The Geological Availability and Emissions of Sulfur and SO₂ from the Typical Coal of China

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ABSTRACT

This study aimed to assess the natural availability of sulfur and SO₂ in coal typical of the Jurassic, Permo-Carboniferous, and Cambrian strata in Shaanxi, China, and their emission rates. A total of 93 samples (39 Binxian Jurassic, 37 Permo-Carboniferous, and 17 Langao Cambrian) were collected and analyzed via the Eschka method (GB/T 214-1996). The results show that the average sulfur content was 2.40%, 2.85%, and 0.92% in the Binxian coal gangue, raw coal, and coal slime, respectively; 1.48%, 2.41%, and 1.5% in the Hancheng Permo-Carboniferous coal gangue, raw coal, and coal slime, respectively; and 0.84% and 2.44% in the Langao Cambrian stone-like coal and black shale rock, respectively. The annual sulfur emissions from the Binxian urban and rural areas totaled 1.5 kt and 9.3 kt (Kilotons), respectively, which contributed 1.4% of the overall SO₂ emitted into the atmosphere. The sulfur emissions from Hancheng urban and rural areas totaled 1.8 kt and 11.9 kt, respectively, which contributed 1.8% of the overall SO₂. The sulfur emissions from Langao urban and rural areas was 0.4 kt and 2.8 kt, respectively, which contributed 0.43% of the overall SO₂. Coal-waste consumption from 1991 to 2015 increased by 23% and 10% in urban and rural areas, respectively, in China, ultimately reducing the debris from coal waste. Raw-coal consumption from 1991 to 2015 decreased from 96% to 73% and from 97% to 87% in urban and rural areas, respectively. SO₂ emissions since 2006 have decreased due to effective desulfurizing technology. According to the results of this study, China has been continuously reducing the emission of SO₂ by adopting a green economy. The study recommends installing desulfurizing equipment in power plants to further reduce the SO₂ emissions.

Keywords: Cambrian; Coal; Jurassic; Shaanxi; Sulfur; SO₂.

INTRODUCTION

Sulfur is one of the major components/impurities of coal, available from 0.2 to 11% both in organic and inorganic form (Ibarra *et al.*, 1994). During coal and other crude oil combustion, sulfur is emitted to atmosphere in the form of SO₂, SO₄, etc., while in the coal waste, sulfur is available in the form of pyrite (FeS₂), gypsum (CaSO₄ · 2H₂O), ferrous sulfate (FeSO₄), barite, and sphalerite (ZnS) (Lu *et al.*, 2010). Among all the forms, the most active and major emitted form is SO₂ and H₂S (Hou *et al.*, 2018). Sulfate aerosol in the atmosphere acting as a cooling inhibitor but their cooling effect will affect though atmospheric CO₂ (Ozkaymak *et al.*, 2017; Samset, 2018). The dominant cooling effect of sulfate aerosol is considered the regional and urban pollutant, which has a wide range of health and environment impacts (Ming *et al.*, 2005). The massive emission of sulfur oxides to the atmosphere may cause air pollution; contribute SO₂

and acid rain, damaging infrastructure, affect plants growth, contribute haze and particulate matters (Saidur *et al.*, 2011). The declining level of the ecosystem and human health affect the life expectancy, which is a great economic loss in long-term (Chen *et al.*, 2018).

China is one of the major energy-consuming country in the world. Due to lack of local oil resources, China used coal resource for the energy purposes, which ultimately contribute a huge amount of the toxic gases (H₂S, CS, SO₂, CS₂, COS etc.) to the atmosphere, which affect the atmosphere and biosphere (Ali *et al.*, 2015; Gu *et al.*, 2017). Among the world major air polluted cities, 16 worst air quality cities are situated in China, while 54% of the river's water is not safe for the human uses. The intense use of coal resource not only released the sulfur but also emits other toxic elements to the environment, which drastically affects the agriculture lands, food crops and water quality (Hussain *et al.*, 2018). Additionally, some poisonous gases were detected in the worst air quality cities, where the higher SO₂ and lung cancer are common (Chen *et al.*, 2018). Kan and Chen (2003) reported that the increasing SO₂ (10 mg m⁻³) emission will lead to an increase in the mortality risk of 1.01% through the respiratory system.

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While some previous epidemiologist reported 0.8–3% dual mortality by emitting $50 \mu\text{m m}^{-3}$ of SO_2 (Katsouyanni and Pershagen, 1997).

To fulfill the energy demand of the country, the Chinese government still planning to install the coal power plants to overcome the energy shortfall. However, these power plants generate more SO_2 . During 1980–1995, China overall SO_2 emission was 23 million tons (Mt) (Yang and Schreifels, 2003), while in 2000, the China coal industries emitted almost 85% (19.9 Mt) of SO_2 (Yang *et al.*, 2002). Due to extreme energy demand in 2006, Chinese government installed 1056 new coal power plants (which generate 63 million kilowatts [MkW] electricity) to achieve the energy goal of 2005 five-year plan (Chen *et al.*, 2018). However, these newly established power plants generate 11.7% SO_2 to the atmosphere (Su *et al.*, 2011). SO_2 emission control is one of the key tasks to all over the world, but very crucial to the China and the USA energy sectors. The Chinese energy cleaning system was improved, but still, the SO_2 emission is greater than 40% (Steinfeld *et al.*, 2009). Chinese industrial SO_2 emission was about 42–52% from 2001 to 2007 (Wang *et al.*, 2018a). Among this emission, $\approx 90\%$ is associated with coal burning both in industrial and commercial uses (Lu *et al.*, 2010).

Some previous studies estimate the sulfur and SO_2 emission of the China and global up to 2008, which shows a drastic increase in the SO_2 emission from 1999 to 2006 (Smith *et al.*, 2011). However, a few studies compiled the SO_2 emission but lack of the sulfur availability and emission from the coal (Tørseth *et al.*, 2012; Wang and Hao, 2012; Klimont *et al.*, 2013). While some of the research has been conducted on the toxic elements released by coal (Zhao and Luo, 2017, 2018), very limited studies are available on the sulfur emission. Sulfur emission is mostly associated with the burning of coal and other fossil fuels, which could cause serious environmental problems, including atmospheric gases unbalancing, deteriorate air quality, increasing global warming and indirectly affect the cryosphere, agriculture land, food crops, and human's health. Therefore, the study aimed to probe and assess the sulfur quantity in the coal and coal waste byproduct (CBWs). The study also aimed to deliver a foundation and explore the critical estimation of the SO_2 emission by China since last 25 years.

METHODOLOGY

Tectonic Setting and Sampling

The field visit and sampling were carried in the three major coalfields (typical coal of China) of Shaanxi Province, China. (1) Langao County (Cambrian coal) is situated between the latitude $32^\circ 26' 13''\text{N}$ to $32^\circ 28' 7''\text{N}$, and longitude $108^\circ 56' 25.5''\text{E}$ to $108^\circ 58' 49''\text{E}$ (Hussain and Luo, 2018). Tectonically, the area is related to Qinling Mountains, where the most visible strata's are late Proterozoic and early Paleozoic, however, a few Jurassic strata's are also expended in some of the areas. The stone-like coal and black shale are widely spread in the Lujiaping formation of the late Proterozoic Lower-Cambrian. The coal and black shale carbonaceous rocks are enriched with sulfide ores,

i.e., molybdenite, pyrite, and chalcopyrite, etc. (Hanjie and Yuzhuo, 1999; Wang *et al.*, 2017; Hussain and Luo, 2018). (2) Weibei coalfield (Hancheng Permo-Carboniferous) is situated in the Ordos Basin between the latitude of $35^\circ 34'\text{E}$ to $35^\circ 38'\text{E}$ and longitudes of $110^\circ 25' 30''\text{E}$ to $110^\circ 31' 28''\text{E}$ (Fig. 1). The major tectonic of the Weibei coalfield are Cambrian, Ordovician, Permian, Carboniferous, Jurassic, Pliocene, Triassic and Pleistocene. The Weibei coal seam is associated with the Carboniferous and Permian geological sequence, which underlying the quaternary sequences (Yao *et al.*, 2009; Hussain *et al.*, 2018). (3) Huanglong coalfield (Binxian Jurassic coalmines) is a part of the Ordos basin located at the latitude of $35^\circ 2' 33''\text{E}$ to $35^\circ 5' 19''\text{E}$ and longitude $107^\circ 50' 42''\text{N}$ to $107^\circ 58' 42''\text{N}$ (Fig. 1). The coal seam of the Huanglong coalfield belongs to the Jurassic (J2), where the most visible formation are Zhiluo Jurassic formation, Yan'an formation, Luohe, Cretaceous, Yijun, Chihuahua, Quaternary and Fuping formation (Ren *et al.*, 2014; Yi *et al.*, 2015; Hussain and Luo, 2018).

The coal, coal wastes and rocks samples were collected from the above three coalfield areas, while the coal slime samples were collected from the washing plants. A total of 93 samples were collected including 39 from the Binxian Jurassic (23 coal gangue, 10 raw coal, 5 coal slime, and 1 ash sample), 37 from the Hancheng Permo-Carboniferous (18 coal gangue, 7 raw coal, 4 coal slime, and 6 spontaneous burning ash), and 17 samples were collected from the Langao Cambrian county (9 stone coal, and 7 Cambrian black shales rock). All these samples were analyzed for sulfur at the Physical and Chemical Laboratory of the Institute of Geographic Sciences and Natural Resource Research, CAS, China.

Experimental Method

Eschka method (GB/T 214-1996) was used to determine the sulfur contents in coal and rocks (Wang *et al.*, 2018b; Zhao and Luo, 2018). The samples were mixed with Eschka reagent (1:2) and then keep in the oven at $800\text{--}900^\circ\text{C}$ for 1–2 hours until all the sample were burnt completely. Then passed the sample through a medium sized filter paper using hot water. Add methyl orange indicator and 2 mL HCl (1:1) and transfer to heat to reduce the total volume up to 200 mL. Add 10 mL BaCl_2 (10%), kept the solution for a night, and then filter the solution using a 0.001% ash size filter paper and finally burnt the filtered materials at 850°C .

Emission Rate

Emission rate for the coal combustion was determined through mass distribution method (Eq. 1) as suggested by Luo *et al.* (2004) and Zhao and Luo (2018), while the emission rate for the sulfur was determined through Eq. (2) and integrated emission rate for different areas, source and time were determined through Eqs. (3) and (4), respectively (Hao *et al.*, 2002):

$$R = \frac{C_{\text{coal}} - C_{\text{ash}} \times C_{\text{fired}}}{C_{\text{coal}}} \times 100 \quad (1)$$

Fig. 1. Tectonic setting and samples location of the study areas.

$$Q_{i,j,t} = C_e \times CC \times R(1 - p) \quad (2)$$

$$Q_{i,j,t} = \sum_j Q_{i,j} \quad (3)$$

$$QT = \sum_t Q_{i,t} \quad (4)$$

In the above equations, R is the emission rate of the coal combustion, Q is the emission rate of the sulfur, C_{coal} , and C_{ash} are the sulfur concentration in coal and ash, respectively. C_{fired} is the total ash contents. While C_e is an element concentration, CC is the coal consumption (NBSC, 2017), and p is the efficiency of elements absorbed at control measure. Where i , j and t are the areas, source (coal, coal gangue etc.), and time (year), respectively.

RESULTS

Huanglong Coalfield

The magnitude of sulfur in the typical coal of Huanglong coalfield (Binxian Jurassic) of Shaanxi Province was interpreted in Table 1. The highest concentration of sulfur in the Binxian Jurassic coal gangue was 6.28%, and lowest was 0.56% with an average of 2.4%, which was higher than those reported by Wang *et al.* (2016) (1.4%) and Luo *et al.* (2005) (1.02%). The average concentration of sulfur

in the Chinese coal gangue was 1.01% (Ren *et al.*, 2006). Similarly, the average quantity of ash was 71.75%, which was equivalent to Wang *et al.* (2016) (70%). The average ash ratio is one-third after the combustion (Zhao *et al.*, 2008). In 2013, the total average production of coal gangue was 750 million tons (Mt), which exotically undergoes in spontaneous combustion and affect the environment badly (NDRCC, 2014; Wang *et al.*, 2016).

The concentration of sulfur in Binxian raw coal ranged 0.1–5.89 with an average of 2.85% (Table 1), which was higher than the Indian coal (0.4–0.6%) (Lu *et al.*, 2013), Weibei coalmines (0.94%), Shenfu coalfield (0.7%), Xishan coalfield (1.3%) and Pingyinan coalfield (1.4%) (Wang *et al.*, 2016). The Binxian Jurassic sulfur concentration was also higher than those reported by Zhao and Luo (2017) (1.52%) and Dai *et al.* (2012) (1.14%). The high consumption of coal released a huge amount of sulfur and other toxic gases, which affect the climatic condition, air quality, human health, land disturbance as well as affect the environmental sustainability (Han *et al.*, 2016; Zhao and Luo, 2017; Samset, 2018; Zhao and Luo, 2018). Similarly, the minimum and maximum concentration of sulfur in the coal slime were 0.61 and 1.45%, respectively, with an average of 0.92% (Table 1). The Binxian coal slime sulfur content was considerably lower than the Shaanxi (2.12%) (Zhao and Luo, 2018). The coal slime is similar to briquettes, which has been recommended by the government to use as a clean coal. These materials are generated during the coal cleaning

Table 1. Sulfur and ash quantity in the Huanglong coalfield (Binxian Jurassic coalmines).

Coal gangue			Raw coal		
sample	Sulfur %	Ash %	Sample	Sulfur %	Ash %
CG1	0.768	44.843	C1	2.186	17.771
CG2	3.158	75.787	C2	5.885	18.881
CG3	6.281	73.730	C3	2.062	16.183
CG4	1.337	63.008	C4	3.518	62.556
CG5	2.730	91.776	C5	1.228	82.614
CG6	5.576	59.041	C6	0.969	57.675
CG7	2.163	39.977	C7	5.860	17.604
CG8	0.699	90.060	C8	3.117	9.753
CG9	2.826	80.260	C9	3.534	16.170
CG10	3.620	75.798	C10	0.098	59.980
CG11	1.698	78.147	Avg.	2.846	35.919
CG12	2.065	70.352	Coal slime		
CG13	1.547	75.789	CM1	0.719	76.108
CG14-1	0.560	78.947	CM2	0.979	74.474
CG14-2	5.599	68.510	CM3	0.611	56.570
CG15	1.534	86.079	CM4	0.854	72.502
CG16	4.192	65.097	CM5	1.454	61.446
CG17	1.777	62.261	Avg.	0.924	68.220
CG18	1.628	82.595	S-Ash		
CG19	1.948	39.200		0.754	59.342
WCG1	1.561	78.878	GBW1111H-1		
WCG2	1.082	79.438		2.956	
CGF1	0.856	90.780	GBW1111H-2		
Avg.	2.400	71.755		3.066	

and washing. The domestic coals are mixed with coal slime, which has been used in the household (Li *et al.*, 2014). Nowadays, the coal slime is separated from the raw coal. The coal slime is using in the household for burning purpose as well as also recommended for the poor agriculture soil.

Weibei Coalfield

The aggregate and average concentration of sulfur and coal ash in the Hancheng Permo-Carboniferous and Langao Cambrian coalmines were given in Table 2 and Fig. 2. The concentration of sulfur in the Hancheng coal gangue ranged 0.28–7.37% with an average of 1.48%, which was equivalent to the Zhao and Luo (2017) (1.5%) and the Bindong coal (1.48%) (Wang *et al.*, 2016). The sulfur concentration of the Hancheng Permo-Carboniferous coal gangue was lower than the Yunnan coal wastes byproducts (CWBs) (1.9%) (Zhao and Luo, 2018), while higher than the Linyou (0.38%), Hancheng (0.9%), Shenfu (0.7%) and Xishan coalmines (1.2%) (Wang *et al.*, 2016).

The magnitude of sulfur in the Hancheng raw coal ranged 0.26–4.26% with an average of 2.4% (Table 2 and Fig. 2). The Hancheng raw coal sulfur contents were higher than the world coal (2%) (Ketris and Yudovich, 2009), Shenmu coalmine Shanxi (0.3%) (Hou *et al.*, 2018), Shanxi (0.8%) (Querol *et al.*, 2008), Anhui (1.32%) (Zhou *et al.*, 2014), Jiangsu (0.64%) (Li *et al.*, 2006), Liaoning (0.11%) (Wang *et al.*, 2016) and Yuegang *et al.* (2015) (1.14%).

The concentration of sulfur in the Permo-Carboniferous coal slime ranged 0.46–2.57% with an average of 1.5% (Table 2 and Fig. 2). The average sulfur contents of the

Permo-Carboniferous coal slime were lower than the world coal (2%) (Ketris and Yudovich, 2009) and the Brazil pyrite coal (3–7%) (Kalkreuth *et al.*, 2010), while higher than the earth crust (0.07%) (Wedepohl, 1995), Indonesian coal (< 1%) (Widodo *et al.*, 2010) and Wang study (1.01%) (Wang *et al.*, 2016). However, the coal slime sulfur contents were equivalent to those reported by Zhao and Luo (2017) (1.5%), Inner Mongolia coal (1.5%) (Chou, 2012) and the Brazil Permian coal (1.59%) (Kalkreuth *et al.*, 2006).

Langao Coalfield

The concentration of sulfur in the Langao Cambrian stone-like coal ranged 0.17–1.67% with an average of 0.84% (Table 2). The average concentration of the Langao sulfur was lower than the world coal (2%) (Ketris and Yudovich, 2009), Brazil pyrite coal (3–7%) (Kalkreuth *et al.*, 2010), Kirovskaya coalmine, Kazakhstan (3.1%) (Pak *et al.*, 2016). However, the Langao Cambrian coal sulfur contents were higher than the Shanxi coal (0.3%) (Hou *et al.*, 2018), Jiangsu coal (0.64%) (Li *et al.*, 2006), and Liaoning coal (0.11%) (Wang *et al.*, 2016). In Langao Cambrian, the contents of sulfur in the black shale rock ranged 0.34–6.45% with an average of 2.4% (Table 2). The Cambrian black shale has a higher concentration than the Permo-Carboniferous coal, while similar to or lower than the Binxian Jurassic coal. The Cambrian black shale sulfur content is significantly higher than the majority of the national and international studies (Kalkreuth *et al.*, 2006; Querol *et al.*, 2008; Ketris and Yudovich, 2009; Zhou *et al.*, 2014; Wang *et al.*, 2016; Zhao and Luo, 2018).

Table 2. Sulfur and ash contents in the Weibei coalfield (Hancheng Permo-Carboniferous coalmines) and the Langao County (Cambrian coalmines).

Weibei coalfield					
Coal gangue			Raw coal		
Samples	Sulfur %	Ash %	Samples	Sulfur %	Ash %
HC3	7.375	72.778	HC20	3.167	41.789
HC4	1.659	87.011	HC22	3.061	67.311
HC5	2.092	90.323	HC23	3.112	44.871
HC6	0.329	88.968	HC24	4.264	51.621
HC7	0.484	66.295	XS1	0.263	76.029
HC8	0.282	73.344	XS7	0.659	54.608
HC9	0.380	74.888	XS9	2.376	51.827
HC10	0.297	65.186	Avg.	2.415	55.437
HC11	0.282	81.911	Coal slime		
HC12	0.417	73.285	HC21	2.396	55.218
HC13	0.849	57.884	HC25	2.573	60.772
HC14	0.397	62.385	XS2	0.802	54.979
HC15	0.563	76.069	XS10	0.464	74.028
HC18	4.610	61.191	Avg.	1.559	61.249
HC28	2.316	69.606	Spontaneous Ash		
XS4	1.328	80.379	HC1	0.22	82.20922
XS5	1.866	78.696	HC2	6.64	41.77081
XS6	1.132	68.144	HC26	0.37	95.42511
Avg.	1.481	73.797	HC27	0.733541	94.2215
Fine-Ash	0.508		XS3	1.732002	72.18859
Course-Ash	0.485		XS11	0.32	55.53513
			Avg.	1.67	73.56
Langao County (Cambrian strata)					
Raw coal			Black shale rock		
ZHC1	1.355	76.897	ZHR1	3.058	85.230
ZHC2	1.077	77.718	ZHR2	0.848	91.324
ZHC3	1.193	73.056	ZHR3	2.051	84.319
ZHC4	1.674	65.770	ZHR4	3.628	79.663
ZHC5	0.757	77.119	ZHR5	6.460	81.129
ZHC6	0.566	83.018	ZHR6	0.694	69.242
ZHC7	0.174	87.444	ZHR7	0.336	93.537
ZHC8	0.375	90.229	Avg.	2.439	83.492
ZHC9	0.396	90.348			
Avg.	0.841	80.178	S-Ash	0.323	89.846

Fig. 2. The average concentration of sulfur in different type typical coalmines, Shaanxi.

The study explored that the Cambrian stone-like coal and the black shale have higher sulfur concentration as compared to the Jurassic and Permian coal. The mode of occurrence of sulfur in the Langao county is sulfide ore, i.e., pyrite, chalcopyrite, and molybdenite (Zhenmin *et al.*, 1997; Hussain and Luo, 2018). The higher sulfur contents in the coal are mostly associated with pyrite ($> 3.5\%$), while lower sulfur coal is associated with organic coal (Pak *et al.*, 2016).

DISCUSSIONS

The sustainability of atmospheric air quality is essential, however, the overuse of fuels (crude oil and coal) altered the air quality greatly (Wang *et al.*, 2018a). The primary air pollutants released by fuels/coals are SO_x , CO_x , NO_x , CH_4 , particulate matter, mercury, etc. This massive emission of gases alternatively affect the crops production,

water quality, the ozone layer, human health and accelerate the global warming (Saidur *et al.*, 2011). The average emission of sulfur to the atmosphere by some typical coalfield of China were given in Tables 3 and 4. Among the different sources, raw coal is the dominant source of sulfur followed by coal gangue and coal slime. A huge amount of coal and coal waste products are used in urban than in rural areas. In Binxian, the rural areas emitted 0.926×10 kilotons (kt) and urban regions emitted 0.15×10 kt of sulfur into the atmosphere, which counted about 1.4% of the total SO_2 emission (Fig. 3). Similarly, the emission of sulfur by Hancheng rural was 1.19×10 kt and urban 0.18×10 kt, respectively, which counted $\approx 1.83\%$ of the SO_2 emission into the atmosphere (Table 3). The total emission of sulfur to the atmosphere by the Langao regions was 0.32×10 kt (urban: 0.04×10 kt, rural: 0.28×10 kt). The estimated SO_2 released by the Langao Cambrian coal was 0.4% (Table 3). The emission of sulfur and SO_2 may affect

Table 3. Regional emission of sulfur to the environment (10 kt).

Areas	Specific	Coal Gangue	Raw coal	Coal slime	Total
Binxian County	<i>Residential</i>	0.137	0.922	0.020	1.078
	<i>Urban</i>	0.032	0.115	0.006	0.153
	<i>Rural</i>	0.105	0.807	0.014	0.926
	<i>Avg.</i>	0.091	0.615	0.013	0.719
	<i>SO₂ %</i>	0.182	1.230	0.026	1.438
Hancheng County	<i>Residential</i>	0.009	1.310	0.056	1.375
	<i>Urban</i>	0.002	0.164	0.017	0.183
	<i>Rural</i>	0.007	1.147	0.039	1.192
	<i>Avg.</i>	0.006	0.874	0.037	0.916
	<i>SO₂ %</i>	0.012	1.747	0.075	1.834
Langao County	<i>Residential</i>		0.320		0.320
	<i>Urban</i>		0.040		0.040
	<i>Rural</i>		0.280		0.280
	<i>Avg.</i>		0.213		0.213
	<i>SO₂ %</i>		0.427		0.427

Table 4. Sulfur contrition in the coal and coal wastes different regions.

Regions	Sulfur %	References
World Coal	2	(Ketris and Yudovich, 2009)
Earth crust	0.07	(Wedepohl, 1995)
Indonesian coal	< 1	(Widodo <i>et al.</i> , 2010)
Brazil Pyrite coal	3–7	(Kalkreuth <i>et al.</i> , 2010)
Brazil Permian coal	1.59	(Kalkreuth <i>et al.</i> , 2006)
Kazakhstan	3.1	(Pak <i>et al.</i> , 2016)
Shanxi coal	0.3	(Guo <i>et al.</i> , 2016)
Shaanxi	0.8	(Querol <i>et al.</i> , 2008)
Anhui	1.32	(Zhou <i>et al.</i> , 2014)
Jiangsu	0.6	(Li <i>et al.</i> , 2006)
North China	1.5	(Zhao and Luo, 2017)
Shaanxi	2.12	(Zhao and Luo, 2018)
Ordos basin, North China	1.48	(Wang <i>et al.</i> , 2016)
Huainan, Anhui	0.07	(Cai <i>et al.</i> , 2008)
Inner Mongolia	1.5	(Chou, 2012)
Beijing	0.12	(Li <i>et al.</i> , 2006)
China	1.0	(Yang and Schreifels, 2003)
China	1.14	(Dai <i>et al.</i> , 2012)

Fig. 3. Average estimated emission of sulfur and SO₂ to the atmosphere by typical coal of China.

the manufactured material, ecological system and the major cause of acid rain ((Eq. 5) explored how the sulfur cause acid rain) (Bhargava and Bhargava, 2013; Vela *et al.*, 2017):

The consumption of coal and coal wastes for the commercial and local purpose is increasing day by day. During coal extraction, a huge amount of coal wastes is generated (coal gangue and coal slime), which ultimately affect the agriculture land, forest, accelerated land degradation, generating coal dust, air and soil pollution (Hussain *et al.*, 2018). From 1991–2015, the coal wastes consumption is increasing, while alternatively decreasing the sulfur emission (NBSC, 2014, 2017; Zhao and Luo, 2018). In 1991, the total China coal waste by-products (CWBs) consumption in urban areas was 4% and in rural areas was 3%, while the raw coal consumption was 96% and 97%, respectively (Fig. 4). In 1997, the State Economic and Trading Commission (SETC) banned the small-scale power generation plants (50-megawatt [MW]), because of emitting a substantial amount of pollutants and generate less electricity. In 2000, SETC was further stringent and banned the 10,000 MW (total capacity) power plants. In this way, the total reduction of coal consumption and SO₂ emission was 10 Mt and 0.4 Mt, respectively (Yang and Schreifels, 2003). Similarly, in 2015 (latest data), the total coal consumption (in China) was reduced to 73% and 87% in urban and rural areas, respectively (Fig. 4). The net reduction of coal consumption from 1991 to 2015 was 23% and 10% in urban and rural China, respectively. However, the coal wastes consumption was increased by 23% in urban and 10% in rural (Fig. 4). The increasing of coal wastes consumption will reduce the gangue debris, air pollution, water contamination,

acid rain and SO₂ emission (Xu *et al.*, 2000; Yao *et al.*, 2016).

By adopting desulfurization policy, China reduced the coal-sulfur emission by 1.2% (1990), 1.17% (1991), 1.09% (1995), 1.05% (1999), and 1.0% (2000). The total change was observed up to 17% between 1990 and 2000 (Yang and Schreifels, 2003). In 2001–2005, the 10th five-year plan set a regulation to reduce the SO₂ emission up to the limit of 23.7 Mt. In 2000, SO₂ emission was 23.7 Mt, while in 2005 the SO₂ emission was 19.5 Mt controlled by potential abatement strategy (Yang and Schreifels, 2003; Lu *et al.*, 2010). In 2005, a significantly low amount of SO₂ was emitted by China, which was considerably lower emission than India and some other Asian countries (Fig. 5). The massive consumption of coal energy and installing the desulfurizing equipment in plants reduced the SO₂ emission up to the significant amount. In 2016, China controlled and reduced the massive emission of SO₂ as compared to 2005 (Fig. 5). The SO₂ reduction in China was mostly observed from 2006, while in other countries SO₂ was increasing, which could affect the atmospheric chemistry and sustainability (Kitayama *et al.*, 2008). By exploring the sulfur and SO₂ emission, the actual sulfur content in the Chinese coal and coal wastes is higher than the majority of international studies (Ren *et al.*, 2006; Ketris and Yudovich, 2009; Zhao and Luo, 2017; Rokni *et al.*, 2018; Wang *et al.*, 2018a; Zhao and Luo, 2018). However, the entire SO₂ emission by China was observed significantly lower than India (Fig. 5) as also reported by NASA (NASA, 2016). The mode of occurrence of sulfur in coal is barite, pyrite, and gypsum (Dinur *et al.*, 1980; Vuthaluru *et al.*, 2000; Ribeiro *et al.*, 2010; Chou, 2012). The high sulfur is associated with pyrite; rosenite, reomerite, jarosite, halotrichite, melanterite, while the low sulfur coal is associated with thenardite, iron sulfate and organic coal (Dinur *et al.*, 1980; Vuthaluru *et al.*, 2000).

By the rapid increase in population and industrialization,

Fig. 4. Consumption of coal and coal wastes in between in 1991 and 2015 in China (data taken from NBSC (2017)).

Fig. 5. Sulfur dioxide emission by China over the last decades (Image modified from NASA (2016)).

China demands more energy over the last decade. Due to lack of the natural gas and oil resources, China used more coal energy compared to another energy resource. According to NBSC, the total national emission of SO_2 in 1991 was 16.22 Mt; 1994, 18.25 Mt; 2000, 19.95 Mt; and finally reached to a peak position of 25.88 Mt in 2006 (Yang *et al.*, 2002; NBSC, 2017). By installing effective sulfur removing equipment, a significant amount of sulfur was reduced from 2006 and onward (Smith *et al.*, 2011) (Fig. 6). Similarly, the consumption of fossil fuels and the emission of SO_2 is increasing in developing countries especially in India (Garg *et al.*, 2002) (Fig. 5). The India coal has a

much lower sulfur content but due to ineffective SO_2 control technology, the emission rate is high (Lu *et al.*, 2011). In China, SO_2 emission both historic and projected was observed reducing (Fig. 6). The historic emission from 2007 to 2016 was 24.68 to ≈ 16.36 Mt, respectively (NBSC, 2017; Zhao and Luo, 2018), while the projection from 2017 to 2028 will be ≈ 16.26 to ≈ 8.0 Mt (Fig. 6). The study expects more reduction of SO_2 in the future than the projected because of using effective desulfurization technology in China. Shuhua *et al.* (2012) reported that if the desulfurization of gases and final emission reached to a 60%, a significantly less amount (0.6 Mt) of sulfur will be released to the atmosphere.

Fig. 6. Annual emission and projection of SO₂ by China (data generated NBSC from 1991 to 2016).

In China, the coal consumption is still too more than the other countries. In northeastern China, more than 70% of families using coal, in northwestern more than 68% of families using coal resource for heating, cooking and other purposes (Duan *et al.*, 2014). In most of the western part of China, a renewable and hydel-energy is using; therefore western China SO₂ emission is lower than eastern China (Fig. 7). In 2015, the highest SO₂ emission was observed in the Shandong Province (1.53 Mt) followed by Inner Mongolia (1.23 Mt) and the Henan (0.032 Mt), Tibet (0.0054 Mt) being lowest one (Fig. 7). However, China also using gas energy (methane, liquid petroleum gas [LPG], etc.). In 2015 (latest data), China using man-made coal gas 108,306 10K m³, natural gas 2,080,061 10K m³, and LPG 5,871,062 tons (Zhao and Luo, 2018).

The study clearly explored that the use of coal and other fossil fuel is increasing continuously, while alternatively release a huge amount of greenhouse gases and SO₂. Most of the countries thrive on their economy and disturbing the natural environment badly. However, China is one of the most responsible country to boost their economy by following green economy practices. Almost 70% of the natural sulfur is bonded with pyrite coal and some other minerals. By the coal beneficiation, approximately 25% of the total sulfur and 50% pyritic-sulfur can be removed (World Bank, 1998). The historical emission and future estimation of SO₂ are important to improve the fossil fuels quality and consumption. Besides, it could be helpful to search the alternative source of clean energy that might sustain our natural environment, air quality, and human health.

Fig. 7. Annual regional sulfur emission to the atmosphere by China in 2015 modified from (NBSC, 2017; Zhao and Luo, 2018).

CONCLUSION

The typical coal of Shaanxi, China, emits a large amount of sulfur and SO₂ into the atmosphere. The average sulfur content was higher in the Permo-Carboniferous (Weibei coalfield) raw coal (2.4%) and lower in the coal gangue (1.48%) and coal slime (1.5%) than in coal worldwide (2%), whereas it was higher in the Jurassic (Huanglong coalfield) raw coal (2.85%) and coal gangue (2.4%) and lower in the coal slime (0.92) than in coal worldwide. The sulfur content in the Cambrian stone-like coal and black shale was 0.84% and 2.4%, respectively. The amount of SO₂ released by the Binxian Jurassic, Hancheng Permo-Carboniferous, and Langao Cambrian coal was 1.4%, 1.8%, and 0.4%, respectively, of the total SO₂ emitted into the atmosphere.

The average consumption of coal waste in 1991 was 4% and 3% in urban and rural areas, respectively. In 2015, the consumption of coal waste and coal briquettes was 15% and 12%, respectively, in urban areas (27%), and 8% and 7%, respectively, in rural areas (15%). The high consumption reduced the debris from coal waste in an environmentally friendly way. The highest SO₂ emissions from China were recorded in 2006 (25.88 Mt), after which they continuously declined, resulting in 16.36 Mt of observed emissions in 2016. Hence, we expect to reduce the amount of emitted SO₂ below the projected values (16.26 Mt and ~8.0 Mt for 2017 and 2028, respectively) due to the prompt installation of desulfurizing equipment. A ~63% reduction in SO₂ has been observed in China between 2006 and now. The adoption of a green economy and commercially feasible technology will further decrease the level of SO₂ and other pollutants. The current study recommend using the desulfurized natural gas or LPG, which is economically viable and socially acceptable, as an alternative to coal.

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COMPLIANCE WITH ETHICAL STANDARDS

The authors declared no conflict of interest and both the authors contributed equally in the respective research work.

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