final Operational Global Analysis data (http://rda.ucar.edu/datasets/ds083.2) with a spatial resolution of 1 × 1° and temporal resolution of 6 h. The Purdue Lin, Yonsei University, and Noah and Grell–Devenyi schemes were chosen as the microphysics, planetary boundary layer, and land-surface and cumulus schemes, respectively. The radiation schemes used were Goddard shortwave radiation and rapid radiative transfer model longwave radiation. For the CAMx configuration, the vertical resolution was configured as 29 layers, which was consistent with the WRF model. For the scheme setting, Euler Backward Iterative, Piecewise Parabolic Method, K-theory, Regional Acid Deposition Mechanism, Carbon Bond Mechanism, Kain–Fritsch, and Mellor–Yamada–Janjic schemes were applied to the chemical solver, horizontal advection solver, vertical diffusion, aqueous-phase chemistry, gas-phase chemistry, cumulus parameterisation, and boundary layer parameterisation schemes, respectively.

2.3.2. Source and receptor setup in PSAT

The PSAT module (version 6.4) was used to investigate the individual concentration contribution of the concerned emission source and as a source apportionment tool to estimate the contributions from a specific emission sector/type or region. In this study, the emissions of two coal types (BIC and ABC), three stove types (TCS, HAC, and ACS), and six coal–stove combinations (BIC + TCS, BIC + ACS, BIC + HAC, ABC + TCS, ABC + ACS, and ABC + HAC) were separately tracked in PSAT to identify the impact of various sources on air pollution and to analyse their impact sensitivity to air pollution. The RRCC emissions from 18 regions were tracked to identify the impact on air pollution of both local and non-local RRCC emissions from different cities. The 18 regions were Jinan, Dezhou, Liaocheng, Tai'an, Heze, Jining, Zaozhuang, Zibo, Binzhou, Laiwu, Dongying, Weifang, Yantai, Weihai, Qingdao, Rizhao, Linyi, and surrounding areas in Domain 2 (Fig. S1). The national air quality monitoring stations in each city were used as receptors.

2.3.3. Model evaluation

The simulated concentrations of each pollutant (i.e. $PM_{2.5}$, SO_2, and NO_x) were compared with observational data for various cities in Shandong (Table S2). The value of correlation coefficients (R) ranged from 0.32 to 0.79, and the normalised mean bias (NMB) of $PM_{2.5}$, NO_2, and SO_2 ranged from −31.6 to 23.2, −2.0 to 1.7, and −2.5 to −0.5%, respectively. The normalised mean errors (NME) of $PM_{2.5}$, NO_2, and SO_2 were 30.5–49.8, 22.0–54.8, and 28.9–56.6%, respectively. Considering the uncertainty of the emission inventory and unavoidable deficiencies of meteorological and air quality models, the model performance was considered acceptable (Zhang et al., 2006; Kwok et al., 2010; Lv et al., 2018).

2.4. Assessment of sensitivity of impacts of different coals, stoves, and coal–stove combinations on air quality

Based on the concentration contribution of different coals, stoves, and coal–stove combinations, we calculated the concentration contribution per unit coal consumption for the RRCC types mentioned above (section 2.2) and analysed the sensitivity of the air quality impact. We defined sensitivity coefficients (SCs) as the average pollutant concentration contribution (e.g. SO_2, NO_x, and $PM_{2.5}$) per unit coal consumption. These values can provide a reference for identifying detailed RRCC emission types (e.g. coal type, stove type, and coal–stove combination) with the highest pollutant concentration contribution under equal coal consumption. The identified RRCC types had larger mitigation effects on air pollution, and their control should be prioritised. The algorithm was as follows:

$$SC_{i,j} = \frac{C_{i,j}}{CC_i} \tag{2}$$

where SC ($\mu g \cdot m^{-3}$/10 kt) represents the sensitivity coefficient of RRCC emission type i to air quality; i represents the RRCC emission type (e.g. BIC or ABC and TCS, ACS, or HAC); j represents the pollutant, which denotes air quality (e.g. SO_2, NO_x, and $PM_{2.5}$); $C_{i,j}$ ($\mu g/m^3$) represents the average concentration contribution for all receptors of emission type i to pollutant j; and CC_i (10 kt) represents the coal consumption amount of RRCC emission type i.

3. Results and discussion

3.1. Rural coal consumption in Shandong

According to the estimation based on large-scale, face-to-face investigations, the total coal consumption of RRCC for household heating during 2016–2017 in Shandong was 19 274 kt. This was approximately 1.9 times higher than the values reported in the China Energy Statistical Yearbook (CESY, 2017). This indicated a considerable underestimation of rural coal consumption in the CESY, which has also been pointed out by other studies. The estimated coal consumption of RRCC in Shandong in 2010, according to Peng et al. (2019), was 2.3 times higher than that reported in the CESY. Such an underestimation was also found in other regions, e.g. Beijing–Tianjin–Hebei (Cheng et al., 2017). Greater differences were found in the consumption of the coal types (Fig. S3). Coal consumption under the flaming state contributed 84.8% of the total coal consumption, which was 4.6 times higher than that of coal consumption under the smoldering state (Fig. S3).

The coal consumption of RRCC for different coal types, stove types, and coal–stove combinations in different prefecture-level cities is shown in Fig. 1. BIC accounted for 44.9–87.9% of total coal consumption in various cities and was the major coal type used for heating (Fig. 1(a)). This may be because rural residents preferred to choose affordable, accessible, and high-calorific-value fossil fuels for heating. Thus, BIC, with these advantages, was preferred by most residents. The ACS stove type, characterised by higher thermal efficiency and larger heating area, accounted for 76.7% of total consumption in various cities. The TCS stove type contributed to 14.7–26.7% of total consumption and was used for raw coal consumption in Zaozhuang, Laiwu, and Tai'an especially.

In terms of the total coal consumption of RRCC in the studied cities, Weifang had the highest coal consumption, accounting for 13.6% of the province total. Yantai (10.1%), Heze (9.7%), and Qingdao (8.6%) also had high coal consumption, mainly because of the larger rural population and heating demands in these regions. BIC + ACS + RA (as described in Text S3) was the main heating combination (i.e. the combination of coal, stove, and heating types; Fig. 1(b)), accounting for 51.4% of all heating combination types in Shandong, especially in Heze (68.3%), Jining (64.0%) and Liaocheng (61.5%). ABC + ACS + RA was another important heating combination (10.2%). We identified 16 combinations that distinguished coal, stove, and heating types during the face-to-face investigation. In addition to the four combinations shown in Fig. 1(b), the

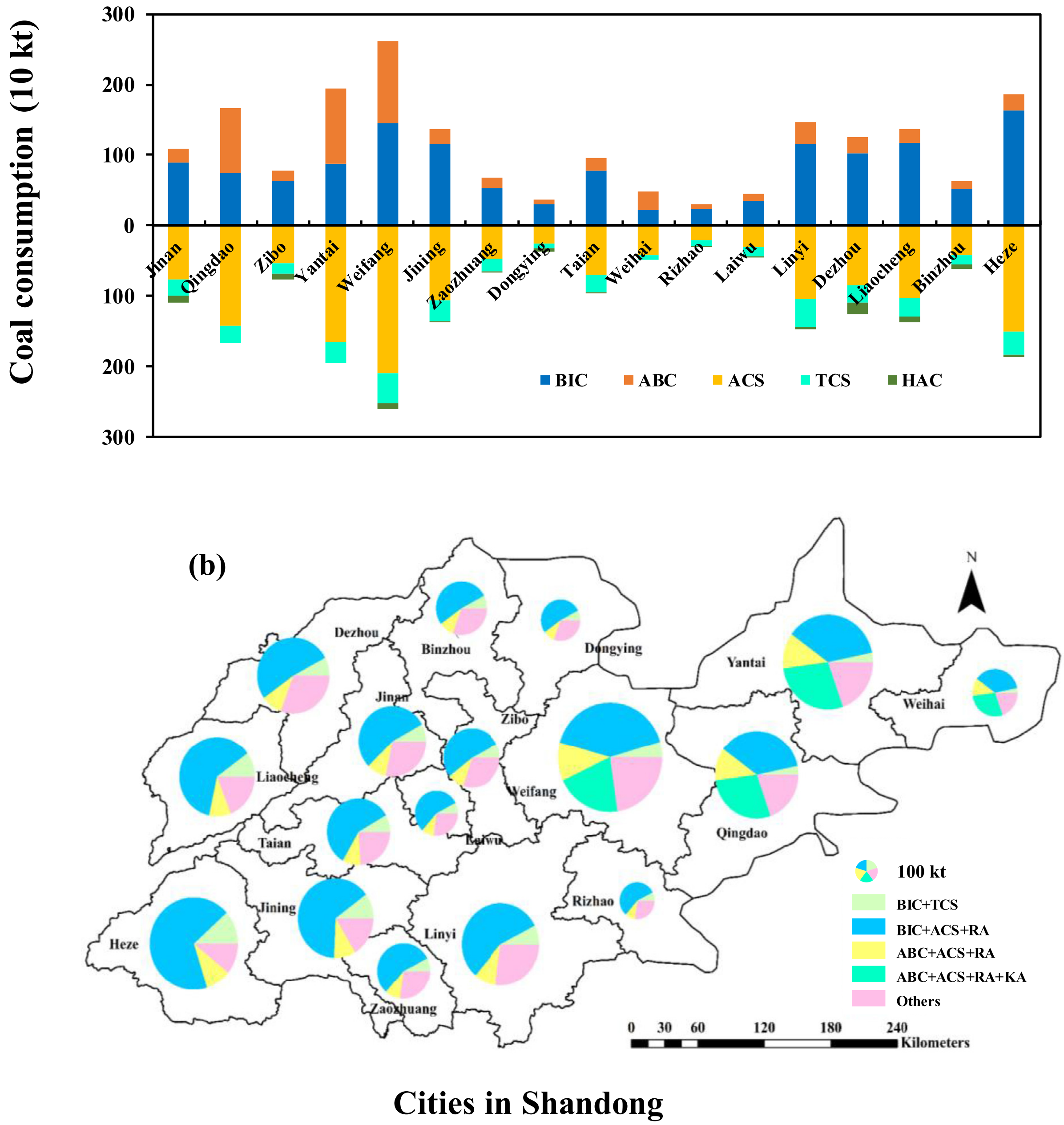

Fig. 1. (a) Rural household coal consumption distinguished by coal and stove type at city level in Shandong province, China. (b) Coal consumption of major heating combinations at city level in Shandong; ABC: anthracite briquette coal; ACS: advanced coal stoves; BIC: bituminous coal; HAC: home Arcola; KA: *kang*; RA: radiator; TCS: traditional coal stoves.

remaining 12 combinations were classified as 'others' because of their lower proportions (0.2–3.9%).

3.2. Air pollutant emissions

The total emissions of SO_2, NO_x, CO, VOC, $PM_{2.5}$, EC, OC, and NH_3 from RRCC for household heating during 2016–2017 in Shandong were 260.1, 33.8, 3 333.1, 82.2, 112.6, 12.6, 26.2, and 5.9 kt, respectively. As shown in Fig. 2, coal consumed in the flaming state emitted more SO_2, NO_x, CO, $PM_{2.5}$, EC, OC, and NH_3 than that consumed in the smoldering state, accounting for 92.5, 92.5, 56.6, 92.5, 99.8, 98.5, and 84.9% of total emissions, respectively. The RRCC emissions under the smoldering state contributed 43.4 and 66.6% of the total CO and VOC emissions, respectively, although the coal consumption under this state accounted only for 15.2% of total coal consumption. The CO and VOC per unit of coal burning under the smoldering state were 3.3 and 10.2 times higher than those under flaming state, respectively. The more complete combustion under the flaming state, with sufficient air supply and high temperature (Du et al., 2016; Liu et al., 2017b), facilitated the formation of SO_2, NO_x, and PM (Du et al., 2016; Liu et al., 2018). Under the smoldering state, stoves have low thermal efficiencies with incomplete coal combustion and lower smoke gas temperatures, which could have promoted the formation of CO and VOC (Du et al., 2016; Liu et al., 2017b, 2018).

The emissions from different coal types and coal–stove combinations are shown in Fig. 2(a) and (b), respectively. BIC combustion contributed 73.4–97.7% of the studied emissions. If BIC were completely replaced by ABC, the emissions of SO_2, NO_x, CO, VOC, $PM_{2.5}$, EC, OC, and NH_3 would be reduced by 66.5, 8.8, 13.3, 41.2, 73.2, 92.1, 77.2, and 32.5%, respectively. The BIC + ACS coal–stove combination was the main contributor (40.5–69.5%) to most of pollutant emissions, except for OC and NH_3, for which the contributions were 35.9 and 17.4%, followed by the BIC + TCS combination (9.7–51.9%). Given that BIC + TCS emitted more pollutants per unit of coal burnt, the government should prioritise replacing this coal–stove combination. Therefore, the use of cleaner coal (i.e. ABC) with ACS stoves could effectively reduce pollutant emissions.

We further analysed the total RRCC emissions and contributions of different coal–stove combinations in different prefecture-level cities (Fig. S4). An uneven distribution of RRCC emissions was found across the cities. High-emission regions were located in Weifang, Heze, and Linyi and accounted for 11.0–13.6, 7.7–10.0, and 7.4–9.0% of the total air pollutant emissions, respectively. In contrast, Laiwu had the lowest emissions, accounting for only 1.2–1.4% of the total emissions in Shandong. Regarding the contribution of different coal–stove combinations to the pollution in different cities, BIC + ACS (54.5–69.2%) contributed the most to pollution emissions across all studied cities (except VOC, OC, and NH_3). BIC + TCS contributed 11.0–67.8% to various pollutants in Heze, Linyi, Zaozhuang, and Jining. ABC + ACS accounted for the second largest proportion of emissions in Qingdao, Yantai, and Weihai.

3.3. Impact of RRCC for household heating on air pollution

3.3.1. Impacts of emissions from coals, stoves, and coal–stove combinations on air pollution

During January 2017, RRCC emissions contributed 19.8, 3.1, and 11.9 $\mu g/m^3$ to the SO_2, NO_x, and $PM_{2.5}$ concentrations in Shandong, respectively, representing 36.1, 9.1, and 16.1% of the monthly average concentration of these pollutants, respectively (Fig. 3). Taking $PM_{2.5}$ as an example, the concentration contribution of RRCC emissions in the heating period exceeded that of vehicle emissions (15.2% (Jiang, 2018)). This indicated that RRCC for heating considerably impacted air pollution. Variations in the contributions of different pollutants to the total pollutant concentration were also determined. The contribution of SO_2 was 4.0 times that of NO_x, mainly because of the relatively low combustion temperature and low efficiency of rural coal, which are not conductive to the formation of NO_x (Wen and Song, 2004). In addition, rural residents tend to use low-quality, low-price raw coal. The ash and sulfur content of low-quality raw coal is high, and coal burning for household usage does not entail dust removal or desulfurisation.

Furthermore, the contribution of RRCC from various coal types, stove types, and coal–stove combinations to overall air pollution varied substantially. The contribution proportions of BIC (7.1–32.5%) were far above those of ABC (1.6–3.6%). ACS was the

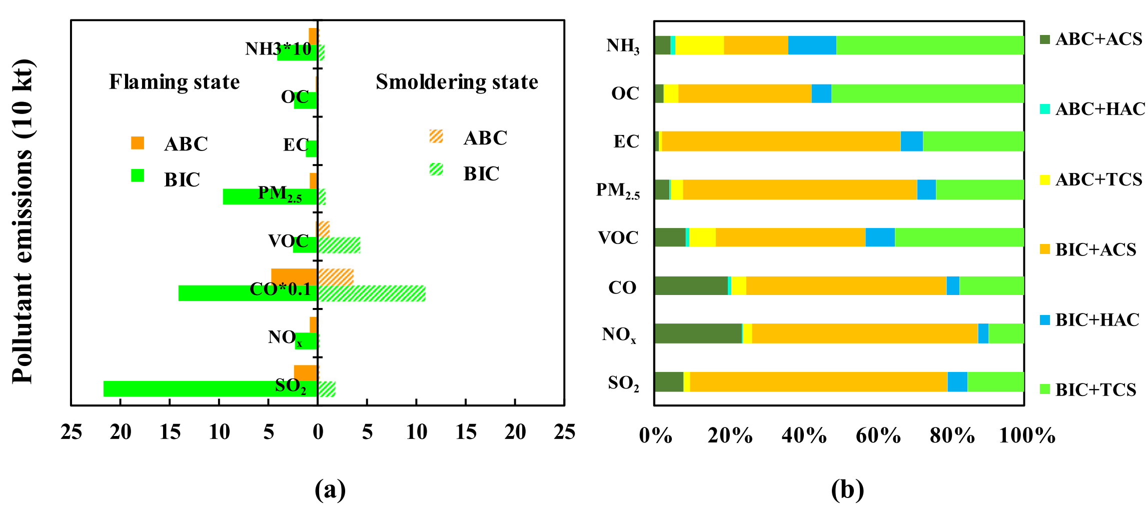

Fig. 2. (a) Air pollutant emissions from rural coal combustion in Shandong province, China, for 2016–2017 heating season. (b) Contributions of different coal–stove combinations to various pollutants; ABC: anthracite briquette coal; ACS: advanced coal stoves; BIC: bituminous coal; EC: elemental carbon; HAC: home Arcola; OC: organic carbon; TCS: traditional coal stoves; VOC: volatile organic compounds.

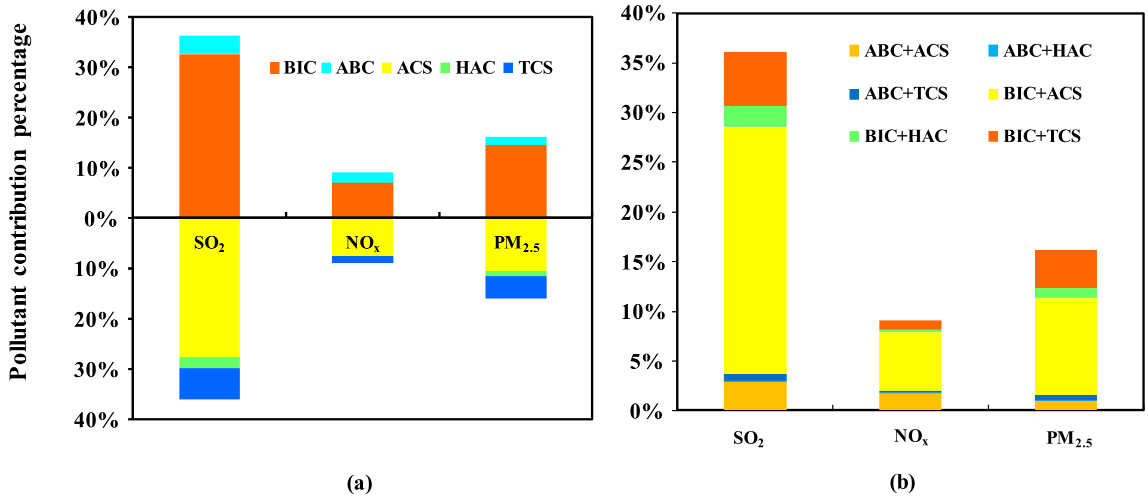

Fig. 3. Contributions of rural residential coal combustion (RRCC) emissions to SO_2, NO_x, and $PM_{2.5}$ concentrations for (a) different coal and stove types, and (b) coal–stove combinations; ABC: anthracite briquette coal; ACS: advanced coal stoves; BIC: bituminous coal; HAC: home Arcola; TCS: traditional coal stoves.

main contributor among the stove types, accounting for 27.8, 7.5 and 10.6% of SO_2, NO_x, and $PM_{2.5}$ emissions, respectively; the emissions from TCS nonetheless made a considerable contribution, especially for SO_2 (6.2%) and $PM_{2.5}$ (4.4%). Although ACS stoves burn more cleanly than TCS stoves do (Krumal et al., 2019), the higher emissions from ACS than TCS in this study were attributed to the greater use of the former in Shandong (see section 3.1). The BIC + ACS combination contributed to more than half of the total RRCC emissions, accounting for 68.8, 64.8, and 60.3% of SO_2, NO_x, and $PM_{2.5}$ emissions, respectively. Consequently, BIC + ACS was identified as the primary coal–stove combination in terms of air pollution. BIC + TCS also contributed to a considerable proportion of the total emissions, especially SO_2 (15.3%) and $PM_{2.5}$ (23.6%) emissions. Given their considerable impact on air pollution, more attention should be paid to the RRCC emissions and coal–stove combinations mentioned above, and corresponding control measures should be formulated.

3.3.2. Impact of coals, stoves, and coal–stove combinations on air pollution in different cities

In addition to analysing the contributions of coal types, stove types, and coal–stove combinations to pollutant concentrations in Shandong, we investigated the contributions to pollutant concentrations in various cities. As shown in Fig S5, the impact of RRCC emissions from Shandong on air pollution varied substantially among the various cities. For $PM_{2.5}$, the RRCC emissions from Shandong contributed the most to $PM_{2.5}$ concentrations in Weifang (15.7 μg/m^3; 22.0% of the total contribution to air pollution). The $PM_{2.5}$ concentrations in Linyi (19.6%), Rizhao (18.9%), and Yantai (18.2%) were also affected by the RRCC emissions from Shandong. The range of contribution concentrations and contribution proportions for various cities were from 5.4 (Weihai) to 16.7 (Liaocheng) μg/m^3 and 11.3% (Dongying) to 22.0% (Weifang), respectively. For SO_2, the concentration contributions were relatively high in Heze (27.3 μg/m^3), Liaocheng (35.8 μg/m^3), Linyi (27.3 μg/m^3), and Zibo (35.2 μg/m^3). Relatively high contribution proportions were found in Heze (53.3%), Liaocheng (54.0%), Zibo (49.3%), and Rizhao (50.5%). The range of contribution concentration and contribution proportions for various cities were from 9.2 (Weihai) to 35.8 (Liaocheng) μg/m^3 and 22.6% (Jining) to 54.0% (Liaocheng), respectively. The range of contribution concentration and contribution proportions for various cities were from 1.5 (Weihai) to 5.7 (Liaocheng) μg/m^3 and 4.8% (Jining) to 15.0% (Rizhao), respectively. Overall, the impact of RRCC on air pollution was relatively strong for Zibo, Liaocheng, and Heze, and the air quality of these cities was relatively poor and subject to severe winter haze events (MEE, 2020b). Therefore, control measures for rural coal combustion may improve the air quality of these cities.

3.4. Sensitivity analyses of impacts of coals, stoves, and coal–stove combinations on air pollution

According to the estimation of the concentration contribution per unit coal consumption (section 2.4), the SCs for the different coal types, stove types, and coal–stove combinations are shown in Fig. 4. The sensitivity of the impact on air pollution of different coal types, stove types, and coal–stove combinations varied considerably. The SC of BIC was generally greater than that of ABC, e.g. 2.1 times for SO_2 and 3.2 times for $PM_{2.5}$, and that of HAC stoves was greater than those of the other stove types for SO_2 (1.2–1.6 times) and $PM_{2.5}$ (1.2–1.7 times). However, for NO_x, the SCs of ABC and ACS were slightly higher than those of the other coal or stove types. The higher calorific value of ABC and higher thermal efficiency of ACS may have caused increased NO_x emissions (Li et al., 2016a). Hence, the government should not ignore NO_x emissions when promoting ABC and ACS. The SC of BIC + HAC (0.89 μg·m^{-3}/10 kt) was 4.0 times higher than that of ABC + HAC for SO_2, that of BIC + TCS (0.67 μg·m^{-3}/10 kt) was 4.1 times higher than that of ABC + ACS for $PM_{2.5}$, and that of ABC + ACS was 3.3 times higher than that of ABC + TCS for NO_x.

The differences in the sensitivity of the impact on air pollution of the various coal and stove types and coal–stove combinations reflected the differences in air pollution mitigation efficiencies. For example, for $PM_{2.5}$, a well-studied pollutant, reducing the use of the BIC + TCS combination under an equal reduction of RRCC emissions should be prioritised. Research on the sensitivity of the air pollution impact is essential for identifying RRCC emission types with high mitigation efficiencies. Effective management measures can consequently be developed to ensure greater environmental and economic benefits.

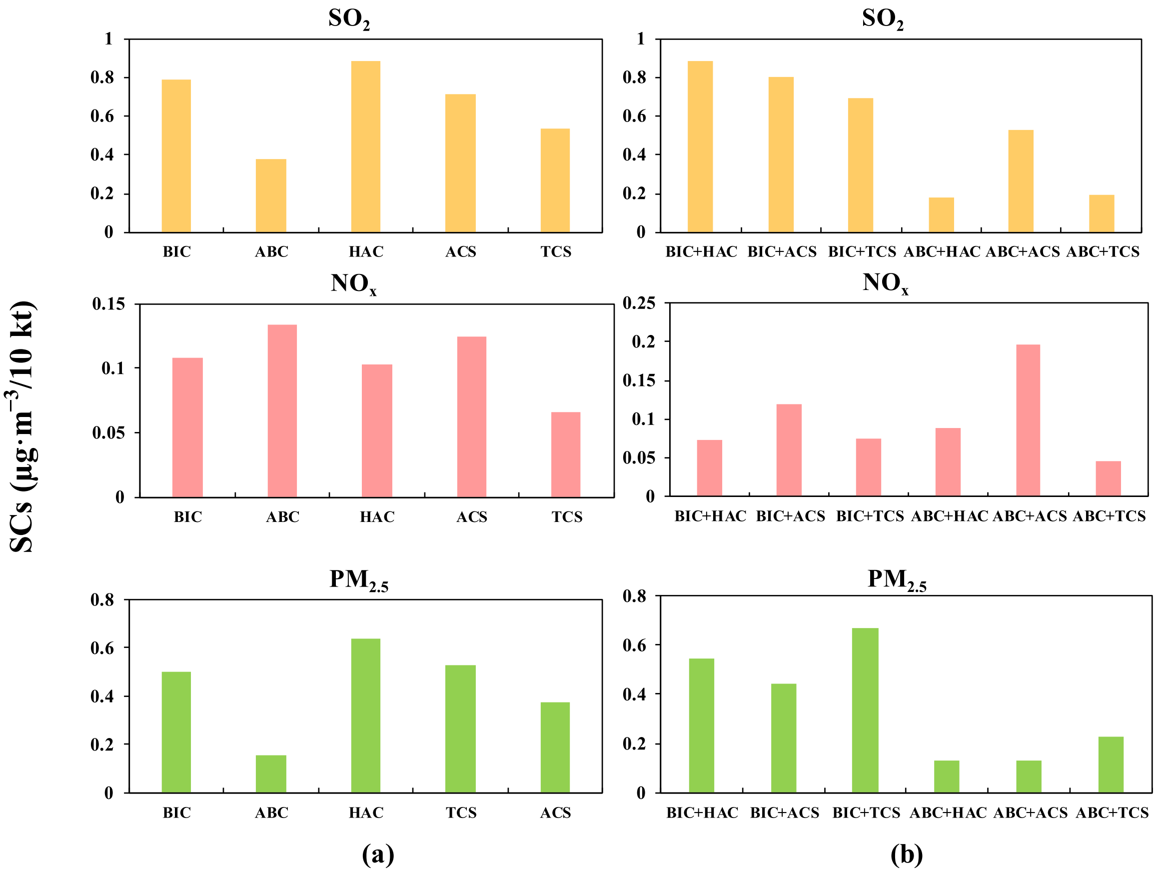

Fig. 4. Sensitivity coefficients (SCs) for different (a) coal and stove types and (b) coal–stove combinations; ABC: anthracite briquette coal; ACS: advanced coal stoves; BIC: bituminous coal; HAC: home Arcola; TCS: traditional coal stoves.

3.5. Impact on air pollution of local and non-local RRCC emissions in different cities

The regional transportation contributions of RRCC in the cities of Shandong to $PM_{2.5}$, SO_2, and NO_x emissions were analysed. As shown in Fig. 5, the regional transport contributions to different pollutants differed between the cities. The regional contribution of non-local RRCC emissions to $PM_{2.5}$, SO_2, and NO_x in the studied cities were 35.3–79.9, 21.8–74.6, and 15.5–72.3%, respectively. In addition, regional contributions to the pollutants varied considerably between the studied cities. Jinan, Binzhou, and Rizhao were greatly affected by regional transport, and the average contributions to $PM_{2.5}$, SO_2, and NO_x in these cities were 72.6, 62.2, and 59.2%, respectively. Taking the city with the least local contribution, Jinan, as an example, the $PM_{2.5}$ concentration contribution from local RRCC emissions was 1.63 $\mu g/m^3$, accounting for 20.1% of the total RRCC contribution (taking the total RRCC concentration contribution as 100%). In contrast, the $PM_{2.5}$ concentration contribution from Dezhou and Tai'an were 2.46 and 0.93 $\mu g/m^3$, respectively, accounting for 30.4 and 11.5%, respectively. For SO_2, regional transport contributed to 74.6%, which was mainly contributed by Dezhou (40.5%) and Tai'an (10.6%). For NO_x, regional transport contributed to 72.3%, and the highest non-local contributors were Dezhou (43.9%) and Tai'an (10.4%), but regional transport contributed 30.7% on average in other cities. In addition to emission sources with emission heights at high altitudes, those with emission heights at low altitudes can also substantially contribute to regional transport. This shows that controlling emission sources with emission heights at low altitudes should not be ignored in the coordinated control of regional pollution.

4. Conclusions

In this study, the impact of RRCC on air quality, with a specific focus on household heating, was carried out based on the specific coal consumption of coal–stove combinations and emissions obtained from a face-to-face survey conducted in Shandong, China. The total and specific contributions of coal and stove type as well as coal–stove combinations to air pollution were identified using the WRF–CAMx–PSAT model. The results showed that RRCC for household heating considerably impacted air pollution in winter, especially SO_2 (36.1%) and $PM_{2.5}$ (16.1%) emissions. Moreover, the contributions of RRCC from various coal types, stove types, and coal–stove combinations to the overall air pollution varied. The concentration contribution of BIC + ACS represented more than half of the total RRCC contribution. Given these considerable impacts on air pollution, RRCC emissions and the coal–stove combinations should be considered and the control measures strengthened.

We further estimated the contribution per unit coal consumption for the RRCC types and found variations in the sensitivity of their impacts on air pollution. The air pollution mitigation efficiencies of various coal–stove combinations under equal coal consumption reduction therefore differ. The mitigation efficiency for atmospheric $PM_{2.5}$ of BIC was 2.2 times higher than that of ABC. If BIC were completely replaced by ABC, $PM_{2.5}$ emissions would be

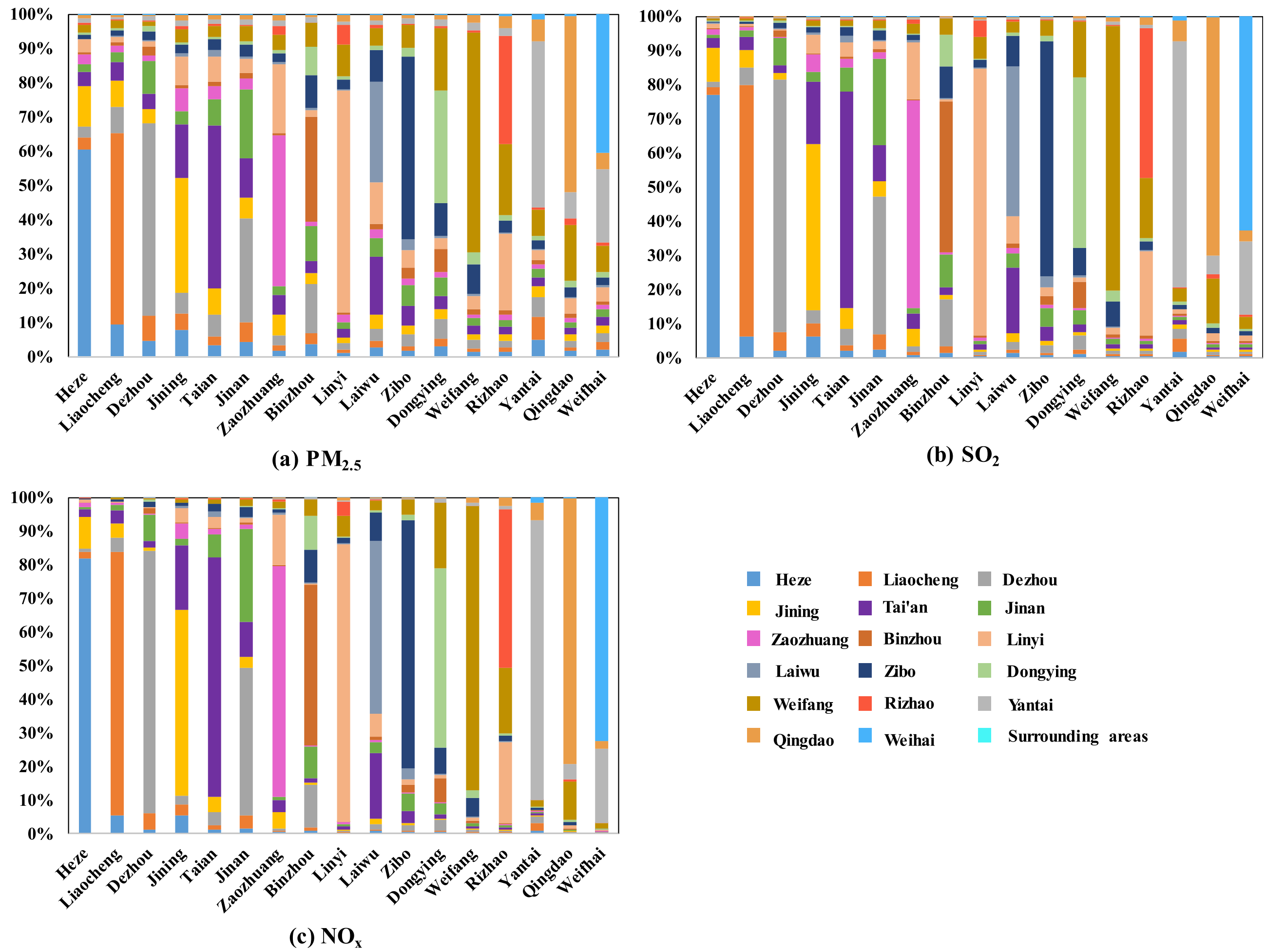

Fig. 5. Regional contribution rates of rural coal combustion emissions of (a) $PM_{2.5}$, (b) SO_2, and (c) NO_x from Shandong province, China, in January 2017.

reduced by 73.2%. The BIC + TCS combination had the highest mitigation efficiency for atmospheric $PM_{2.5}$ pollution, namely 4.1 times higher than that of ABC + ACS. The contributions of RRCC to regional pollution were also analysed. The regional contributions of non-local RRCC emissions to $PM_{2.5}$, SO_2, and NO_x emissions in the studied cities were 35.3–79.9, 21.8–74.6, and 15.5–72.3%, respectively. RRCC is a typical near-surface emission source with low emission height, and RRCC emissions were found to contribute considerably to regional transport.

The results of this study contribute to the understanding of the environmental impacts of rural emissions and provide scientific support for generating robust and effective air pollution mitigation strategies. The investigation of the specific contributions of coal types, stove types, and coal–stove combinations to air pollution was necessary. In addition to the total and abovementioned specific contributions to air pollution, the contribution per unit coal consumption should also be considered in the development of management measures to ensure more environmental and economic benefits. The BIC + TCS combination should be prioritised to reduce RRCC emissions and enhance the efficacy of $PM_{2.5}$ pollution control. Moreover, emission sources with emission heights at low altitudes should be managed for the coordinated control of regional pollution. The mechanism of the contributions of near-surface emission sources to regional air pollution should be investigated in future research.

Credit author statement

Ying Zhou: Conceptualization, Methodology, Writing- Reviewing and Editing. Teng Zi: Writing- Original draft preparation. Jianlei Lang: Supervision, Writing- Reviewing and Editing. Dawei Huang: Investigation, Software; Peng Wei: Software, Validation. Dongsheng Chen: Software, Validation. Shuiyuan Cheng: Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This research was supported by the Natural Sciences Foundation of China (grant no. 51878012) and National Key R&D Program of China (grant no. 2018YFC0213206 & 2017YFC0212202). We gratefully acknowledge the Beijing Municipal Commission of Education and Beijing Municipal Commission of Science and Technology for supporting this work. The authors wish to thank the anonymous reviewers for their insightful comments.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2020.127517.

References

CESY, 2017. China Energy Statistical Yearbook. China Statistics Press, Beijing, China.

Cheng, M., Zhi, G., Tang, W., Liu, S., Dang, H., Guo, Z., Du, J., Du, X., Zhang, W., Zhang, Y., Meng, F., 2017. Air pollutant emission from the underestimated households' coal consumption source in China. Sci. Total Environ. 580, 641–650.

Dai, Q., Bi, X., Song, W., Li, T., Liu, B., Ding, J., Xu, J., Song, C., Yang, N., Schulze, B.C., Zhang, Y., Feng, Y., Hopke, P.K., 2019. Residential coal combustion as a source of primary sulfate in Xi'an, China. Atmos. Environ. 196, 66–76.

Du, Q., Zhang, C., Mu, Y., Cheng, Y., Zhang, Y., Liu, C., Song, M., Tian, D., Liu, P., Liu, J., Xue, C., Ye, C., 2016. An important missing source of atmospheric carbonyl sulfide: domestic coal combustion. Geophys. Res. Lett. 43, 8720–8727.

Guo, H., Kota, S.H., Chen, K.y., Sahu, S.K., Hu, J.l., Ying, Q., Wang, Y., 2018. Source contributions and potential reductions to health effects of particulate matter in India. Atmos. Chem. Phys. 18, 15219–15229.

Huang, R.J., Cheng, R., Jing, M., Yang, L., Li, Y., Chen, Q., Chen, Y., Yan, J., Lin, C., Wu, Y., Zhang, R., El Haddad, I., Prevot, A.S.H., O'Dowd, C.D., Cao, J., 2018. Source-specific health risk Analysis on particulate trace elements: coal combustion and traffic emission as major contributors in wintertime beijing. Environ. Sci. Technol. 52, 10967–10974.

Jiang, X., 2018. Characteristics and Source Analysis of Airborne Particulate Pollution in Laiwu City. Environmental Science and Engineering. Shandong Jianzhu University [D], in Chinese.

Krumal, K., Mikuska, P., Horak, J., Hopan, F., Krpec, K., 2019. Comparison of emissions of gaseous and particulate pollutants from the combustion of biomass and coal in modern and old-type boilers used for residential heating in the Czech Republic, Central Europe. Chemosphere 229, 51–59.

Kwok, R.H.F., Fung, J.C.H., Lau, A.K.H., Fu, J.S., 2010. Numerical study on seasonal variations of gaseous pollutants and particulate matters in Hong Kong and Pearl River Delta Region. J. Geophys. Res. 115.

Li, Q., Jiang, J., Qi, J., Deng, J., Yang, D., Wu, J., Duan, L., Hao, J., 2016a. Improving the Energy efficiency of stoves to reduce pollutant emissions from household solid fuel combustion in China. Environ. Sci. Technol. Lett. 3, 369–374.

Li, Q., Li, X., Jiang, J., Duan, L., Ge, S., Zhang, Q., Deng, J., Wang, S., Hao, J., 2016b. Semi-coke briquettes: towards reducing emissions of primary PM2.5, particulate carbon, and carbon monoxide from household coal combustion in China. Sci. Rep. 6, 19306.

Li, Q., Jiang, J., Wang, S., Rumchev, K., Mead-Hunter, R., Morawska, L., Hao, J., 2017. Impacts of household coal and biomass combustion on indoor and ambient air quality in China: current status and implication. Sci. Total Environ. 576, 347–361.

Li, H., You, S., Zhang, H., Zheng, W., Zou, L., 2018a. Investigating the environmental quality deterioration and human health hazard caused by heating emissions. Sci. Total Environ. 628–629, 1209–1222.

Li, X., Wu, J., Elser, M., Feng, T., Cao, J., El-Haddad, I., Huang, R., Tie, X., Prévôt, A.S.H., Li, G., 2018b. Contributions of residential coal combustion to the air quality in Beijing–Tianjin–Hebei (BTH), China: a case study. Atmos. Chem. Phys. 18, 10675–10691.

Liu, J., Mauzerall, D.L., Chen, Q., Zhang, Q., Song, Y., Peng, W., Klimont, Z., Qiu, X., Zhang, S., Hu, M., Lin, W., Smith, K.R., Zhu, T., 2016. Air pollutant emissions from Chinese households: a major and underappreciated ambient pollution source. Proc. Natl. Acad. Sci. Unit. States Am. 113, 7756–7761.

Liu, C., Zhang, C., Mu, Y., Liu, J., Zhang, Y., 2017b. Emission of volatile organic compounds from domestic coal stove with the actual alternation of flaming and smoldering combustion processes. Environ. Pollut. 221, 385–391.

Liu, P., Zhang, C., Xue, C., Mu, Y., Liu, J., Zhang, Y., Tian, D., Ye, C., Zhang, H., Guan, J., 2017a. The contribution of residential coal combustion to atmospheric PM2. 5 in northern China during winter. Atmos. Chem. Phys. 17, 11503–11520.

Liu, Y., Zhang, Y., Li, C., Bai, Y., Zhang, D., Xue, C., Liu, G., 2018. Air pollutant emissions and mitigation potential through the adoption of semi-coke coals and improved heating stoves: field evaluation of a pilot intervention program in rural China. Environ. Pollut. 240, 661–669.

Lui, K.H., Bandowe, B.A., Tian, L., Chan, C.S., Cao, J.J., Ning, Z., Lee, S.C., Ho, K.F., 2017. Cancer risk from polycyclic aromatic compounds in fine particulate matter generated from household coal combustion in Xuanwei, China. Chemosphere 169, 660–668.

Lv, Z., Liu, H., Ying, Q., Fu, M., Meng, Z., Wang, Y., Wei, W., Gong, H., He, K., 2018. Impacts of shipping emissions on PM2.5 pollution in China. Atmos. Chem. Phys. 18, 15811–15824.

Ma, Q., Cai, S., Wang, S., Zhao, B., Martin, R.V., Brauer, M., Cohen, A., Jiang, J., Zhou, W., Hao, J., Frostad, J., Forouzanfar, M.H., Burnett, R.T., 2017. Impacts of coal burning on ambient PM2.5 pollution in China. Atmos. Chem. Phys. 17, 4477–4491.

MEE, 2020a. Ministry of Ecology and Environment of the People's Republic of China): the Quality of Surface Water and Ambient Air of China. Ministry of Ecology and Environment of the People's Republic of China available at: http://www.mee.gov.cn/xxgk2018/xxgk/xxgk15/202001/t20200123_760936.html (in Chinese).

MEE, 2020b. (Ministry of Ecology and Environment of the People's Republic of China): National Air Quality Report. Ministry of Ecology and Environment of the People's Republic of China available at: http://www.mee.gov.cn/hjzl/dqhj/cskqzlzkyb/ (in Chinese).

NBS, 2017. Shandong Statistics Yearbook. China Statistics Press, Beijing.

Peng, L., Zhang, Q., Yao, Z., Mauzerall, D.L., Kang, S., Du, Z., Zheng, Y., Xue, T., He, K., 2019. Underreported coal in statistics: a survey-based solid fuel consumption and emission inventory for the rural residential sector in China. Appl. Energy 235, 1169–1182.

Shen, F., Zhang, L., Jiang, L., Tang, M., Gai, X., Chen, M., Ge, X., 2020. Temporal variations of six ambient criteria air pollutants from 2015 to 2018, their spatial distributions, health risks and relationships with socioeconomic factors during 2018 in China. Environ. Int. 137, 105556.

Wen, Z., Song, J., 2004. Effect of boiler combustion adjustment on NOx emission. Guangdong Electric Power 17, 25–27, 77 (in Chinese).

Xue, Y., Zhou, Z., Nie, T., Wang, K., Nie, L., Pan, T., Wu, X., Tian, H., Zhong, L., Li, J., Liu, H., Liu, S., Shao, P., 2016. Trends of multiple air pollutants emissions from residential coal combustion in Beijing and its implication on improving air quality for control measures. Atmos. Environ. 142, 303–312.

Yan, Q., Kong, S., Yan, Y., Liu, H., Wang, W., Chen, K., Yin, Y., Zheng, H., Wu, J., Yao, L., Zeng, X., Cheng, Y., Zheng, S., Wu, F., Niu, Z., Zhang, Y., Zheng, M., Zhao, D., Liu, D., Qi, S., 2020. Emission and simulation of primary fine and submicron particles and water-soluble ions from domestic coal combustion in China. Atmos. Environ. 224, 117308.

Zhang, Y., Liu, P., Queen, A., Misenis, C., Pun, B., Seigneur, C., Wu, S.-Y., 2006. A comprehensive performance evaluation of MM5-CMAQ for the Summer 1999 Southern Oxidants Study episode—Part II: gas and aerosol predictions. Atmos. Environ. 40, 4839–4855.

Yang, J., Kang, S., Ji, Z., Yin, X., Tripathee, L., 2020. Investigating air pollutant concentrations, impact factors, and emission control strategies in western China by using a regional climate-chemistry model. Chemosphere 246, 125767.

Zhang, Y., Jamesjayschauer, Zhang, Y., Zeng, L., Wei, Y., Liu, Y., Shao, M., 2008. Characteristics of Particulate carbon emissions from real-world Chinese coal combustion. Environ. Sci. Technol. 42, 5068–5073.

Zhang, H., Wang, Y., Hu, J., Ying, Q., Hu, X.M., 2015. Relationships between meteorological parameters and criteria air pollutants in three megacities in China. Environ. Res. 140, 242–254.

Zhang, Z., Wang, W., Cheng, M., Liu, S., Xu, J., He, Y., Meng, F., 2017. The contribution of residential coal combustion to PM 2.5 pollution over China's Beijing-Tianjin-Hebei region in winter. Atmos. Environ. 159, 147–161.