Emission Characteristics of Volatile Organic Compounds from Typical Coal Utilization Sources: A Case Study in Shanxi of Northern China

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ABSTRACT

China, where coal is the dominant energy resource, accounted for 50.5% of the world's coal consumption—or 1906.7 million tons of oil equivalent—during 2018. As a major source of volatile organic compounds (VOCs), coal utilization also led to high national emissions of these pollutants. In this study, we investigated the profiles; benzene, toluene, ethylbenzene, and xylene (BTEX) ratios; ozone formation potential (OFP); and secondary organic aerosol (SOA) formation potential of VOCs generated by coal-utilizing steel plants, power plants, coking plants, and residential stoves in northern China. Among the detected VOCs, the results identified 1-butene as the most abundant species for both the power plants (36.7%) and the residential stoves (41.7%) as well as a significant contributor at the steel plants (7.3%), and alkenes, alkanes, and aromatics composed the largest groups for the power plants (42.0%) and residential stoves (60.2%); steel plants (59.2%); and coking plants (66.1%), respectively. Additionally, the VOC profiles for power plants employing the same coal source or combustion technology exhibited strong similarities, although the BTEX ratios varied more between plants using different coal sources than those using different combustion technologies. Finally, alkenes were primarily responsible for the ozone formation (73.1%, 59.0%, and 87.6% for the power plants, steel plants, and residential stoves, respectively), whereas aromatics were primarily responsible for the SOA formation (more than 94.0% for all four coal-utilizing sources).

Keywords: Volatile organic compounds, Source profiles, BTEX, Ozone formation potential, Coal utilization source

1 INTRODUCTION

The tropospheric ozone concentration in China has continually increased in recent years. Compared to 2018, the average concentrations of PM_{2.5}, PM₁₀, SO₂, NO₂, and CO in 337 major cities declined in 2019 by 0%, 1.6%, 15.4%, 0%, and 0%, respectively, while only ozone concentration increased by 6.5% (Ministry of Ecological Environment, 2020). As the main precursor of ozone, VOC pollution is the main cause of serious tropospheric ozone pollution problems. Moreover, VOCs play an important role in complex atmospheric pollution formation, which is linked to the formation of peroxides, aldehyde, ketone, peroxyacyl nitrates (PANs), and SOA (Yao *et al.*, 2006). Atmospheric VOCs can also adversely affect human health, plants, animals and the ecological environment (Zou *et al.*, 2013). The U.S. EPA (2020) identified 389 hazardous air pollutants that

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have severe negative impacts on human health and the environment. The priority control pollutants list includes benzene series and many other VOC species because of their strong irritating, carcinogenic, and toxic effects. Therefore, controlling VOC emissions can help reduce concentration of ozone and PM_{2.5}, decreasing photochemical smog pollution (Liu *et al.*, 2008a) and mitigating the adverse impact of air pollution on human health.

As the largest coal producer and consumer in the world, China consumed 2.80 billion tons of standard coal in 2019, accounting for 57.6% of the total primary energy consumption (National Bureau of Statistics, 2020). Coal consumption is one of the most important energy sources in China, and the national total coal consumption is expected to continue to increase (Dai, 2017). China's coal-dominated energy consumption is predicted to continue for at least 20 years (Liu, 2014). Power plant, steel plant, and coking plant are the key industries of national economy. The electric energy production of coal-fired power plant accounted for 64.2% of national electricity generation in 2019 (China Electricity Council, 2020). In addition, China is the largest coke and steel producer in the world (China Coking Industry Association, 2019). According to the data from the National Bureau of Statistics (2020), 95.8% of the total coal consumption was used for industrial purposes in China, and the industries (production and supply of electric and heat power, mining and washing of coal, smelting and pressing of ferrous metals) related to coal-fired power generation, steelmaking and cokemaking accounted for 61.8% of the total coal consumption in 2018 (National Bureau of Statistics, 2019). As significant coal usage industries in China, coal-fired power generation, steel, and coking industries have attracted widespread attention. In addition, coal is the main fuel for rural residential cooking and heating stoves in China. A previous study showed that there are approximately 60 million residential coal-fired heating stoves in rural China (Wang *et al.*, 2016). However, due to its direct combustion mode, exhaust mode operations, low combustion efficiency, lack of stack gas dedusting and desulfurization equipment (Tsai *et al.*, 2003; Zhang and Smith, 2007; Wang *et al.*, 2016), the flue gas pollution produced by residential coal usage is one of the main sources of winter haze in northern China, which also poses a serious indoor threat to the health of rural residents (Liu *et al.*, 2017). The VOCs produced from the coal utilization processes mentioned above are significant sources of atmospheric VOC pollution. A previous study identified coal-fired thermal power generation (Yan *et al.*, 2016) and flue gas from coking process (Li *et al.*, 2014) to be the main anthropogenic sources of VOCs in the northern city of China.

Many studies have analyzed the VOC emission characteristics from vehicles (Kawashima *et al.*, 2006; Olson *et al.*, 2009; Yao *et al.*, 2016; Zhang *et al.*, 2016), biomass burning (James *et al.*, 2001; Evtyugina *et al.*, 2013; Jumpponen *et al.*, 2013), solvent (Niedojadlo *et al.*, 2007; Yuan *et al.*, 2010; Mo *et al.*, 2015; Tian, 2015), petrochemical (Chen *et al.*, 2006; Zhan, 2015), and refining (Cetin *et al.*, 2003; Lin *et al.*, 2004), but research on VOC emissions from coal utilization sources is limited. In previous studies, Fernández-Martínez *et al.* (2001) studied the characteristics of 16 VOC species emitted during the coal utilization process in five Spanish coal power plants and found that aromatics, aliphatic hydrocarbons, and chlorinated compounds were the main VOC species formed during coal utilization. Shi *et al.* (2015) analyzed the VOC emission characteristics of coal-fired industry plants in Liaoning Province (northern China), and concluded that halogenated hydrocarbons were the representative VOC groups from iron smelt and coke production plants, while aromatics were the largest VOC group from heating stations. Li *et al.* (2018) studied the VOC emission characteristics of coal-fired coking plants, and power plants in Hebei Province (northern China), and the results showed that benzene, acetylene, and ethane were the three most abundant VOC species emitted from both power plants and coking plants. Santos *et al.* (2004) analyzed 36 VOC species from the stack gases of coal-fired power stations in Brazil, and found benzene to be the highest VOC species. In southern Taiwan, Tsai *et al.* (2008) investigated 68 VOC species emitted from four types of technology processes (including coke manufacturing, sintering, hot forming, and cold forming) in an integrated steelworks, and found that toluene, 1,2,4-trimethylbenzene, isopentane, *m,p*-xylene, 1-butene, ethylbenzene, and benzene were the dominant VOC species in these processes.

Scholars (Liu *et al.*, 2008a; Mo *et al.*, 2016; Wu and Xie, 2017) have already conducted significant research on VOC emission characteristics from different VOC emission sources and regions. Although the VOC emission characteristics of typical coal usage sources in China have been initially established, there are still many limitations and concerns regarding the accuracy of these

VOC source profiles: The types of investigated VOC emission sources are limited and insufficient, the measurement and analysis methods of the source profiles are not unified, and species selected for VOC source profiles are not consistent. VOC source profiles represent the chemical composition of VOC species and the relative contribution of a certain source, which can be used to identify the VOC emission sources and evaluate their influence on ozone formation. Previous studies (Mo *et al.*, 2014; Hu, 2017; Huang *et al.*, 2020) identified significant differences between some VOC source profiles from the same industrial emission source due to various factors, such as differences in technology and raw material. Relevant research (Mo *et al.*, 2016; Wu and Xie, 2017) also identified regional differences in VOC emission characteristics. Therefore, a systematic, standard and localized VOC source profile database is required to better understand the VOC characteristics of typical emission sources and solve current photochemical smog and ozone pollution problems. In this study, we investigated the VOC characteristics in five coal-fired power plants with different coal source usages and different coal combustion technologies. These findings have not been reported in previous studies. This study also collected VOC samples from two steel plant production stages to produce more refined VOC profiles from steel plants. We also conducted a comparative analysis of VOC profiles between four different coal utilization processes. The emission characteristics of VOCs from various coal utilization processes (coal-fired power plants, steel plants, coking plants, and residential stoves) were investigated in this study. The research objectives were to (1) conduct a comparative analysis of VOC source profiles from four types of coal utilization processes, (2) calculate BTEX ratios to identify the emission sources, (3) study the ozone formation potential of individual VOC species to investigate their influence on atmospheric ozone, and (4) estimate the SOA formation potential of VOC species emitted from coal utilization processes.

2 METHODS

The study area, Shanxi Province, is a typical coal consumption province in China. This province only accounts for 1.6% of China's land area but contributes approximately 11.8% of the total national coal consumption (National Bureau of Statistics, 2019). In this study, three types of industrial coal utilization processes (coking plants, steel plants, and power plants) and rural residential coal utilization processes were chosen to investigate the VOC emission characteristics in northern China.

2.1 Basic Information

Thermal power generation was the main electricity supply in China in 2019, accounting for 64.2% of the total electricity production, while coal-fired power plants accounted for 96.8% of the total thermal power generation (China Electricity Council, 2020). Five coal-fired power plants were selected in this study, which covered the two main types of power plant coal-burning boilers: Three power plants adopted pulverized coal-fired boiler technology, and two power plants adopted circulating fluidized bed technology (Ma *et al.*, 2015). China is the largest steel producer and consumer in the world. In steel plants, sintering and blast furnace ironmaking emit a significant mass of pollutants, including VOCs, for coal utilization. VOC samples were collected from both sintering and blast furnace ironmaking processes to study the emission composition characteristics of the steel plant stack exhaust gas. Cokemaking is another important VOC emission source in China; therefore, the flue gas from the coke oven of coking oven plants was also collected to analyze the VOC characteristics. In this study, we also investigated the VOC emission characteristics of rural cooking and heating stoves. The unqualified coal, low combustion efficiency, lack of dedusting and desulfurization facilities of rural stoves could result in serious atmospheric VOC pollution and even indoor air pollution. Detailed information on the chosen VOC emission sources is presented in Table 1.

2.2 Sampling Collection

The sample sites were located in exhaust chimneys or pollutant-monitoring points. VOC samples were collected using 1 L Tedlar bags. The studied compound concentration should be lower than the detection limit of the method. The VOC sample was collected into the sampling

Table 1. Detailed information of four types of VOC emission sources.

	Utilization type	Boiler tonnage (T)	Installed capacity (MW)	Denitration	Desulfurization	Dedusting	Coal source
Power Plant A	Pulverized coal-fired boiler	1025	300	Low-nitrogen-burning SCR	Wet limestone flue gas desulfurization (WLFGD)	Wet electrostatic precipitator (WESP)	Middle area of Shanxi
Power Plant B	Pulverized coal-fired boiler	1065	300	Low-nitrogen-burning SCR	WLFGD	Electrostatic precipitator (ESP)	Middle area of Shanxi
Power Plant C	Pulverized coal-fired boiler	1020	300	SNCR	Limestone flue gas desulfurization (LFGD)	WESP	South area of Shanxi
Power Plant D	Circulating fluidized bed	80	-	SNCR	LFGD	ESP	South area of Shanxi
Power Plant E	Circulating fluidized bed	130	58	SCR	LFGD	ESP	South area of Shanxi
Steel plant	Sintering furnace	-	-	-	LFGD	ESP and bag filter	-
Coking plant	Blast furnace	-	-	-	LFGD	ESP and bag filter	-
Residential coal usage	Coke oven	-	-	-	LFGD	ESP	Middle area of Shanxi
	Home-made cooking or heating stoves	-	-	-	-	-	South area of Shanxi

bag using a sampling pump with a flow rate of 0.5 L min⁻¹. The joint between the pump and pipe of the Tedlar sampling bag was airtight during sampling. Close the valve and label the Tedlar bag after sampling. Relevant sampling information was recorded for the subsequent analyses. After sampling, the samples were preserved in the laboratory at room temperature and in the absence of light. All samples were analyzed within 72 hours. The detailed sampling methods are as follows.

2.2.1 Coal-fired power plant

The pollutant-monitoring points of exhaust chimney in power plants were chosen to be sampling sites, which were about 10 m from the ground. A total of 12 samples were collected in the coal-fired power plants.

2.2.2 Steel plant

The pollutant-monitoring points of sintering and blast furnaces in steel plants were chosen to be sampling sites, which were about 15 m from the ground. As the water content of flue gas is relatively high in steel plants, a water absorption device was adopted for dehumidification. A total of 6 samples were collected in the steel plants.

2.2.3 Coking plant

The flue gas samples from air outlet of coke oven were collected in coking plant. A total of 6 samples were collected in the coking plants.

2.2.4 Residential stove

The flue gas samples were collected directly from the chimney of residential coal-fired heating or cooking stoves by using sampling pipe and pump. The height of sample sites was 5 m from ground. A total of 3 samples were collected.

2.3 Sample Analysis

VOC analysis was conducted using the U.S. EPA TO-15 method. The sample was analyzed by preconcentration (Model 7100; Entech Instruments) combined with Gas Chromatography-Mass Spectrometer (6890 GC/5973 MSD; Agilent). 300 mL of gas from the sample bags or canister were pumped into the preconcentrator and

injected into the GC/MS through a preconcentration sampling system. With super-pure helium as the carrier gas, HP-VOC capillary column chromatography (film thickness of 60 m × 0.32 mm × 1.8 mm; Agilent) was used to isolate the compounds. The GC oven temperature was set to 40°C, held for 2 min, then increased to 220°C at a rate of 5°C min⁻¹, holding for 2 min.

Based on electron ionization (EI), MSD runs were conducted in the scanning data acquisition mode, and the mass range was set to 35–300 amu. The method detection limit was less than 0.1 ppbv, and the repeatability was less than 10%. The detected variation in the same standard sample was less than 10%.

The target VOCs were identified according to retention time and standard mass spectrometry and then quantitatively analyzed using the external standard method. The standard gas mixture (1.0 ppm) was diluted with zero air and then sampled and analyzed under the same conditions as the field samples. A seven-point calibration curve (0.0, 1.0, 5.0, 10.0, 50.0, 100.0 and 200.0 ppbv) was used to quantitatively analyze the VOCs in air samples.

2.4 Quality Assurance and Quality Control

Before sampling, the pipe was purged with clean air to remove any impurities from the pipe surface. The pipe was inflated with the sample gas and connected to the VOC sampling device for sampling after the pipe was filled with smoke.

The pipelines of the VOC sampling system are made of Teflon materials for reducing the adsorption effect of the sampling pipe on VOC components. The sampling flowmeter was calibrated before sample collection to ensure the accuracy and stability of the flow rate.

The Tedlar sampling bags were washed at least five times before sampling. At least 1 of the 10 samples was injected with high-purity nitrogen to confirm the effectiveness of cleaning. The VOCs should be undetected or below the detection limit during the cleaning verification experiment. Before every VOC sample analysis, a blank test with high-purity nitrogen was conducted to ensure that the instrument system was not contaminated.

When drawing the standard curve, the relative standard deviation (RSD) of the response factors of each VOC species was less than 30%. A total of 47 species were analyzed in this study, and “total VOCs” refers to the sum of the 47 VOCs.

2.5 Data Analysis

According to the target list developed by the U.S. EPA Photochemical Assessment Monitoring Stations (PAMS) program, the VOC species in this study were selected based on their reactivity to photochemical smog and tropospheric ozone formation. The OFP is related to the chemical reaction activity of VOCs in the atmosphere, and three main methods are typically used to research organic reactivity at the current level: the propylene-equivalent concentration method, OH radical reactivity method, and maximum increment reactivity method (MIR). The former two methods only consider the OH radical and VOC reaction rate and ignore the complicated subsequent reaction. The MIR coefficient method was selected to calculate the OFP of VOCs in this study because it is more objective with regards to the ozone formation reaction mechanism (Cheng *et al.*, 2012). The MIR coefficient method can be used to identify VOC species with high chemical reactivity because the MIR value refers to the ozone formation ability of VOC species under optimum reactive conditions.

The OFP of 47 VOC species were calculated by Eq. (1):

$$\text{OFP}_i = C_i \times \text{MIR}_i \quad (1)$$

where OFP_i is the ozone formation potential of VOC species i ($\mu\text{g-ozone m}^{-3}$); C_i is the mass concentration of VOC species i ($\mu\text{g m}^{-3}$); MIR_i is the corresponding maximum increment reactivity of VOC species i ($\mu\text{g-ozone m}^{-3}$) (Carter, 2010).

The fractional aerosol coefficient (FAC) was used to calculate the potential formation of SOA of VOCs. The SOA formation potential of 18 VOC species were calculated by Eq. (2):

$$\text{SOA}_p = \text{VOC}_0 \times \text{FAC} \quad (2)$$

where SOA_p is the SOA formation potential of VOC species ($\mu\text{g m}^{-3}$); VOC_0 is the initial mass

concentration or emissions of VOC species ($\mu\text{g m}^{-3}$); FAC is the corresponding fractional aerosol coefficient of VOC species (Grosjean, 1992; Lv *et al.*, 2009).

3 RESULTS AND DISCUSSION

3.1 VOCs Emission Characteristics and Source Profiles

The 47 VOC species were classified into four groups, including 26 alkanes, 10 alkenes, 10 aromatics, and 1 alkyne. As this study focused on the VOC source profile of a typical coal utilization process, the mass fraction of the 47 VOC species was calculated to investigate the VOC emission characteristics of typical coal utilization sources. The mass fractions of the 47 VOC species emissions from each coal utilization source are presented in Fig. 1, and the VOC group weight percentages from emission sources are shown in Fig. 2.

3.1.1 Power plant

The main VOC species in the flue gas of power plants were 1-butene, propane, isobutane, acetylene, toluene, and styrene. 1-Butene was the largest contributing VOC species of Power

Fig. 1. VOC source profiles of coking plant, steel plants, power plants and residential stoves.

Fig. 2. VOC group weight percentages from different emission sources.

Plants A, C, D, and E (weight fractions of 57.0%, 25.8%, 44.9%, and 29.8%, respectively), and the second-largest contributor in Plant B (with 26.0% of weight fraction). As shown in Fig. S1, the high weight percentage of 1-butene in this study was consistent with the previous results from a coal-fired power plant in Shanxi Province (Yan *et al.*, 2016), but much higher than that in Liaoning (Shi *et al.*, 2015) and Hebei Province (Li *et al.*, 2018), with the values of 1.4% and 0.6%, respectively. The characteristics of some VOC species showed the obvious differences between the two types of coal combustion technology. The weight fractions of the major VOC species propylene, propane, acetylene, benzene, and toluene from power plants with circulating fluidized bed boilers were apparently higher than those from pulverized coal-fired boilers; these results were the same as the findings observed in circulating fluidized bed boilers and pulverized coal-fired boilers in power plants in Hebei Province (Table S1; Li *et al.*, 2018). In addition, due to the different sources of coal, the percentages of some VOC species showed large differences between Power Plants A and B (with coal sourced from the middle area of Shanxi) and Power Plants C, D, and E (with coal sourced from the southern area of Shanxi). For instance, the weight fraction of styrene was significantly higher in Power Plants A and B (19.6% and 29.5%, respectively), while the percentages of styrene in Power Plants C, D, and E were all less than 1.0%. The weight fractions of benzene in Power Plants A and B were less than 1.0%, which were lower than those of Power Plants C, D, and E (6.4%, 2.1%, and 2.5%, respectively). The same situation was also observed for toluene, with toluene percentages of 0.5% and 1.4% for Power Plant A and B, respectively, and 16.0%, 5.6%, and 5.5% for Power Plants C, D, and E, respectively.

The major VOC groups of the three power plants with pulverized coal-fired boilers (A, B, and C) were alkenes and aromatics, accounting for 89.7%, 79.9%, and 71.0%, respectively. The main VOC groups of the two power plants with a circulating fluidized bed boiler (C and D) were alkenes and alkanes, in total accounting for 82.1% and 73.4%, respectively. Alkyne from the three pulverized coal-fired boiler power plants were all less than 1%, while those of two circulating fluidized bed boiler power plants were 11.6% and 3.4%, respectively. According to SPECIATE 5.1 (U.S. EPA, 2020), aromatics were the main VOC group (64.8%) from the stack flue gas of coal-burning boiler electric generation, followed by alkanes (23.4%) and alkenes (11.8%). In comparison with the SPECIATE 5.1 data, we measured more alkenes and fewer aromatics in the coal-fired power plants in this study. In contrast, previous studies identified alkanes (Shi *et al.*, 2015; Yan *et al.*, 2016; Li *et al.*, 2018) to be the largest VOC group in China. Therefore, these variations were likely caused by regional differences in coal combustion technology and coal quality. Our VOC source profile of power plants suggests that the VOC emission characteristics were largely affected by both coal utilization technology and coal sources.

3.1.2 Steel plant

The main VOC species in the flue gas of blast furnace ironmaking were 2-methylpentane, 2,3-dimethylbutane, cyclopentane, 1-butene, and toluene, while the main VOC species in the sintering process were ethane, benzene, propylene, 1-butene, and propane. Benzene and toluene were also found to be the main species in the ironmaking process in Liaoning (Shi *et al.*, 2015) and Taiwan (Tsai *et al.*, 2008) steel plants, as shown in Fig. S2 and Table S2. The largest VOC species from the ironmaking process was 2-methylpentane, accounting for 26.5%, but this species was not detected in the ironmaking process in Liaoning (Shi *et al.*, 2015). As for the sintering process, the weight fraction of benzene was 14.4%, which is consistent with the weight fraction of 16.1% reported by Miao (2017). Propylene (10.2%), 1-butene (7.2%), toluene (6.1%), propane (5.4%), ethylene (5.3%), and *n*-butane (3.5%) were the main species in the flue gas during sintering process. Among these species, benzene, 1-butene, *n*-butane, and toluene were also the most abundant species in the sintering process of Taiwan steel plants (Tsai *et al.*, 2008). Notably, ethane was the largest VOC species in the flue gas of the sintering process in this study. According to SPECIATE 5.1 (U.S. EPA, 2020), ethane was also the major compound in the iron sintering process but was not detected in some steel plant studies (Tsai *et al.*, 2008; Shi *et al.*, 2015; Miao, 2017). Alkane was the most abundant VOC group emitted from both the blast furnace ironmaking process and sintering process, accounting for 69.5% and 48.9%, respectively (Fig. 2). Previous studies identified aromatics to be the largest VOC group in Taiwan and Liaoning (Tsai *et al.*, 2008; Shi *et al.*, 2015). The weight fractions of aromatics, alkenes, and alkynes in the blast furnace ironmaking process were 17.3%, 13.1%, and 0.1%, respectively, and in the sintering process were 26.8%,

22.1%, and 2.2%, respectively (Fig. 2). The different combustion technologies, combustion temperature, burning materials, and VOC source profiles in this study suggest that the emission characteristics of VOCs differed between the two coal utilization processes of steel plants.

3.1.3 Coking plant

The main VOC species from the coking plant were benzene, styrene, ethylene, ethane, and propane. Benzene was the largest VOC species contributor, contributing 32.1% to the total VOCs. As shown in Fig. S3 and Table S3, styrene was the main VOC species in the coking plant in this study (mass fraction of 35.6%), which is contradictory to previous findings of similar studies that determined styrene mass fractions of less than 1% (Jia *et al.*, 2009; Shi *et al.*, 2015; Wang *et al.*, 2017; Li *et al.*, 2018). Benzene was also the main VOC species in the coking plant, with a weight fraction of 17.6%, and was also the main species (with a weight fraction of 20.5%) in Hebei Province (Li *et al.*, 2018). In other similar studies (Jia *et al.*, 2009; Shi *et al.*, 2015; Wang *et al.*, 2017), the percentage of benzene was much lower than in this study, with 8.1% reported by Shi *et al.* (2015), 4.5% reported by Wang *et al.* (2017), and 0.2% and 0.4% reported by Jia *et al.* (2009). The weight fraction of ethene was 9.6% and was identified as the main VOC emission species from coking plants in other studies (Jia *et al.*, 2009; Wang *et al.*, 2017; Li *et al.*, 2018). The weight fraction of ethane was only 3.4% in this study and less than 0.1% in Liaoning Province (Shi *et al.*, 2015), but its value in other studies ranged from 18.5% to 28.9% (Jia *et al.*, 2009; Wang *et al.*, 2017; Li *et al.*, 2018). The VOC characteristics in coking plants showed large differences between different regions due to the different coke oven models, materials, and coking temperatures applied. Among the species, aromatics were the largest contributors (66.4%), followed by alkanes (19.1%), alkenes (14.1%), and alkynes (0.9%).

3.1.4 Residential stove

In this study, 1-butene was the largest contributor to the total VOC emissions from residential coal-burning stoves, accounting for 44.9%. 1-Butene was also the main VOC species identified in three different types of coal (raw anthracite, raw bituminous coal, and briquette of bituminous coal) (Liu *et al.*, 2008a). The second-largest species in the stack gas of residential stoves was ethylene, accounting for 12.5%, which was consistent with the results of other residential coal utilization studies, including 12.6% from Liu *et al.* (2008a) and 14.4% from Liu *et al.* (2017). As shown in Fig. S4 and Table S4, a higher weight fraction of ethylene was also found in many previous residential coal usage studies (Tsai *et al.*, 2003; Wang *et al.*, 2013; Mo *et al.*, 2016). In addition, the abundant VOC species of residential coal usage included styrene (12.6%), 2,2-dimethyl butane (5.5%), propylene (4.5%), acetylene (2.5%), *n*-pentane (2.5%), ethane (2.3%), and benzene (1.9%). Among these species, ethane, acetylene, propylene, and benzene have also been observed as the dominant species in many similar studies (Tsai *et al.*, 2003; Liu *et al.*, 2008a; Wang *et al.*, 2013; Mo *et al.*, 2016; Liu *et al.*, 2017). Alkenes were the major VOC group in the flue gas of residential coal-burning stoves, followed by aromatics, alkanes, and alkynes, accounting for 60.1%, 19.5%, 17.9%, and 2.5%, respectively. Some previous studies also found alkenes to be the most abundant VOC group in residential coal usage (Wang *et al.*, 2013; Mo *et al.*, 2016), while Liu *et al.* (2017) found alkanes to be the largest VOC group in residential coal usage, regardless of smoldering or flaming combustion processes. The results from Taiwan also showed that alkanes were the largest VOC group from residential coal usage (Tsai *et al.*, 2003).

3.2 BTEX Ratio

The ratios of two particular VOC species of benzene series (benzene, toluene, ethylbenzene, and xylene) have been widely used to investigate the emission characteristics of VOCs. In a previous study, B/T ratios higher than 1 were considered to be associated with coal-fired industrial emissions (Santos *et al.*, 2004; Liu *et al.*, 2008a). As shown in Table 2, the average B/T ratios in this study varied greatly in different coal utilization processes. The B/T ratios in the power plant, sintering process of the steel plant, ironmaking process of the steel plant, coking plant, and residential stove were 0.47, 2.34, 0.56, 7.03, and 2.99, respectively.

The B/T ratios of five power plants ranged from 0.37 to 0.57, which are higher than those of coal-fired power stations in Liaoning Province (Shi *et al.*, 2015) and lower than those of power

plants in Korea (Pudasainee *et al.*, 2010), Brazil (Santos *et al.*, 2004) and northern China (Yan *et al.*, 2016). As shown in Table 2, the BTEX ratios showed large differences between power plants that used different sources of coal. For instance, the B/E ratios were between 2.14 and 2.43 for Power Plants C, D, and E, and only 0.16 and 0.31 for the other two power plants. The B/E ratios were 0.29 and 0.55 for Power Plants A and B, respectively, and > 5 for Power Plants C, D, and E. The same observations were also noted for the B/T, T/E, T/X, and E/X ratios. Dagaut and Cathonnet (1998) showed that benzene and toluene were formed from low-molecular unsaturated hydrocarbons, which cracked macromolecules in the fuel, while xylene was directly formed from the incomplete combustion of macromolecules; therefore, the different coal qualities may have caused the observed differences in BTEX values. The B/T ratio of steel plants in our studies was 2.34 for the sintering process and 0.56 for the blast furnace ironmaking process, which are within the range of previous studies. As previously mentioned, the contribution rates of benzene to VOC concentrations in the stack gases of coking plants were much higher than those of other coal-fired industries. The B/T ratios of coking plants in this study were 7.03 compared with 0.14–1.28 in other studies. The B/T ratio of residential coal-burning stoves was 2.99, which is consistent with the B/T ratio (2.86) of residential coal usage determined by Wang *et al.* (2013). Compared with previous studies, the B/T ratio of residential coal usage in this study was lower than that reported by Mo *et al.* (3.47; 2016) but higher than that reported in other similar studies (Tsai *et al.*, 2003; Liu *et al.*, 2008a; Liu *et al.*, 2017).

Table 2. BTEX ratios from different VOC emission sources.

Source	Reference	B:T	B:E	B:X	T:E	T:X	E:X	Notes
Power plants	Power Plant A (this study)	0.57	0.16	0.08	0.29	0.14	0.49	Pulverized coal-fired boiler
	Power Plant B	0.57	0.31	0.11	0.55	0.20	0.36	Pulverized coal-fired boiler
	Power Plant C	0.40	2.43	0.68	6.09	1.70	0.28	Pulverized coal-fired boiler
	Power Plant D	0.37	2.14	0.60	5.84	1.64	0.28	Circulating fluidized bed boiler
	Power Plant E	0.45	2.36	0.61	5.20	1.34	0.26	Circulating fluidized bed boiler
	Pudasainee <i>et al.</i> (2010)	1.73						Anthracite coal
		0.80						Bituminous coal
	Santos <i>et al.</i> (2004)	8.53	33.06	55.71	3.88	6.53	1.69	
	Shi <i>et al.</i> (2015)	0.17	2.75	1.20	15.80	6.88	0.44	Pulverized coal-fired boiler
	Yan <i>et al.</i> (2016)	1.02	0.19	0.11	0.19	0.11	0.58	Anthracite coal
Steel plants	Li <i>et al.</i> (2018)	2.84	5.16	3.30	1.81	1.16	0.64	
	This study	2.34	60.53	12.83	25.90	5.49	0.21	Sintering
		0.56	3.61	0.86	6.47	1.54	0.24	Ironmaking process
	Tsai <i>et al.</i> (2008)	0.25	1.86	0.89	7.52	3.62	0.48	Sintering
	Miao (2017)	3.61	12.15	5.77	3.37	1.60	0.48	Sintering
	Shi <i>et al.</i> (2015)	0.16	3.29	1.37	20.56	8.59	0.42	
Coking plants	This study	7.03	7.17	3.29	1.02	0.47	0.46	Machine coke oven
	He <i>et al.</i> (2005)	0.22	1.73	0.32	8.01	1.45	5.46	Traditional coke oven
		0.48	4.18	1.22	8.65	2.53	3.41	Machine coke oven
	Tsai <i>et al.</i> (2008)	0.14	0.91	0.50	6.66	3.67	0.55	Cokemaking
	Jia <i>et al.</i> (2009)	1.27	5.00	0.79	3.93	0.62	0.16	58-II type
		1.28	15.95	1.75	12.42	1.36	0.11	JN43-80 type
	Shi <i>et al.</i> (2015)	0.29	3.11	1.84	10.58	6.27	0.59	
	Li <i>et al.</i> (2018)	3.06	68.33	9.76	22.33	3.19	0.14	
	Wang <i>et al.</i> (2017)	1.28	9.49	1.26	7.40	0.99	0.13	
	This study	2.99	1.48	0.92	0.50	0.31	0.62	
Residential stove	Wang <i>et al.</i> (2013)	2.86						
	Liu <i>et al.</i> (2008)	2.07		5.68		2.74		Raw anthracite
	Liu <i>et al.</i> (2017)	1.13		3.72		3.29		Smoldering combustion
		1.91						Flaming combustion
	Mo <i>et al.</i> (2016)	3.47	13.57	6.25	3.91	1.80	0.46	
	Tsai <i>et al.</i> (2003)	1.99	16.48	2.17	8.26	1.09	0.13

According to previous studies that assessed B/T ratios (Suthawaree *et al.*, 2012), the B/T value can be used to distinguish traffic and non-traffic sources, with B/T ratios of approximately 0.5 regarded as traffic sources. However, in this study, the B/T ratios of the five power plants and ironmaking processes in steel plant blast furnaces were both approximately 0.5, and the B/T ratio of the coke oven in He *et al.* (2005) was 0.48. Therefore, the use of the B/T ratio to identify vehicular emission sources may not be sufficient.

3.3 OFP and SOA

As VOCs are the main precursors of ozone formation and SOAs, we analyzed the OFP and SOA formation potential of selected VOC species to investigate the influence of typical coal utilization processes on atmospheric ozone (Yao *et al.*, 2006; Liu *et al.*, 2008a).

3.3.1 OFP

The major ozone-forming species in the power plants was 1-butene, contributing approximately 67.2% to the ozone formation. Alkenes and aromatics were the main contributors to ozone formation in this study, accounting for 72.6% and 20.0%, respectively; they were also identified as major OFP VOC species in coal-burning power plants in previous studies (Shi *et al.*, 2015; Yan *et al.*, 2016). Among the five power plants, the OFP results differed between power plants with pulverized coal-fired boilers, while 2-methylpentane and cyclopentane were the main OFP VOC species in both power plants that use circulating fluidized beds. In coking plants, the two most abundant oxygen-forming species were ethylene and styrene, contributing 46.2% to the total ozone formation. Although ethylene only accounted for 9.6% of the total VOC emissions from coking plants, the OFP of ethylene contributed 27.0% to ozone formation. In comparison, the OFP of alkenes and aromatics accounted for 37.2% and 47.0%, respectively. No alkanes or alkynes were observed in the top 10 OFPs of VOC species in the coking plant. As for steel plants, propylene, 1-butene, toluene, *cis*-2-butene, and *m*-xylene were the main species contributing to the OFP in both the blast furnace ironmaking process and the sintering process. Notably, although alkenes accounted for only 22.8% and 9.9% of the total VOC emissions from the blast furnace ironmaking process in

Fig. 3. Contribution of VOC groups and the top 10 VOC species to the OFP in coking plants, steel plants, power plants, and residential stoves.

Table 3. Weight percentages of SOA formation for VOC species.

VOC species	FAC (%)	CONTRIBUTION RATE (%)								
		Plant A	Plant B	Plant C	Plant D	Plant E	Sintering furnace	Blast furnace	Coking plant	Residential usage
Isoprene	2	0.23	0.54	0.02	0.12	0.20	0.12	0.03	0.98	0.84
Methylcyclopentane	0.17	0.05	0.04	0.03	0.09	0.13	0.11	0.18	0.03	0.06
Cyclohexane	0.17	0.19	0.29	0.02	0.08	0.12	0.15	0.18	0.17	0.07
<i>n</i> -Heptane	0.06	0.06	0.03	0.02	0.08	0.12	0.02	0.09	0.12	0.22
Methylcyclohexane	2.7	0.46	0.51	0.91	2.01	3.20	1.69	3.57	0.22	0.25
2-Methylheptane	0.5	0.14	0.13	0.12	0.35	0.51	0.04	0.30	0.03	0.04
3-Methylheptane	0.5	0.12	0.11	0.09	0.31	0.52	0.07	0.35	0.03	0.05
<i>n</i> -Octane	0.06	0.04	0.04	0.02	0.04	0.07	0.01	0.04	0.01	0.06
<i>n</i> -Nonane	1.5	3.22	3.14	0.62	1.08	0.81	0.09	0.65	0.80	2.25
Benzene	2	1.31	1.77	7.50	6.79	8.09	40.97	10.22	36.50	13.30
Toluene	5.4	6.18	8.43	50.84	50.03	48.11	47.33	49.43	14.02	12.00
Ethylbenzene	5.4	21.52	15.28	8.34	8.57	9.24	1.83	7.64	13.74	24.23
<i>m</i> -Xylene	4.7	16.56	15.88	15.07	14.57	13.65	5.38	15.89	15.59	20.54
<i>p</i> -Xylene	1.6	2.59	2.73	1.80	2.35	3.93	0.42	2.42	0.78	0.80
<i>o</i> -Xylene	5	14.72	13.65	6.11	5.35	6.40	0.94	5.16	8.75	11.92
1,3,5-Trimethylbenzene	2.9	9.69	10.63	1.31	1.44	1.07	0.19	0.84	2.18	3.65
1,2,4-Trimethylbenzene	2	8.02	9.37	2.99	2.90	1.48	0.25	1.34	2.84	5.13
1,2,3-Trimethylbenzene	3.6	14.91	17.42	4.17	3.84	2.35	0.39	1.67	3.20	4.57

steel plants, the OFP of alkenes in the top 10 VOC species contributed 76.0% and 37.2% to ozone formation in the two processes. In contrast, the alkane contribution rate to VOC emissions from the two steel plant processes was 41.0% and 55.3%, respectively, while the contribution of alkanes to ozone formation declined to 2.3% and 25.4%, respectively. This suggests that the chemical reactivity of alkenes during ozone formation is much higher than that of alkanes. Aromatics from steel plants contributed 11.7% and 18.6% to the OFP in the two processes, respectively. The main contributing species to the OFP in residential coal usage were 1-butene, ethylene, and propylene, accounting for 61.0%, 16.9%, and 7.9%, respectively. Moreover, alkenes had the highest contribution rate to ozone formation at 68.7%, followed by aromatics (8.0%) and alkanes (1.0%).

The contribution of different VOC species to ozone formation was influenced by both their concentration mass fractions and their maximum incremental reactivity coefficient value. Low-concentration mass fraction VOC species may have a high contribution rate to ozone formation; therefore, both high-concentration mass fractions and high-OFP VOC species should be considered for the effective control of VOC pollution. Our results showed that alkenes were the dominant oxygen-forming VOC group in the four types of coal utilization processes, and aromatics were the major ozone-forming VOC group in coal-fired steel plants and power plants. Alkenes were the largest contributor to the OFP in all five power plants, with an alkene percentage of more than 50%, and the major VOC species were similar among power plants. To improve the effectiveness of ambient ozone control measures, policymaking agencies should focus more on alkenes and aromatics when establishing related policies for controlling VOCs.

3.3.2 SOA

VOCs also chemically react with atmospheric oxidants (OH, NO₃, and O₃) in the atmosphere, and the oxidation products subsequently form SOA, which influences atmospheric visibility. The potential formation of SOA in this study was estimated using fractional aerosol coefficients (Grosjean, 1992). According to Table 3, the top 5 VOC species contributing to the total SOA formation in the flue gases of coal utilization processes are all in the benzene series. In the four types of coal utilization processes, aromatics accounted for more than 94.0% of the total SOA formation. *n*-Nonane was the major VOC species contributing to SOA formation from power plants and residential stoves, and methylcyclohexane was the major VOC species of SOA formation from power plants and steel plants.

4 CONCLUSION

We evaluated the emission characteristics, OFP-related chemical reactivity, and SOA formation potential of VOCs originating from four types of coal-utilizing sources in northern China, namely, steel plants, power plants, coking plants, and residential stoves. The VOC profiles significantly varied between different the coal utilization processes. Both the coal source and the combustion technology influenced the profiles for the five studied power plants. However, based on the BTEX ratios, the former factor played a larger role in shaping the BTEX emissions. Furthermore, at the steel plants, sintering generated alkanes and alkenes as the predominant VOC species (48.9% and 26.8% of the total VOCs, respectively), but blast furnace ironmaking generated alkanes and aromatics (69.5% and 17.3%). We also found far higher B/T ratios for coking plants than those previously reported but lower ratios for power plants (0.37–0.57) than the one commonly recognized (1.0). As we collected all of our samples from Shanxi Province, subsequent research should assess whether these values are location-specific.

Additionally, we identified the largest contributors to the OFP of the emitted VOCs as alkenes for the steel plants, power plants, and residential stoves (59.0–87.6%) but as aromatics for the coking plants (55.8%). Notably, alkenes accounted for 33.6% of the total OFP at the steel plants (ironmaking process) but only 0–8.4% for the other sources. Lastly, above 94.0% of the SOA formation was attributable to aromatics across all of the studied sources.

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DISCLAIMER

The authors declare no competing financial interests.

SUPPLEMENTARY MATERIAL

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