



Emission Factors of NO_x, SO₂, PM and VOCs in Pharmaceuticals, Brick and Food Industries in Shanxi, China

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ABSTRACT

The acquisition of accurate emission factors (EFs) of pollutants is an inevitable step to the establishment of emission inventories for development of pollution control policies. The current studies were focused on large-scale industries (LSIs) although tremendous pollutants emitted from the small-scale industries (SSIs) with small coal-fired boilers (SCFBs) ascribe to the deficiency of pollutant removal facilities (RFs). A systematic field sampling and measurements conducted in 51 enterprises involving production of pharmaceuticals, brick and food to obtain the EFs of SO₂, NO_x, PM, and VOCs (SNPV) associated with coal consumption (EF_I), industrial output (EF_{II}), and product yield (EF_{III}). Among them, PM-RFs were all equipped except for 3 brick factories, no NO_x- and VOCs-RFs were installed, and SO₂-RFs were installed in part. Obvious fluctuations existed in EF_I and EF_{II} values among 51 companies owing to the differences of pollutant removal efficiencies, coal compositions, annual outputs, production processes, and products. Co-burning of coal and coal gangue (raw material) in brick production weakened the correlation between sulfur contents in coal and SO₂ EF_I values. The using of organic solvents in drug making process promoted the emission of VOCs. SO₂ EFs in factories with RFs were much lower than those factories without RFs. SO₂ EFs dominated over those of PM and NO_x among three kinds of enterprises, especially in brick companies. For EF_I (in kg t⁻¹), food industry possessed highest value for SO₂, PM, and NO_x, while the maximum value for VOCs occurred at pharmaceuticals industry. Due to the low output values of brick companies, their SNPV possessed the highest EF_{II} compared to the other two kinds of factories. NO_x EFs experienced lessen fluctuations than other pollutants among all the factories due to the different formation mechanism and no installation of NO_x RFs. EF_{III} showed various fluctuations due to the different product types.

Keywords: Emission factor; NO_x; SO_x; VOCs; Pharmaceutical industry; Brick industry.

INTRODUCTION

In recent years, serious atmospheric quality deterioration had occurred in cities of Asian, European and North American, especially in rapidly developing China (Fang *et al.*, 2009; Pascal *et al.*, 2013; Li *et al.*, 2017). At present, the common air pollutants in China are SO₂, NO_x and PM emitted from various factories, which have caused great harm to the environment and human health (Delmelle *et al.*, 2002; Srivastava *et al.*, 2005; Lepeule *et al.*, 2012; Tam *et al.*, 2016; Guttikunda and Jawahar, 2018; Hu *et al.*, 2018;

Zhao *et al.*, 2018; Zhai *et al.*, 2019).

PM, especially PM_{2.5} should be responsible for Chinese 1.1 million excess deaths in 2015 (Zhai *et al.*, 2019). The sulphate and nitrate originated from their precursors of SO₂ and NO_x were main contributors to PM_{2.5} (Yao *et al.*, 2014). PM has the potential effects to human health due to their deposition capabilities in the body. Besides, there are many evidences for the impacts of PM on the human respiratory system (Crouse *et al.*, 2012; Lepeule *et al.*, 2012). The increasing SO₂ emission will lead to an increase in the mortality risk of 1.01% through the respiratory system (Hussain and Luo., 2018). Acid rain caused by sulfate acid deposition can be detrimental to ecosystems, plants and animals, both aquatic and terrestrial (Cronin *et al.*, 2002; Tam *et al.*, 2016). NO_x can promote the formation of photo-chemical smog, visibility reduction, and acid rain, and ozone depletion. As the main compositions of NO_x,

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NO and NO₂ seriously injured people by invading the bronchioles and alveoli in the deep part of the respiratory tract (Yang, 2017). VOCs in ambient air are an increasing concern because many of them have been identified to be human carcinogens and precursors of both secondary organic aerosols and O₃ (Zhang *et al.*, 2015; Hu *et al.*, 2018).

Along with China's economic development, emissions of PM, NO_x, SO₂, and VOCs in developed areas have increased rapidly. The acquisition of detailed source emission inventories rely on accurate EFs data, which is premise for development of control policies of these pollutants (Li *et al.*, 2018). Prior researches on emission factors mainly focused on LSIs, such as coal-fired power generation, cement production, coke making, and iron and steel production (Guo *et al.*, 2017; Li *et al.*, 2018; Ni *et al.*, 2018). But the SSIs should be paid more attentions because of tremendous emissions owing to the lack of advanced technology and facilities for pollutants removal (Li *et al.*, 2018).

Coal burning associated with SSIs is main source of air pollutants with its emissions is depend on pollutant removal efficiencies, coal quality, and the amount of coal combustion (Li *et al.*, 2018). The SSIs related to coal burning including pharmaceuticals production, food processing, and brick making should be more concerned.

A large amount of heat was need during the synthetic and fermentation processes, the mostly common steps for pharmaceuticals production. In pharmaceuticals companies, SNP originated mainly from coal combustion, while VOCs was also formed from evaporation of organic solvent used in extraction and solvent recovery processes (Guo *et al.*, 2014).

Food processing industry is the pillar industry of the national economy and the basic industry to guarantee people's livelihood (Ma *et al.*, 2019). The food industries use SCFBs for heating with drastic pollutants released due to the lack of RFs. VOCs with strong odors were also released from food processing, which imposed seriously adverse effects on surrounding environment and human health (Xi *et al.*, 2014; Li *et al.*, 2017a). Previous researches were mainly focused on the smell of the food and cooking fumes, while fewer researches were conducted on the VOCs from SCFBs applied in food processing (Liu *et al.*, 2016; Li *et al.*, 2017b; Xu *et al.*, 2017; Gao *et al.*, 2018).

As a developing country, China has a well-developed construction industry and a strong demand for bricks, which yielded abundant air pollutants during coal combustion for production and pose threat to environment (Toledo *et al.*, 2004; Kan *et al.*, 2012; Huang *et al.*, 2014; Wang *et al.*, 2014; Kadir *et al.*, 2015). There are approximately 80,000 brick factories in China, 90% of which are the traditional type and characterized as utility of low-quality coal and lack of lack of pollutant RFs (General Administration of Quality Supervision, 2013). However, the emission inventories of brick production in China counted on the EFs of other countries to a great extent, regardless of scarce reported data related to NO_x and PM released from brick factories in China (Zhao *et al.*, 2013).

In a word, the EFs associated systematically researches

related to coal combustion in SSIs should be paid more attentions. To our knowledge, fewer data related to pollutant EFs were available in China for SCFBs used in SSIs such as pharmaceuticals production, food processing, and brick making. The establishment of pollutant inventories for these enterprises relied on the foreign data, while considerable bias occurred due to the significantly regional discrepancies in coal quality and combustion mode, production process, and pollutant RFs (Huang *et al.*, 2014; Wang *et al.*, 2014). Greater uncertainties existed in the establishment of emission inventories due to the absence of detailed EF values (Yue *et al.*, 2018). Localization and particularization of EFs related to these enterprises were urgently needed in China (Horák *et al.*, 2018).

In this study, a total of 51 related enterprises about brick making, pharmaceuticals manufacturing, food processing were investigated for annual output, product yield, and coal quality. Also the field measurements of SO₂, PM, and NO_x, and VOCs sampling and subsequently instrumental analysis were conducted. All of these works aimed to achieve following purposes: 1) Acquisition of localized EFs associated with SO₂, PM, NO_x, and VOCs for these enterprises; 2) Improvement of practicability by the output, product yields, and coal consumptions associated EFs; 3) Refinement of EFs based on specific products.

METHODOLOGY

Investigation of Related Enterprises

In this study, 51 companies in Shanxi Province including 34 for brick making, 8 for pharmaceuticals manufacturing, and 9 for food processing were on-site investigated for their production and coal related information. Meanwhile, SO₂, PM, and NO_x were real-time monitored using a flue gas sampler, while VOCs were collected and subsequent analyzed by a GC-MS system.

The information about product types and yields, raw materials, output, and coal compositions were detailed described in Tables 1, 2, and 3. The abbreviations used in this study including RE, CC, and MY were removal efficiency, coal consumptions and millionaire yuan (RMB), respectively. It should be noted that all of factories were not equipped with NO_x and VOCs RFs, while PM RFs were all installed for all of them except for 3 brick factories. The SO₂ RFs were erected for 3 of 8 pharmaceuticals factories, 3 of 9 food plants, and 13 of 34 brick enterprises. PM RE values showed not the correlation with annual output of enterprises, which suggested more advanced PM removal technologies were not always adopted by enterprises with high output. The sulfur contents contained in coal were 0.456 ± 0.118 for pharmaceuticals, $0.792 \pm 0.444\%$ for food, and $0.661 \pm 0.817\%$ for brick, the highest corresponding value (2.5%) occurred at brick factories of 32, 33, and 34. The main products of pharmaceuticals plants were vaccine, injection, pills, and sterile powder. Coal gangue, coal burning fly ash, and clay were used as raw materials in 31, 1, and 2 of 34 brick factories and the outputs were ranged from 0.150 to 69.41 MY. The main products vinegar, steamed bread slices, bean products, sour milk, and liquor were contained

Table 1. Statistics about coal components, output, and yield for pharmaceuticals plants.

	S _{ad} (%)	A _{ad} (%)	CC (t a ⁻¹)	SO ₂ RE (%)	PM RE (%)	Output (MY)	Product	Yield (t a ⁻¹)
P1	0.70	20.0	400	0	93.0	60.0	Vaccine	200
P2	0.30	6.00	5400	19.8	88.0	1009	Injection	130
P3	0.45	17.0	950	0	77.0	29.8	Pill	31
P4	0.35	17.8	6790	27.9	77.0	150	Oral liquid	330
P5	0.45	17.0	150	0	99.0	5.20	Injection	3.5
P6	0.45	17.0	300	0	95.0	10.0	Sterile powder	12
P7	0.45	17.0	800	0	95.0	26.7	Injection	60
P8	0.50	15.0	3920	61.1	87.7	1963	Injection	8.8

Table 2. Statistics about coal compositions, output, and yield for food processing.

	S _{ad} (%)	A _{ad} (%)	CC (t a ⁻¹)	SO ₂ RE (%)	PM RE (%)	Output (MY)	Product	Yield (t a ⁻¹)
F1	0.35	17	393	0	73.5	23.3	White spirit	579
F2	0.35	17	120	0	95.0	3.00	Steamed bread slices	320
F3	0.45	17	589	0	99.0	8.91	Bean	1563
F4	0.98	16	620	0	70.0	383	Sour milk beverage	15000
F5	1	20	1360	0	87.0	21.9	Sour milk beverage	9860
F6	0.5	15	1428	0	90.0	5.40	Vinegar	10000
F7	0.6	21	1182	71.4	67.0	260	Meat	2400
F8	1.4	17	379	85.0	79.0	251	Feed	85535
F9	1.5	17	1100	70.0	95.0	130	Feed	50000

in food factories, which possessed largely variable outputs from 3.00 to 383 MY.

Sampling and Measurement of SO₂, NO_x, and PM

The pollutants such as gaseous SO₂, NO_x and CO, and PM originated from all the enterprises were analyzed by a flue gas analyzer (Laoying-3012H, Qingdao LaoYing Environmental Science and Technology, Co., Ltd.) installed at outlet of flue gas after pollutant RFs aimed to investigate the pollutants actually discharged into atmosphere. In addition, the extra information related to temperature and flow velocity (m s⁻¹) of flue gas was also provided. A pilot tube was used to the measurement of flue gas velocity and collection of PM. PM mass concentration was calculated as PM mass divided by the volume of sampled flue gas. The gas analyzers were calibrated with zero gas and targeted by standard gases (NO_x, SO₂ and O₂) before the test day to eliminate the possible interferences. The pollutant mass emissions were obtained using pollutant concentrations multiplied by volume of flue gas.

The proximate and ultimate analysis of coal was carried on for access to the information about contents of sulfur, carbon, hydrogen, nitrogen, oxygen, and water. All the analysis processes subordinated to the Chinese standard methods of GB/T 212-2008 and GB/T 476-2001. The same calculation methods of EFs described by Li *et al.* (2018) were adopted in this study.

Calculation of Emission Factors Associated with Coal Consumption, Output, and Product

In order to enhance the practical purpose, EFs were provided based on coal consumptions (EF_I reported in kg t⁻¹), annual output (EF_{II} reported in kg MY⁻¹), and product (reported in kg t⁻¹). All the EF forms were calculated using

Eqs. (1)–(3).

$$EF_I (\text{kg t}^{-1}) = C (\text{kg m}^{-3}) \times V_{FA} (\text{m}^3 \text{kg}^{-1}) \times 1000 (\text{kg t}^{-1}) \quad (1)$$

where C was mass concentrations of pollutants in flue gas and V_{FA} is the actual flue gas volume originated from combustion of 1 kg coal.

$$EF_{II} (\text{kg MY}^{-1}) = (C (\text{kg m}^{-3}) \times 1000 (\text{kg t}^{-1}) \times V_{FA} (\text{m}^3 \text{kg}^{-1}) \times \text{Coal consumption (t a}^{-1})) / (\text{Output (MY a}^{-1})) \quad (2)$$

$$EF_{III} = (C (\text{kg m}^{-3}) \times 1000 (\text{kg t}^{-1}) \times V_{FA} (\text{m}^3 \text{kg}^{-1}) \times \text{Coal consumption (t a}^{-1})) / (\text{Product yeild (t a}^{-1})) \quad (3)$$

V_{FA} was derived from the theoretical air amount required by 1 kg of coal combustion (V_{AT}) calculated by Eq. (4).

$$V_{AT} = 0.0889\omega(C_{ar}) + 0.2567\omega(H_{ar}) + 0.0333\omega(S_{ar}) + 0.0762\omega(N_{ar}) - 0.0333\omega(O_{ar}) \quad (4)$$

V_{AT} is the air amount needed for complete burning of 1 kg coal (m³ kg⁻¹), elements C, H, O, N, and S marked by “ar” in the lower right corner indicate the contents of corresponding element in coal on the received basis.

The theoretical flue gas volume generated from burning of 1 kg of coal was calculated using Eq. (5).

$$V_{FT} = V_{CO_2} + V_{SO_2} + V_{NO_2} + V_{N_2} + V_{H_2O} \quad (5)$$

$$V_{CO_2} + V_{SO_2} + V_{NO_2} = 0.01867\omega(C_{ar}) + 0.007\omega(S_{ar}) + 0.0016\omega(N_{ar}) \quad (6)$$

where V_{CO₂}, V_{SO₂}, and V_{NO₂} refer to volumes of CO₂, SO₂,

Table 3. Statistics about coal compositions, output, and yield of brick factories.

	S _{ad} (%)	A _{ad} (%)	CC (t a ⁻¹)	SO ₂ RE (%)	PM RE (%)	Output (MY)	Raw material	Yield (t a ⁻¹)
B1	0.36	17	147000	0	99	2.69	Gangue	92100
B2	0.3	13	1400	0	83	0.16	Gangue	2000
B3	0.3	16.5	7000	0	98	0.98	Gangue	12500
B4	0.3	17	54000	0	83	1.35	Gangue	33750
B5	0.3	17	38000	0	83	2.40	Gangue	30000
B6	0.3	17	3500	0	85	0.30	Gangue	5000
B7	0.3	16.5	1221	0	95	3.80	Coal fly ash	66787
B8	0.3	17	1200	0	83	0.15	Gangue	1500
B9	0.34	13.3	9400	88.9	99	9.02	Gangue	35000
B10	0.38	27	56000	84.0	99	24.0	Gangue	42500
B11	0.25	27	18000	0	95	19.0	Gangue	27500
B12	0.25	27	18000	77.3	95	13.0	Gangue	50000
B13	0.25	27	10100.6	0	99	7.00	Gangue	18750
B14	0.25	27	3000	30.0	99	13.0	Gangue	45000
B15	0.25	27	25366	82.4	99	17.0	Gangue	40000
B16	0.25	27	22000	0	99	15.0	Gangue	40000
B17	0.25	17	10000	0	99	9.90	Gangue	37500
B18	0.25	27	22000	0	99	14.0	Clay	40000
B19	0.2	28	31752	0	90	3.36	Gangue	43500
B20	0.2	30.4	15190	0	0	3.00	Gangue	37500
B21	0.62	25	17500	0	0	3.00	Gangue	25000
B22	0.3	13.2	126000	32.5	99.9	8.00	Gangue	150000
B23	0.88	13.8	56000	48.1	41	6.48	Gangue	150000
B24	3.5	13.1	10000	0	97	7.11	Gangue	17500
B25	0.3	14	70802	0	0	69.4	Gangue	17808
B26	1.3	13	16000	57.0	99.6	9.20	Gangue	15000
B27	0.5	17	1000	50.0	75	0.50	Clay	2500
B28	0.5	17	10000	70.5	99	5.00	Gangue	14750
B29	0.5	17	16000	50.0	99	6.60	Gangue	40000
B30	0.5	17	20000	50.0	95	8.10	Gangue	50000
B31	0.5	17	4400	51.1	85	1.90	Gangue	10750
B32	2.5	30	15000	0	85	7.50	Gangue	31250
B33	2.5	30	16000	0	85	7.50	Gangue	45000
B34	2.5	30	35280	0	85	15.1	Gangue	70000

and NO₂ derived from burning of coal containing carbon, sulfur, and nitrogen, V_{N₂} is the N₂ volume in theoretical air volume (V_{AT}) and calculated as 0.79V_{AT}, and V_{H₂O} is the water vapor volume originated from combustion of hydrogen in coal (0.112ω(H_{ar})), vaporization of water in coal (0.00124ω(M_{ar})), and vapor in air (0.0161V_{AT}).

Finally, the actual flue gas generated from 1 kg of coal burning was calculated by Eq. (7).

$$V_{FA} = V_{FT} + (\alpha - 1)V_{AT} + 0.0161(\alpha - 1)V_{AT} \quad (7)$$

α is the excess air coefficient and provided by corresponding enterprises.

VOCs Sampling and Analysis

VOCs sampling and analysis procedure was conducted according to standard method designated by Chinese Ministry of Environmental Protection (HJ 734-2014) for VOCs in flue gas from stationary sources. The whole procedure was simply divided into three steps, adsorption by a stainless pipe for TD-100 system (Markes International,

UK), thermal desorption by TD-100, and measurement by a HP6890 GC/5973i MS system. The sampling flow-sheet was shown in Fig. S1. A adsorption pipe placed behind the sampling gun (volume 2 L and at a flow rate of 40 mL min⁻¹) was used to collected VOCs in flue gas. An ice bath impact bottle placed before adsorption pipe to eliminate the water interference. The adsorption pipes pumped into a cryofocus trap (-40°C) to concentrate VOCs and rapidly h desorbed by a TD-100 system, and then transferred to GC-MS system equipped with a DB-1 capillary column (60 m × 0.32 mm × 1.0 mm, Agilent Technologies, USA). The GC oven temperature was set at 35°C, increased to 140°C at 6°C min⁻¹, and then ramped to 220°C at 15°C min⁻¹ and held for 3 min. The calibration standards were prepared by diluting 100 ppbv of PAMS (Photochemical Assessment Monitoring Station, USA) certified gas to 0.5, 1, 5, 15, 50, and 100 ppbv (Yan *et al.*, 2016; Widiana *et al.*, 2017). A sum of 57 VOC species identified and measured in this study was used to represent the total VOCs.

For quality assurance and quality control, all the samples were analyzed within 24 h, and a series of experiments

including penetration of adsorption pipe and field blank were conducted every 8 samples. The sampling pipes were heated in highly pure N_2 gas (99.999%) at 350°C for 15 min with a flow rate of 40 mL min^{-1} for activation prior to sampling. 1,4-difluorobenzene and chlorobenzene-d5 were selected as internal standards in samples. A reference standard (1 ppbv) was injected before every testing day to ensure the instrument stability. The method detection limits (MDLs) (reported in $\mu\text{g m}^{-3}$) for 57 species ranged from 0.1 of C_6H_{14} (CAS: 75–83–2) to 1 of C_3H_6O (CAS: 67–64–1) with the mean value as 0.45 ± 0.19 (Table 1S).

RESULT AND DISCUSSION

Emission of Air Pollutants from Pharmaceutical Industries

Although much lower amount of coal linked with heating and concentrating of drug production were consumed compared to coal burning boilers used in LSIs, huge quantities of air pollutants discharged from them due to the lack of pollutant RFs. The information about ash content and sulfur content, and coal consumptions were listed in Table 1. The coal consumptions (reported in t a^{-1}) of 8 pharmaceuticals plants ranged from 150 to 6790 with a mean value as 2339, and showed a weak correlation with their output ($R^2 = 0.13$, $p < 0.05$), indicated the influence

of drug prices. Generally the SO_2 RFs occurred in factories with high output values such as P2, P4, and P8, while NO_x and VOCs RFs were not equipped among all the 8 factories. As a contrast, all the factories installed PM RFs with the PM RE ranged from 77% to 99%. The emission amounts increased along with the increasing of coal consumptions. Figs. 1 and 2, and Tables S2–S5 listed three types of EFs for each pharmaceuticals factory and mean value of EFs for 8 factories.

The key influencing factors on the SO_2 emissions were sulfur content of coal and removal efficiency of SO_2 (Li et al., 2018). The drastic fluctuations of EF_I and EF_{II} for SO_2 occurred among 8 plants and the weak correlation was found between EF_I and EF_{II} ($R^2 = 0.12$, $p < 0.05$), which reflect the greater impact of product prices of different factories. The EF_I values ranged from 1.91 to 11.2 kg t^{-1} (mean: 5.86 ± 2.95), and they were $5.01\text{--}231\text{ kg MY}^{-1}$ (mean: 130 ± 88.3) for EF_{II} (Figs. 1 and 2). The highest SO_2 EF_I of 11.2 kg t^{-1} occurred at P1, while the lowest value of 1.91 kg t^{-1} was attributed to P4. The high S_{ad} (0.70%) of coal and the deficiency of SO_2 RFs for P1 were explanations. For SO_2 EF_{II} values, the highest and lowest EF_{II} were owned by P3 (231 kg MY^{-1}) and P8 (5.01 kg MY^{-1}), respectively.

The EFs of SO_2 for 8 plants could be identified as two categories based on with or without SO_2 RFs. Class I

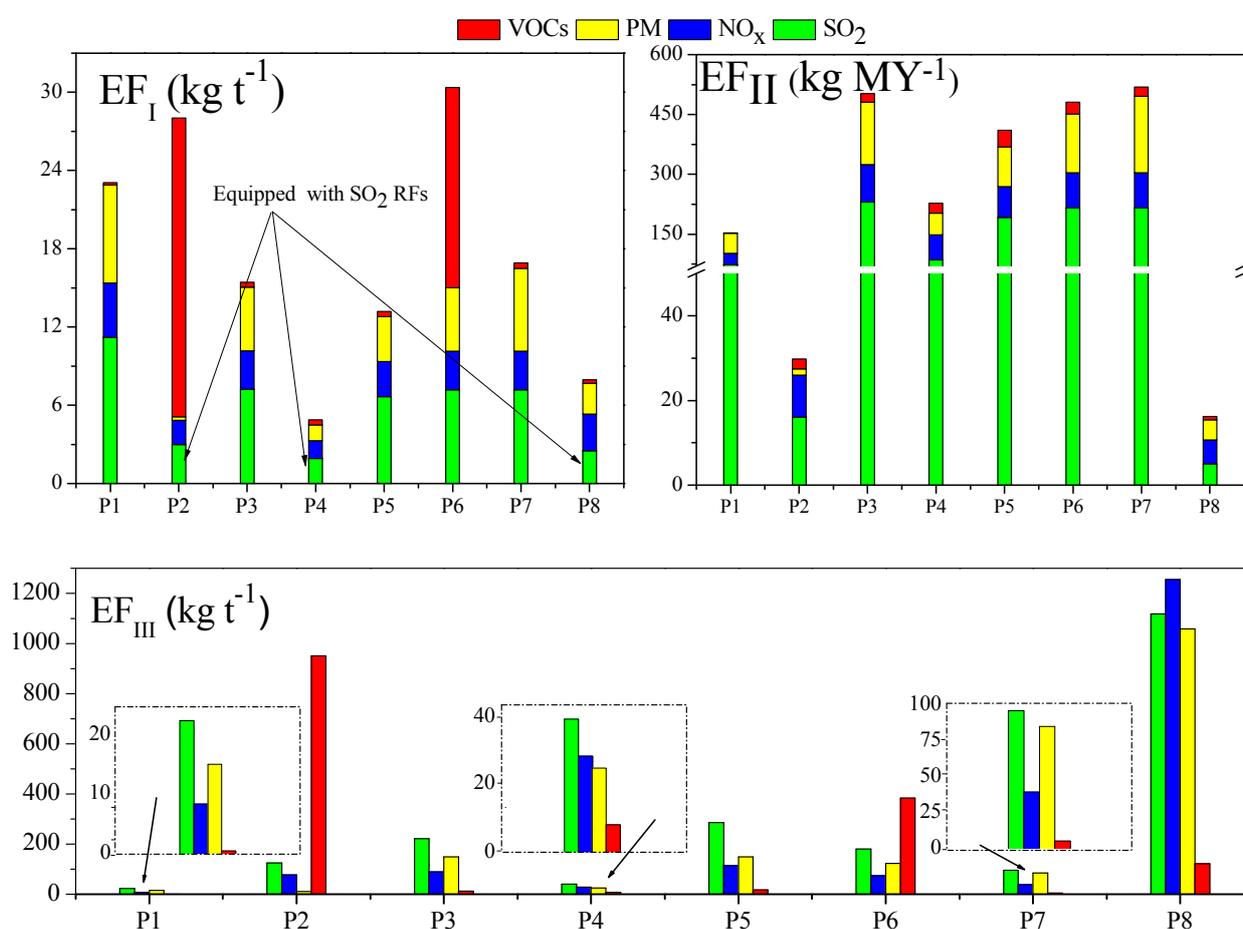


Fig. 1. Statistical values of EF_I , EF_{II} , and EF_{III} for 8 pharmaceuticals making factories.

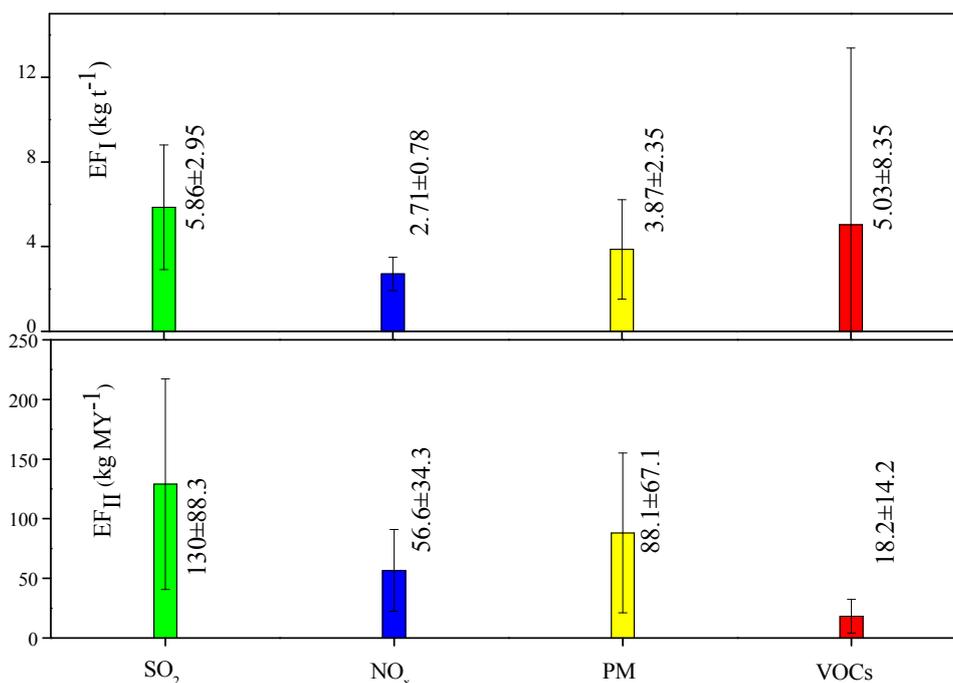


Fig. 2. Mean values of pollutant EF_I for 8 pharmaceuticals factories.

including P2, P4, and P8 had a low EF_I value (3.01, 1.91 and 2.51 kg t⁻¹) with installed SO₂ RFs and high outputs, while high EF_I values (6.67 to 11.2 kg t⁻¹) were possessed by Class II (P1, P3, P5, P6, and P7) with low output values (Table 1). The EF_I (7.90 kg t⁻¹) and EF_{II} (35.8 kg MY⁻¹) of SO₂ for Class II were 3.20 and 5.20 times of corresponding values of Class I. EF_I values were well correlated with sulfur contents of coal among 5 plants in Class II ($R^2 = 0.92$), while the weak correlation was found among 8 factories ($R^2 = 0.52$), indicating more strong influence of SO₂ RE than sulfur content. The 3 factories within Class I possessed much lower EF_{II} of 35.8 kg MY⁻¹ than mean value of 186 kg MY⁻¹ of 5 plants in Class II. EF_{II} was more affected by product price than coal compositions.

There was little fluctuation for NO_x EF_I values among the 8 plants compared to corresponding values for SO₂, from 1.38 of P4 to 4.18 kg t⁻¹ of P1 with the average value as 2.72 kg t⁻¹. No NO_x removal equipment among all the 8 factories should be underlined. The highest NO_x EF_I occurred at P1 with the coal consumption as 400 t a⁻¹. Drastic emissions of fuel-NO_x related to burning of high nitrogen content of coal and thermal NO_x associated with high burning temperature in P1 was impetus for the highest NO_x EF_I (Löffler *et al.*, 2005). EF_{II} values (reported in kg MY⁻¹) of NO_x varied from 5.63 of P8 to 93.8 of P3 with the mean value as 56.6 (Fig. 1 and 2). Low NO_x EF_{II} values were found for P2 and P8 as 9.96 kg MY⁻¹ and 5.63 kg MY⁻¹, respectively, owing to the high price of products from two factories. The rest 6 factories possessed high NO_x EF_{II} values from 27.83 to 93.7 kg MY⁻¹. It should be noted that NO_x emission was less influenced by coal quality than SO₂ emission due to the different formation mechanisms between NO_x and SO₂.

In regard to PM emissions, the EF_I of the 8 factories

were in the range of 0.26–7.5 kg t⁻¹, with an average of 3.87 kg t⁻¹ (Fig. 2). Among 8 factories, 4 ones possessed higher PM EF_I than 3.87 kg t⁻¹, and the highest PM EF_I (7.5 kg t⁻¹) appeared at P1 ascribe to the highest ash content (20%) (Table 1). The coal with medium ash content based on A_d (ash content on dry basis) was used in these 8 plants, and the highest A_d of 20% in P1 resulted in the highest EF_I. P2 owned the lowest PM EF_I value resulted from low A_d (6%) of coal. In a word, The PM EF_I was mainly subjected to ash content of coal and PM RE of RFs. The PM EF_{II} values exhibited a similar trend with NO_x EF_{II}, P2 and P8 owned low values of 1.40 and 4.75 kg MY⁻¹, while they varied from 50 to 191.3 kg MY⁻¹ with a mean as 109 kg MY⁻¹. The product price and coal quality were main influencing factors on PM EF_{II}. So the application of ash removal technologies prior to coal burning was a key factor to reduce the PM emissions.

For the VOCs emissions, P2 and P6 owned much higher EF_I values as 22.9 and 15.3 kg t⁻¹ than 0.18–0.40 kg t⁻¹ of remaining 6 factories. A large amount of organic solvents application in two plants and roasting of Chinese medicinal herbs would be contributors to VOCs. In addition, the V_{ad} (volatile contents on air dried basis) values were similar among all the 8 factories and ranged from 28.5% to 31.1%, which further verified the high contributions of organic solvents and related production processes. P1, P2, and P8 exhibited lower EF_{II} levels (0.90, 2.39 and 0.88 kg MY⁻¹) compared to the corresponding values (21.85–41.54 kg MY⁻¹) of the rest 5 factories, which implied the stronger influence of product price and output than nitrogen content and production processes on VOCs EF_{II} values.

In regard to EF_{III} values of SNPV for these 8 factories, the average of the 3 factories which equipped SO₂ RFs was 86.6 ± 35.5 kg t⁻¹, it was much lower than those 5 factories

with the average as $161 \pm 92.7 \text{ kg t}^{-1}$. Due to the different drug production processes, higher fluctuation was showed in NO_x , PM and VOCs values which ranged from 8.35 to 1256 kg t^{-1} , $10.9\text{--}1059 \text{ kg t}^{-1}$ and $0.36\text{--}951 \text{ kg t}^{-1}$ with the average as 396 ± 211 , 328 ± 202 and $313 \pm 187 \text{ kg t}^{-1}$. In addition, P1 and P5 produced veterinary drugs, P2, P3, and P4 made Chinese patent medicines, while P6, P7, and P8 intended to make western medicines by chemical method.

In addition, three types were also identified based on products as Class I for veterinary drug production (P1 and P5), Class II for production of (P2, P3, and P4), and Class III for making of western medicines by chemical method (P6, P7, and P8). Due to difference of measurement unit of products among different enterprises, it was difficult to conduct a comparing analysis of each other. EF_I and EF_{II} values showed not the obvious differences among 8 factories with different target products.

Emission of Air Pollutants from Food Industries

In this study, coal compositions and pollutant removal efficiencies of SCFBs applied in 9 food factories, and their outputs and products were investigated to obtain the EFs. Although only small amount of coal consumed for heating in 9 food industries, the pollutants produced could not be

ignored owing to the pollutant removal equipments behind the times and inferior coal. The coal consumptions (reported in t a^{-1}) of 9 food factories ranged from 120 to 1428 with a mean value as 796, lower than pharmaceutical factories and much lower than brick plants, which showed a weak correlation with the industrial output ($R^2 = 0.14$) due to the influence of product price. 3 of 4 factories with high output values were equipped with SO_2 RFs such as P7, P8, and P9, while the rest F4 without SO_2 RFs installed regardless of its highest output of 383 MY. The PM RFs installed in all the 9 plants with the PM RE ranged from 67% to 99%, while all the factories were not possessed the NO_x and VOCs RFs. Fig. 3, and Table S6–S9 listed three types of EFs for each food factory.

The Pearson's correlation coefficient between sulfur content and SO_2 EF_I values for 6 factories without SO_2 RFs was 0.98, indicated that sulfur content had a great influence on SO_2 EF_I . The 3 factories owning SO_2 RFs possessed much lower SO_2 EF_I ($6.94 \pm 2.51 \text{ kg t}^{-1}$) than that ($10.5 \pm 4.05 \text{ kg t}^{-1}$) of the other factories. The highest EF_I occurred in F5, as high as 16.0 kg t^{-1} , might be related to the high sulfur content in coal (1%). The average EF_I of 9.31 kg t^{-1} for food processing plants was higher than corresponding value of 5.86 kg t^{-1} , which was attributed to

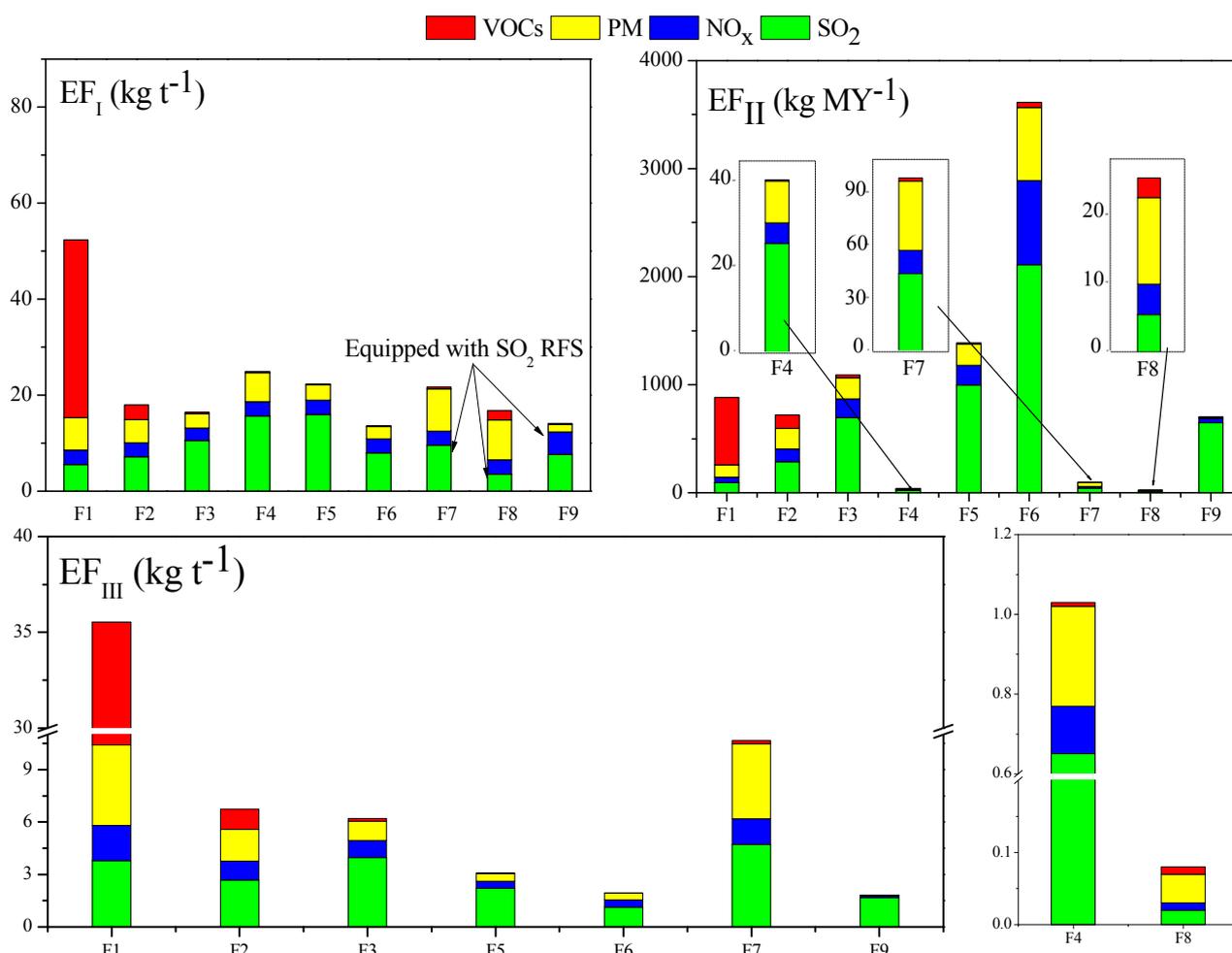


Fig. 3. Values of pollutant EF_I , EF_{II} , and EF_{III} for 9 food processing plants.

high sulfur content of coal in food plants. Considering SO₂ EF_{II}, it showed a different trend compared to EF_I, implied the influence of product price beside aforementioned sulfur content and SO₂ RE. The average SO₂ EF_{II} of the 3 companies with installed SO₂ RFs was 326 kg MY⁻¹, far less than that of the companies without SO₂ RFs (607 kg MY⁻¹). The highest EF_{II} owner was F6 (2111 kg MY⁻¹) among 9 enterprises, which might be ascribed to its cheap product (vinegar), while the lowest value of occurred at F. Among 3 factories containing SO₂ RFs, F9 possessed the highest SO₂ EF_{II} of 647 kg MY⁻¹, while P8 owned the lowest value of 5.39 kg MY⁻¹, which were attributed to the differences of coal consumption, product price, sulfur content, and SO₂ RE of RFs.

All the 9 enterprises were not equipped with NO_x RFs, so the NO_x formation processes became the determining factors for NO_x emissions. Unlike SO₂ forming by combustion of sulfur in coal, NO_x mainly originated from oxidation of air N₂ at high temperature regardless of that small proportion of NO_x formed by burning of fuel-nitrogen (Löfller *et al.*, 2005), so SO₂ emissions were more depended on the coal compositions than NO_x due to the different formation mechanisms between them. Therefore, NO_x EF_I values experienced minor fluctuation compared to SO₂ EF_I, which ranged from 2.58 to 4.72 kg t⁻¹ with the mean value as 3.09 kg t⁻¹. F9 had the maximum NO_x EF_I (4.72 kg t⁻¹) owing to its high level of nitrogen content in coal, and the minimum value was found in F8 (2.58 kg t⁻¹). Unlike NO_x EF_I, huge fluctuation existed in NO_x EF_{II} values (reported in kg MY⁻¹), varied from 4.44 of F8 to 777 of F6 with the mean as 151 kg MY⁻¹, which was in part resulted from the output difference among 9 enterprises.

The EF_I values were mainly determined by two factors such as PM RE values and ash content of coal. The factories with PM REs < 80% owned much higher PM EF_I of 7.49 kg t⁻¹ than 2.67 kg t⁻¹ of the other enterprises with PM REs > 85%. The PM EF_{II} values varied significantly from 9.83 to 678 kg MY⁻¹ with an average of 162 ± 194 kg MY⁻¹, which was similar to NO_x.

It should be mentioned that VOCs EF_I of F1 (37.0 kg MY⁻¹) dominated over the remaining 8 factories (0.18–3.08 kg t⁻¹), which was explained by not only the lacks of the VOCs RFs, but also the production process of white spirit. The average plus standard deviation of VOCs EF_{II} was 93.1 ± 184 kg MY⁻¹, which exhibited a significant fluctuation. F1 was the highest among the 9 companies and as high as 624 kg MY⁻¹.

For EF_{III} values, F1 owned the maximum values of SNPV, the correspondingly lower values occurred at F4 and F8, which reflected the influence of product yield. The EF_{III} values of 3 companies equipped with SO₂ RFs were 4.73, 0.02 and 1.68 kg t⁻¹ respectively with the average was 1.52 ± 2.32 kg t⁻¹, while they ranged from 0.650 to 3.97 kg t⁻¹ with mean value as 2.41 ± 1.35 kg t⁻¹ for the rest factories. Unlike NO_x EF_{II}, EF_{III} values also possessed large fluctuation (0.010–2.00 kg t⁻¹) among 9 enterprises due to the differences of product yields among 9 enterprises. Due to the high emission in F1, the EF_{III} of VOCs (25.1 kg t⁻¹) was far more than the other factories (0.010–1.16 kg t⁻¹).

Li *et al.* (2018) discussed the EFs of SO₂, NO_x, and PM for coal-fired power plants (CFPPs), cement factories, and coking enterprises. Comparing the corresponding values with abovementioned large-scale coal-fired power plants, food factories possessed higher EF_I values of SO₂, NO_x, and PM due to the perfect pollutant RFs in power generation industries. The EF_I in food factories (report in kg t⁻¹) were 9.31 ± 4.23 (SO₂), 3.10 ± 0.56 (NO_x), and 5.01 ± 2.36 (PM), while the cement factories were 4.35 ± 3.01, 7.73 ± 5.98, and 50.7 ± 77.1. SO₂ EF_I of food factories was lower than cement enterprises due to the SO₂ RFs installed in all the cement factories, while higher NO_x and PM EF_I values occurred at cement industries due to the difference of production processes.

Emission of Air Pollutants from Brick Factories

In this study, 34 small-size brick manufacturing factories using coal gangue, clay, and coal fly ash as raw materials were investigated to analyze the emissions of pollutants and calculate the EFs. Coal consumptions, output values, and product yields differed significantly among different companies. The coal consumptions of these plants ranged from 1.00 × 10³ t a⁻¹ to 1.47 × 10⁵ t a⁻¹, the product outputs varied from 1,500 to 150,000 t a⁻¹, and the annual outputs were in the range of 15.0 to 6.94 × 10³ MY. In regard to raw materials, 31 of 34 companies applied coal gangue in production process, and 1 and the rest 2 factories used the fly ash used clay in production. Among the 34 factories, only 13 of them equipped with SO₂ RFs with the removal rate as 30.0%–88.9%. Owing to the small enterprise size, none of these factories installed the devices to remove the NO_x and VOCs. For PM, 31 of them had the DRs with REs ranged from 41.0% to 99.0%.

All the EF values expressed as EF_I, EF_{II}, and EF_{III} for each brick factory were listed in Tables S10–S13. The EF_I values about SO₂ for 34 brick factories were weak correlated with their sulfur contents in coal, which might be related to the co-burning of coal and coal gangue process and unique smoke venting mode in brick production. Coal gangue containing sulfur contents also resulted in the SO₂ formation, which could weaken the correlation between coal containing sulfur contents with SO₂ EF_I. Co-emission of water vapor from drying process of unfired brick and flue gas from roasting process decreased the temperature of flue gas, further reduce the SO₂ REs. Significant difference existed between companies with and without SO₂ RFs, the mean value of SO₂ EF_I for both companies were 3.73 and 9.29 kg t⁻¹, respectively (Fig. 4). SO₂ EF_I values ranged from 0.65 and 35.0 kg t⁻¹, the highest value occurred in B24 (35.0 kg t⁻¹). The lack of the SO₂ RFs and the highest sulfur content of 3.5% could be the reason. EF_I values of NO_x, PM, and VOCs varied from 0.08 to 2.94 from 0.03 to 6.27 kg t⁻¹; and from 0.02 to 5.33 kg t⁻¹. It should be noted that B7 produced with fly ash emitted the highest PM, which proved the influence of raw materials on pollutant emissions. The significant difference between whether installed the SO₂ RFs could explain the rapid EF values.

Fig. 5 showed the EF_{II} (reported in t MY⁻¹) of these factories, they ranged from 0.68 to 400, 0.42 to 9.41, 0 to

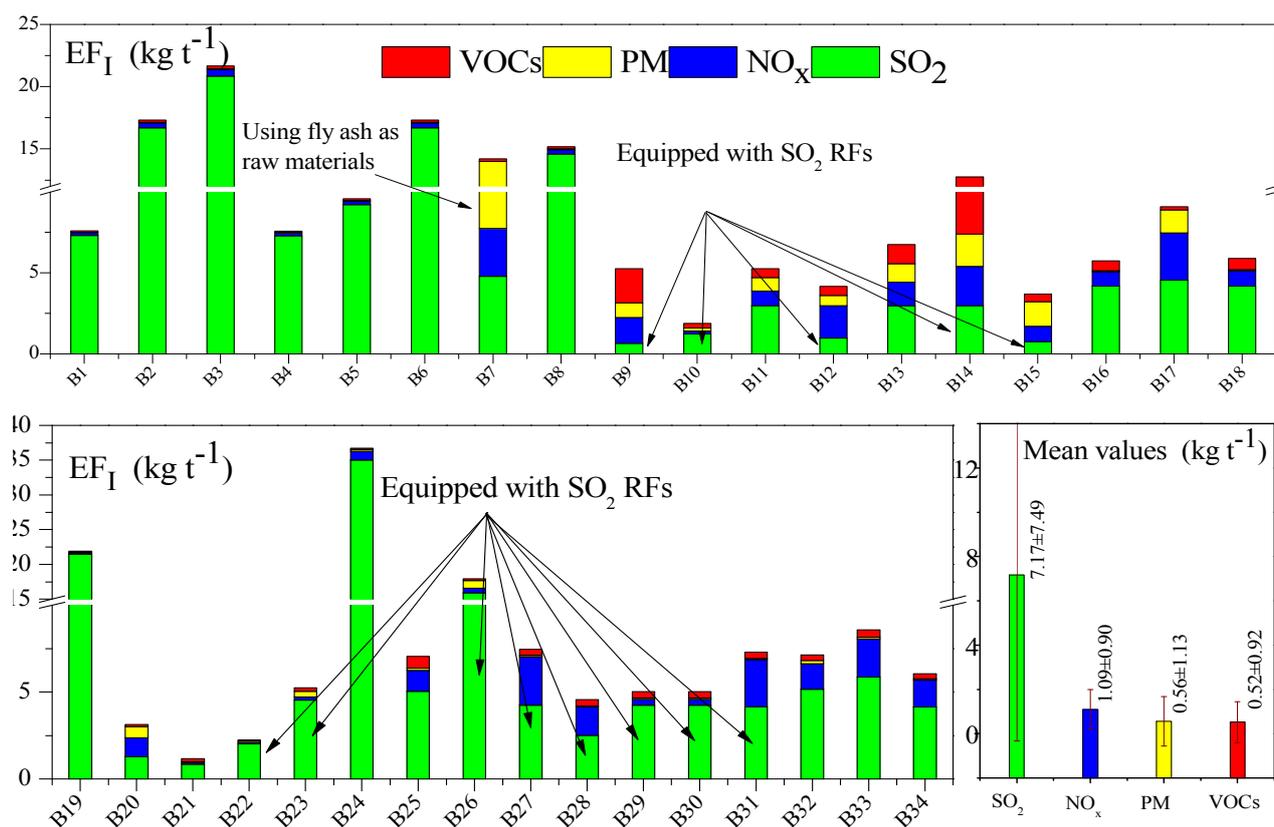


Fig. 4. EF_I values for 34 brick production factories.

3.24 and 0.06 to 4.52 for SO₂, NO_x, PM and VOCs, respectively. Fig. 6 showed the EF_{III} values and for SNPV, the values were ranged from 0.09 to 20.0, 0.05 to 4.82, 0.03 to 1.17 and 0.01 to 2.75 kg t⁻¹, respectively. 13 factories with SO₂ RFs showed a lower EF_{III} values than the other 21 factories without SO₂ RFs installed, the mean values were 3.85 ± 4.25 and 7.54 ± 6.53 kg t⁻¹. It should be noticed that the highest EF_{III} values about SO₂, NO_x and VOCs all occurred in B25 (20.0, 4.82 and 2.75 kg t⁻¹), the low product yield and the high amount of emission would be the explanations.

Figs. 4, 5 and 6 also listed the mean values of EF_I, EF_{II}, and EF_{III} of 4 air pollutants. For EF_I, the mean values were 7.17 ± 7.49 for SO₂, 1.09 ± 0.90 for NO_x, 0.56 ± 1.13 for PM and 0.52 ± 0.92 for VOCs. Considering EF_{II}, the mean values were 56.5 ± 94.1 for SO₂, 2.74 ± 2.08 for NO_x, 0.86 ± 0.82 for PM and 1.15 ± 0.89 for VOCs. In regard to EF_{III}, the mean values were 137 ± 156 for SO₂, 14.8 ± 19.9 for NO_x, 4.79 ± 6.72 for PM and 6.79 ± 11.4 for VOCs. In regard to EF_{III}, the mean values were 5.48 ± 6.25 for SO₂, 0.59 ± 0.80 for NO_x, 0.19 ± 0.27 for PM and 0.27 ± 0.45 for VOCs.

The obvious fluctuation of EF_I occurred among 34 companies owing to the influence of the pollutant removal rates, sulfur contents within coal and coal gangue, ash contents, and conditions of coal combustion.

The EF_I values for SO₂ and NO_x for the CFPPs were much lower than those of brick making factories, the corresponding values in CFPPs and brick factories were

1.03 ± 1.25 vs. 1.04 ± 0.69, and 7.17 ± 7.49 vs. 1.09 ± 0.90, respectively (Li *et al.*, 2018). However, the average PM EF_I of brick companies was lower than that of CFPPs. The EF_I of cement plants were 4.35 ± 3.02, 7.73 ± 5.98, 50.8 ± 77.1 kg t⁻¹, respectively (Li *et al.*, 2018). SO₂ EF_I values of brick factories were higher than those of cement ones due to the installation of effective SO₂ RFs in cement companies, while higher NO_x and PM EF_I values occurred at cement industries.

CONCLUSIONS

In this study, 51 companies in Shanxi Province detailed described in methodology part were surveyed for their coal compositions and consumptions, annual outputs, product yields, and pollutant RFs. Also, SO₂, PM, and NO_x were on-site measured, and VOCs were sampled and analyzed in laboratory aimed to obtain the pollutant EF_I associated with coal consumptions, annual output, and product yield.

Overall the SO₂ EF_I dominated all the other three pollutants among all the three kinds of companies. The key influencing factors on the SO₂ emissions were sulfur content of coal and SO₂ RE for food and pharmaceuticals factories, while the sulfur content in coal gangue was also an important factor for brick companies. Increased VOCs emissions could be resulted from the utility of organic solvents in small fractional pharmaceuticals factories and production of white spirit. Unlike SO₂ completely originated from burning of sulfur in coal, NO_x also could be formed

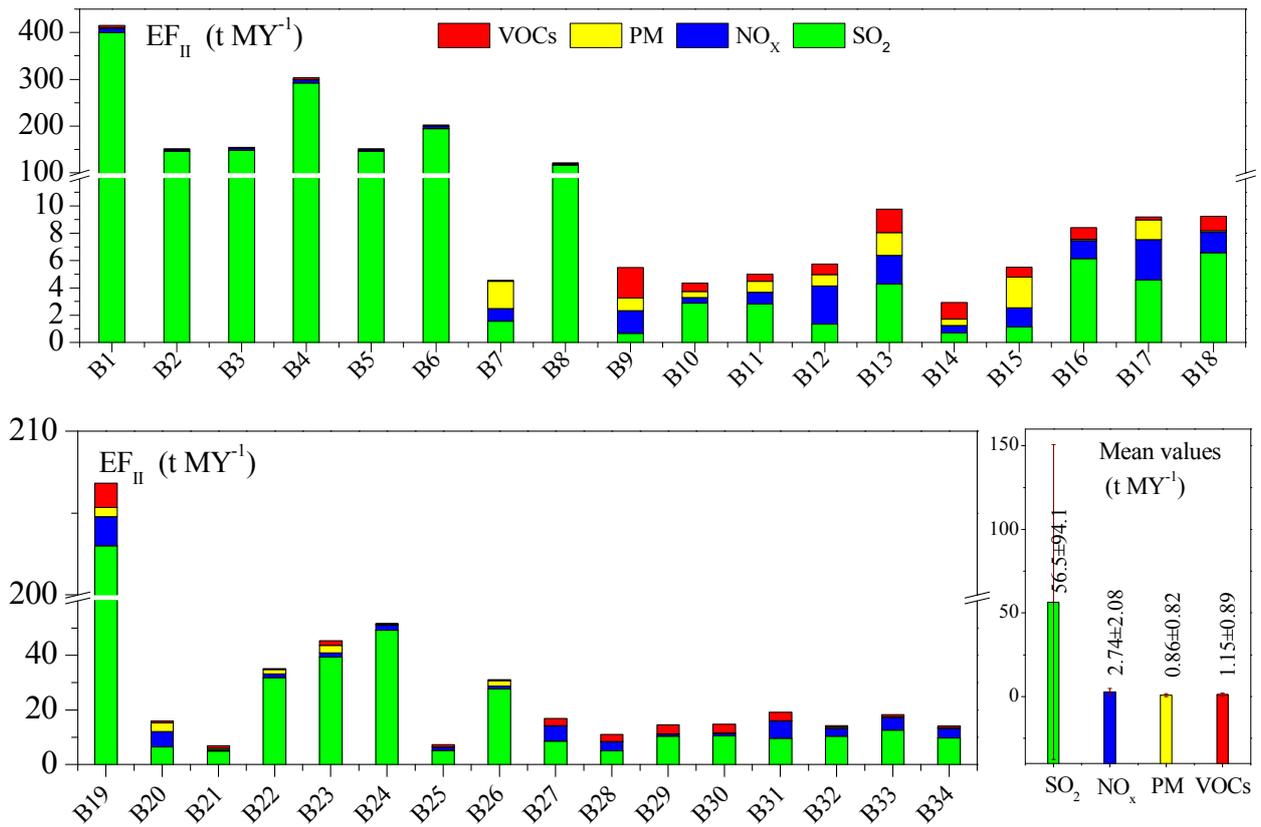


Fig. 5. EF_{II} values for 34 brick production factories.

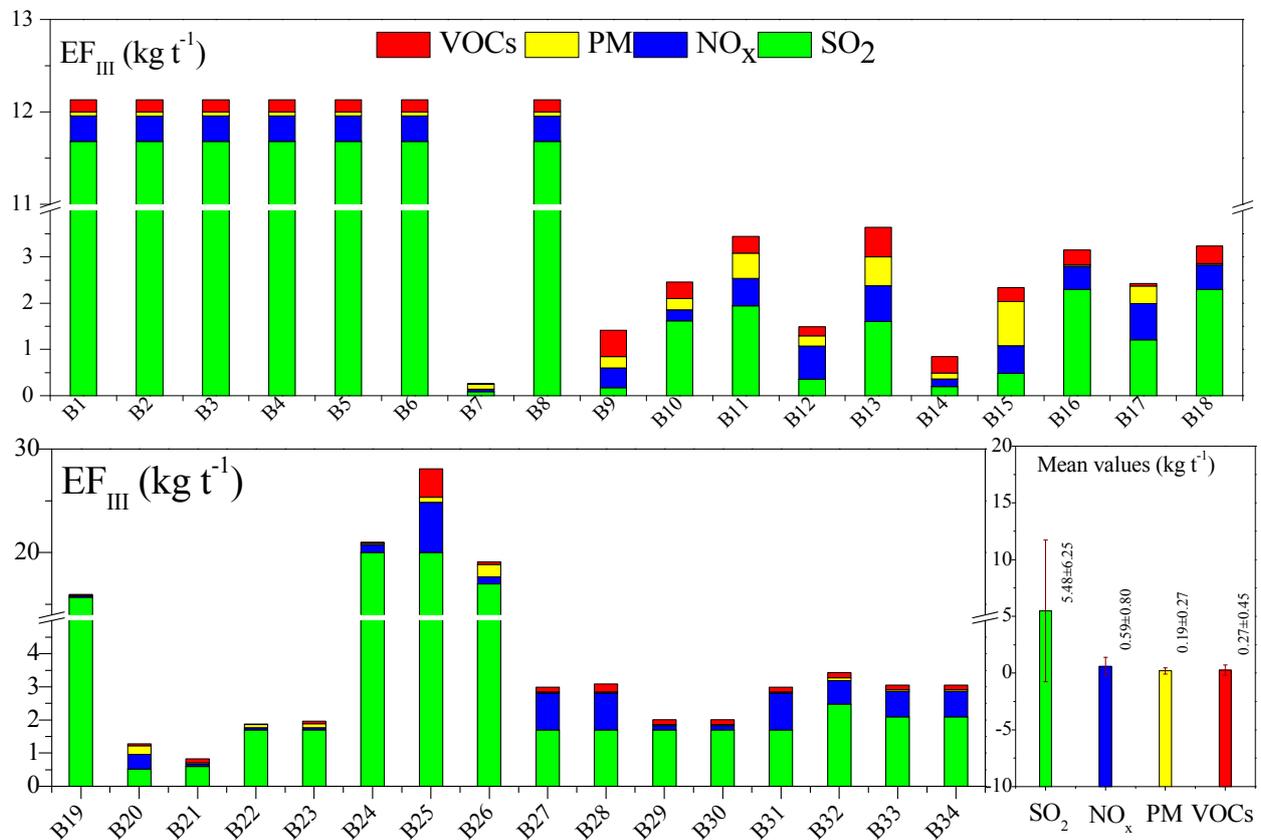


Fig. 6. EF_{III} values for 34 brick production factories.

by oxidation of air N₂ at high combustion temperature, NO_x EF_I values experienced minor fluctuations within the same industries compared to the other air pollutants. EF_{II} values for 4 air pollutants for brick factories dominated over the other two kinds of industries due to its low output.

3 of 8 pharmaceuticals factories with SO₂ RFs (P2, P4, and P8) possessed SO₂ EF_I values of 3.00, 1.91 and 2.51 kg t⁻¹, and SO₂ EF_{II} values of 16.1, 86.3 and 5.01 kg MY⁻¹. The values of EF_I and EF_{II} of SO₂ for rest 5 factories were in the range of 6.67 to 11.2 kg t⁻¹, and 74.7 to 231 kg MY⁻¹. The same trends occurred in food and brick factories, the EF_I and EF_{II} in food factories were 6.94 ± 2.51 kg t⁻¹, 37.9 ± 24.6 kg MY⁻¹ with SO₂ RFs, and 10.5 ± 4.05 kg t⁻¹, 702 ± 715 kg MY⁻¹ without SO₂ RFs. In brick factories, the EF_I and EF_{II} values were 3.73 ± 3.80 kg t⁻¹, 11.5 ± 12.5 kg MY⁻¹ with SO₂ RFs, and 9.29 ± 8.37 kg t⁻¹, 84.4 ± 110 kg MY⁻¹.

The mean value of NO_x EF_I (in kg t⁻¹) was 2.71 ± 0.78, 3.10 ± 0.56, and 1.09 ± 0.90 for pharmaceuticals, food, and brick companies, and the corresponding NO_x EF_{II} values were 56.6 ± 34.3 kg MY⁻¹, 151 ± 223 kg MY⁻¹, and 2.74 ± 2.08 t MY⁻¹.

In terms of coal quality in three types of companies, PM emissions depended on not only the dust RE, but also the ash content in coal. The ash content of coal used in P1 reached 20.0%, and the PM EF_I was the highest (7.5 kg t⁻¹) among the 8 pharmaceuticals companies, while their average value was 3.87 ± 2.35 kg t⁻¹. The corresponding PM EF_I and EF_{II} were 5.01 ± 2.36 kg t⁻¹ vs. 162 ± 194 kg MY⁻¹ in food factories, and 0.55 ± 1.12 kg t⁻¹ vs. 0.84 ± 0.83 t MY⁻¹ in brick factories.

The 2nd and 6th pharmaceuticals factories possessed a prominent VOCs EF_I values compared with other plants due to the utility of organic solvents which were 22.9 and 15.3 kg t⁻¹, while the average was 5.03 ± 8.35 kg t⁻¹. The mean value of VOCs EF_I in food and brick factories (in kg t⁻¹) were 4.83 ± 10.9 and 0.52 ± 0.92, and EF_{II} were 93.2 ± 184 kg MY⁻¹, 1.15 ± 0.89 t MY⁻¹. The 1st food factory owned the highest EF_I value as 37.0 kg t⁻¹ due to the production process of white spirit.

In general, compared with LSIs, SO₂ EFs for these SSIs were much higher than those of LSIs such as CFPPs and cement factories, NO_x EFs were at the same level with those of LSIs, and PM EFs were much lower than cement factories and similar to CFPPs.

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SUPPLEMENTARY MATERIAL

Supplementary data associated with this article can be found in the online version at <http://www.aaqr.org>.

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