Emission Factors of NOx, 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, NOx, PM, and VOCs (SNPV) associated with coal consumption (EFI), industrial output (EFII), and product yield (EF III). Among them, PM-RFs were all equipped except for 3 brick factories, no NOx- 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 owning 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 EFI 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 NOx 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 NOx, 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. NOx EFs experienced lessen fluctuations than other pollutants among all the factories due to the different formation mechanism and no installation of NOx RFs. EF III showed various fluctuations due to the different product types.

Keywords: Emission factor; NOx; SOx; 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, NOx, 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 PM2.5 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 NOx were main contributors to PM2.5 (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). NOx can promote the formation of photo-chemical smog, visibility reduction, and acid rain, and ozone depletion. As the main compositions of NOx,
NO and NO\(_2\) 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 O\(_3\) (Zhang et al., 2015; Hu et al., 2018).

Along with China's economic development, emissions of PM, NO\(_x\), SO\(_2\), 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 owning 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 SO\(_2\), 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 SO\(_2\), 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, SO\(_2\), 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 SO\(_2\) 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 ± 0.444% for food, and 0.661 ± 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.

<table>
<thead>
<tr>
<th></th>
<th>Sa (%)</th>
<th>Aad (%)</th>
<th>CC (t a⁻¹)</th>
<th>SO₂ RE (%)</th>
<th>PM RE (%)</th>
<th>Output (MY)</th>
<th>Product</th>
<th>Yield (t a⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>P1</td>
<td>0.70</td>
<td>20.0</td>
<td>400</td>
<td>0</td>
<td>93.0</td>
<td>60.0</td>
<td>Vaccine</td>
<td>200</td>
</tr>
<tr>
<td>P2</td>
<td>0.30</td>
<td>6.00</td>
<td>5400</td>
<td>19.8</td>
<td>88.0</td>
<td>1099</td>
<td>Injection</td>
<td>130</td>
</tr>
<tr>
<td>P3</td>
<td>0.45</td>
<td>17.0</td>
<td>950</td>
<td>0</td>
<td>77.0</td>
<td>29.8</td>
<td>Pill</td>
<td>31</td>
</tr>
<tr>
<td>P4</td>
<td>0.35</td>
<td>17.8</td>
<td>6790</td>
<td>27.9</td>
<td>77.0</td>
<td>150</td>
<td>Oral liquid</td>
<td>330</td>
</tr>
<tr>
<td>P5</td>
<td>0.45</td>
<td>17.0</td>
<td>150</td>
<td>0</td>
<td>99.0</td>
<td>5.20</td>
<td>Injection</td>
<td>3.5</td>
</tr>
<tr>
<td>P6</td>
<td>0.45</td>
<td>17.0</td>
<td>300</td>
<td>0</td>
<td>95.0</td>
<td>10.0</td>
<td>Sterile powder</td>
<td>12</td>
</tr>
<tr>
<td>P7</td>
<td>0.45</td>
<td>17.0</td>
<td>800</td>
<td>0</td>
<td>95.0</td>
<td>26.7</td>
<td>Injection</td>
<td>60</td>
</tr>
<tr>
<td>P8</td>
<td>0.50</td>
<td>15.0</td>
<td>3920</td>
<td>61.1</td>
<td>87.7</td>
<td>1963</td>
<td>Injection</td>
<td>8.8</td>
</tr>
</tbody>
</table>

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

<table>
<thead>
<tr>
<th></th>
<th>Sa (%)</th>
<th>Aad (%)</th>
<th>CC (t a⁻¹)</th>
<th>SO₂ RE (%)</th>
<th>PM RE (%)</th>
<th>Output (MY)</th>
<th>Product</th>
<th>Yield (t a⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>F1</td>
<td>0.35</td>
<td>17</td>
<td>393</td>
<td>0</td>
<td>73.5</td>
<td>23.3</td>
<td>White spirit</td>
<td>579</td>
</tr>
<tr>
<td>F2</td>
<td>0.35</td>
<td>17</td>
<td>120</td>
<td>0</td>
<td>95.0</td>
<td>3.00</td>
<td>Steamed bread slices</td>
<td>320</td>
</tr>
<tr>
<td>F3</td>
<td>0.45</td>
<td>17</td>
<td>589</td>
<td>0</td>
<td>99.0</td>
<td>8.91</td>
<td>Bean</td>
<td>1563</td>
</tr>
<tr>
<td>F4</td>
<td>0.35</td>
<td>17</td>
<td>620</td>
<td>0</td>
<td>70.0</td>
<td>383</td>
<td>Sour milk beverage</td>
<td>15000</td>
</tr>
<tr>
<td>F5</td>
<td>1</td>
<td>20</td>
<td>1360</td>
<td>0</td>
<td>87.0</td>
<td>21.9</td>
<td>Sour milk beverage</td>
<td>9860</td>
</tr>
<tr>
<td>F6</td>
<td>0.5</td>
<td>15</td>
<td>1428</td>
<td>0</td>
<td>90.0</td>
<td>5.40</td>
<td>Vinegar</td>
<td>10000</td>
</tr>
<tr>
<td>F7</td>
<td>0.6</td>
<td>21</td>
<td>1182</td>
<td>71.4</td>
<td>67.0</td>
<td>260</td>
<td>Meat</td>
<td>2400</td>
</tr>
<tr>
<td>F8</td>
<td>1.4</td>
<td>17</td>
<td>379</td>
<td>85.0</td>
<td>79.0</td>
<td>251</td>
<td>Feed</td>
<td>85535</td>
</tr>
<tr>
<td>F9</td>
<td>1.5</td>
<td>17</td>
<td>1100</td>
<td>70.0</td>
<td>95.0</td>
<td>130</td>
<td>Feed</td>
<td>50000</td>
</tr>
</tbody>
</table>

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

**Sampling and Measurement of SO₂, NOₓ, and PM**

The pollutants such as gaseous SO₂, NOₓ, 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 (NOx, 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 (EF₁ reported in kg t⁻¹), annual output (EF₂ reported in kg MY⁻¹), and product (reported in kg t⁻¹). All the EF forms were calculated using Eqs. (1)–(3).

EF₁ (kg t⁻¹) = C (kg m⁻³) × VFA (m³ kg⁻¹) × 1000 (kg t⁻¹)

(1)

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

EF₂ (kg MY⁻¹) = (C (kg m⁻³) × 1000 (kg t⁻¹) × VFA (m³ kg⁻¹) × Coal consumption (t a⁻¹))/(Output (MY a⁻¹))

(2)

EF₃ (kg t⁻¹) = (C (kg m⁻³) × 1000 (kg t⁻¹) × VFA (m³ kg⁻¹) × Coal consumption (t a⁻¹))/(Product yield (t a⁻¹))

(3)

VFA was derived from the theoretical air amount required by 1 kg of coal combustion (VAT) calculated by Eq. (4).

V_AT = 0.0889ω₁(Car) + 0.2567ω₂(Har) + 0.0333ω₃(Sar) + 0.0762ω₄(Nar) – 0.0333ω₅(Oar)

(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_CO₂ + V_SO₂ + V_N₂O + V_N₂ + V_H₂O

(5)

V_CO₂ + V_SO₂ + V_N₂O = 0.01867ω₁(Car) + 0.007ω₂(Har) + 0.0016ω₃(Sar)

(6)

where V_CO₂, V_SO₂, and V_N₂O refer to volumes of CO₂, SO₂,
Table 3. Statistics about coal compositions, output, and yield of brick factories.

<table>
<thead>
<tr>
<th>Sample</th>
<th>( S_a ) (%)</th>
<th>( A_a ) (%)</th>
<th>( CC ) (t a(^{-1}))</th>
<th>( SO_2 ) RE (%)</th>
<th>( PM ) RE (%)</th>
<th>Output (MY)</th>
<th>Raw material</th>
<th>Yield (t a(^{-1}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>B1</td>
<td>0.36</td>
<td>17</td>
<td>147000</td>
<td>0</td>
<td>99</td>
<td>2.69</td>
<td>Gangue</td>
<td>92100</td>
</tr>
<tr>
<td>B2</td>
<td>0.3</td>
<td>13</td>
<td>1400</td>
<td>0</td>
<td>83</td>
<td>0.16</td>
<td>Gangue</td>
<td>2000</td>
</tr>
<tr>
<td>B3</td>
<td>0.3</td>
<td>16.5</td>
<td>7000</td>
<td>0</td>
<td>98</td>
<td>0.98</td>
<td>Gangue</td>
<td>12500</td>
</tr>
<tr>
<td>B4</td>
<td>0.3</td>
<td>17</td>
<td>54000</td>
<td>0</td>
<td>83</td>
<td>1.35</td>
<td>Gangue</td>
<td>33750</td>
</tr>
<tr>
<td>B5</td>
<td>0.3</td>
<td>17</td>
<td>38000</td>
<td>0</td>
<td>83</td>
<td>2.40</td>
<td>Gangue</td>
<td>30000</td>
</tr>
<tr>
<td>B6</td>
<td>0.3</td>
<td>17</td>
<td>3500</td>
<td>0</td>
<td>85</td>
<td>0.30</td>
<td>Gangue</td>
<td>50000</td>
</tr>
<tr>
<td>B7</td>
<td>0.3</td>
<td>16.5</td>
<td>1221</td>
<td>0</td>
<td>95</td>
<td>3.80</td>
<td>Coal fly ash</td>
<td>56787</td>
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<tr>
<td>B8</td>
<td>0.3</td>
<td>17</td>
<td>1200</td>
<td>0</td>
<td>83</td>
<td>0.15</td>
<td>Gangue</td>
<td>1500</td>
</tr>
<tr>
<td>B9</td>
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<td>13.3</td>
<td>9400</td>
<td>88.9</td>
<td>99</td>
<td>9.02</td>
<td>Gangue</td>
<td>35000</td>
</tr>
<tr>
<td>B10</td>
<td>0.38</td>
<td>27</td>
<td>56000</td>
<td>84.0</td>
<td>99</td>
<td>24.0</td>
<td>Gangue</td>
<td>42500</td>
</tr>
<tr>
<td>B11</td>
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<td>27</td>
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<td>0</td>
<td>95</td>
<td>19.0</td>
<td>Gangue</td>
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</tr>
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<td>27</td>
<td>18000</td>
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<td>Gangue</td>
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</tr>
<tr>
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<td>27</td>
<td>10100.6</td>
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<td>7.00</td>
<td>Gangue</td>
<td>18750</td>
</tr>
<tr>
<td>B14</td>
<td>0.25</td>
<td>27</td>
<td>3000</td>
<td>30.0</td>
<td>99</td>
<td>13.0</td>
<td>Gangue</td>
<td>45000</td>
</tr>
<tr>
<td>B15</td>
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<td>27</td>
<td>25366</td>
<td>82.4</td>
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<td>17.0</td>
<td>Gangue</td>
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<tr>
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<td>0.25</td>
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<td>22000</td>
<td>0</td>
<td>99</td>
<td>15.0</td>
<td>Gangue</td>
<td>40000</td>
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<td>B17</td>
<td>0.25</td>
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<td>10000</td>
<td>0</td>
<td>99</td>
<td>9.90</td>
<td>Gangue</td>
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<td>0.25</td>
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<td>0</td>
<td>99</td>
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<td>B19</td>
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<td>31752</td>
<td>0</td>
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<td>3.36</td>
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<td>3.00</td>
<td>Gangue</td>
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<td>8.00</td>
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<td>48.1</td>
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<td>6.48</td>
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<td>7.11</td>
<td>Gangue</td>
<td>17500</td>
</tr>
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<td>B25</td>
<td>0.3</td>
<td>14</td>
<td>70802</td>
<td>0</td>
<td>0</td>
<td>69.4</td>
<td>Gangue</td>
<td>17808</td>
</tr>
<tr>
<td>B26</td>
<td>1.3</td>
<td>16000</td>
<td>57.0</td>
<td>99.6</td>
<td>9.20</td>
<td>Gangue</td>
<td>15000</td>
<td></td>
</tr>
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and NO\(_2\) derived from burning of coal containing carbon, sulfur, and nitrogen, \( V_{N2} \) is the \( N_2 \) 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 \( (0.112\omega(H_2)) \), vaporization of water in coal \( (0.00124\omega(M_2)) \), and vapor in air \( (0.0161V_{AT}) \).

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

\[
V_{FA} = V_{FT} + (\alpha - 1)V_{AT} + 0.0161(\alpha - 1)V_{AT} \tag{7}
\]

\( \alpha \) 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 \((-40^\circ C)\) to concentrate VOCs and rapidly desorbed by a TD-100 system, and then transferred to GC-MS system equipped with a DB-1 capillary column \((60 \text{ m} \times 0.32 \text{ mm} \times 1.0 \text{ mm}, \text{Agilent Technologies, USA})\). 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 N₂ 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 µg m⁻³) for 57 species ranged from 0.1 of C₆H₁₄ (CAS: 75–83–2) to 1 of C₃H₆O (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 (R² = 0.13, p < 0.05), indicated the influence of drug prices. Generally the SO₂ RFs occurred in factories with high output values such as P2, P4, and P8, while NOₓ 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 SO₂ emissions were sulfur content of coal and removal efficiency of SO₂ (Li et al., 2018). The drastic fluctuations of EF₁ and EFgröße II for SO₂ occurred among 8 plants and the weak correlation was found between EF₁ and EFgröße II (R² = 0.12, p < 0.05), which reflect the greater impact of product prices of different factories. The EF₁ values ranged from 1.91 to 11.2 kg t⁻¹ (mean: 5.86 ± 2.95), and they were 5.01–231 kg MY⁻¹ (mean: 130 ± 88.3) for EFgröße II (Figs. 1 and 2). The highest SO₂ EF₁ 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 Sad (0.70%) of coal and the deficiency of SO₂ RFs for P1 were explanations.

For SO₂ EFgröße II values, the highest and lowest EFgröße II were owned by P3 (231 kg MY⁻¹) and P8 (5.01 kg MY⁻¹), respectively.

The EFs of SO₂ for 8 plants could be identified as two categories based on with or without SO₂ RFs. Class I
including P2, P4, and P8 had a low EF value (3.01, 1.91 and 2.51 kg t⁻¹) with installed SO₂ RFs and high outputs, while high EF 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 EF₁ (7.90 kg t⁻¹) and EFⅡ (35.8 kg MY⁻¹) of SO₂ for Class II were 3.20 and 5.20 times of corresponding values of Class I. EF₁ values were well correlated with sulfur contents of coal among 5 plants in Class II (R² = 0.92), while the weak correlation was found among 8 factories (R² = 0.52), indicating more strong influence of SO₂ RE than sulfur content. The 3 factories within Class I possessed much lower EFⅡ of 35.8 kg MY⁻¹ than mean value of 186 kg MY⁻¹ of 5 plants in Class II. EFⅡ was more affected by product price than coal compositions.

There was little fluctuation for NOₓ EF₁ 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 NOₓ removal equipment among all the 8 factories should be underlined. The highest NOₓ EF₁ occurred at P1 with the coal consumption as 400 t a⁻¹. Drastic emissions of fuel-NOₓ related to burning of high nitrogen content of coal and thermal NOₓ associated with high burning temperature in P1 was impetus for the highest NOₓ EF₁ (Löffler et al., 2005). EFⅡ values (reported in kg MY⁻¹) of NOₓ varied from 5.63 of P8 to 93.8 of P3 with the mean value as 56.6 (Fig. 1 and 2). Low NOₓ EFⅡ values were found for P2 and P8 as 9.96 kg MY⁻¹ and 5.63 kg MY⁻¹, respectively, owning to the high price of products from two factories. The rest 6 factories possessed high NOₓ EFⅡ values from 27.83 to 93.7 kg MY⁻¹. It should be noted that NOₓ emission was less influenced by coal quality than SO₂ emission due to the different formation mechanisms between NOₓ and SO₂.

In regard to PM emissions, the EF₁ 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 EF₁ than 3.87 kg t⁻¹, and the highest PM EF₁ (7.5 kg t⁻¹) appeared at P1 ascribe to the highest ash contend (20%) (Table 1). The coal with medium ash content based on Aₐ (ash content on dry basis) was used in these 8 plants, and the highest Aₐ of 20% in P1 resulted in the highest EF₁. P2 owned the lowest PM EF₁ value resulted from low Aₐ (6%) of coal. In a word, The PM EF₁ was mainly subjected to ash content of coal and PM RE of RFs. The PM EFⅡ values exhibited a similar trend with NOₓ EFⅡ; 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 EFⅡ. 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 EF₁ 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 Vₐd (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 EFⅡ 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 EFⅡ values.

In regard to EFⅢ 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 ± 92.7 kg t⁻¹. Due to the different drug production processes, higher fluctuation was showed in NOx, PM and VOCs values which ranged from 8.35 to 1256 kg t⁻¹, 10.9–1059 kg t⁻¹ and 0.36–951 kg t⁻¹ with the average as 396 ± 211, 328 ± 202 and 313 ± 187kg 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. EF₁ and EF₂ 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 owning 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 (R² = 0.14) due to the influence of product price. 3 of 4 factories with high output values were equipped with SO₂ RFs such as P7, P8, and P9, while the rest F4 without SO₂ 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 NOx 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 SO₂ EF₁ values for 6 factories without SO₂ RFs was 0.98, indicated that sulfur content had a great influence on SO₂ EF₁. The 3 factories owning SO₂ RFs possessed much lower SO₂ EF₁ (6.94 ± 2.51 kg t⁻¹) than that (10.5 ± 4.05 kg t⁻¹) of the other factories. The highest EF₁ occurred in F5, as high as 16.0 kg t⁻¹, might be related to the high sulfur content in coal (1%). The average EF₁ 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
high sulfur content of coal in food plants. Considering SO$_2$ EF$_{II}$, it showed a different trend compared to EF$_{I}$, implying the influence of product price beside aforementioned sulfur content and SO$_2$ RE. The average SO$_2$ EF$_{II}$ of the 3 companies with installed SO$_2$ RFs was 326 kg MY$^{-1}$, far less than that of the companies without SO$_2$ 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 9 factories containing SO$_2$ RFs, F9 possessed the highest SO$_2$ 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 SO$_2$ 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 SO$_2$ forming by combustion of sulfur in coal, NO$_x$ mainly originated from oxidation of air N$_2$ at high temperature regardless of that small proportion of NO$_x$ formed by burning of fuel-nitrogen (Löffler et al., 2005), so SO$_2$ 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 SO$_2$ 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$_{II}$ values showed large fluctuation in existed 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$_{II}$ 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$_{II}$ 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$_{II}$ 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 lack of the VOCs RFs, but also the production process of white spirit. The average plus standard deviation of VOCs EF$_{II}$ was 93.1 ± 4.23 (SO2), 3.10 ± 0.56 (NOx), and 5.01 ± 2.36 (PM), which was far more than the other factories (0.010–1.16 kg t$^{-1}$).

Li et al. (2018) discussed the EFs of SO$_2$, NO$_x$, and PM for co-fired power plants (CFPPs), cement factories, and coking enterprises. Comparing the corresponding values with aforementioned large-scale coal-fired power plants, food factories possessed higher EF$_{I}$ values of SO$_2$, 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 (SO$_2$), 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. SO$_2$ EF$_{I}$ of food factories was lower than cement enterprises due to the SO$_2$ 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 × 10$^3$ t a$^{-1}$ to 1.47 × 10$^5$ 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 × 10$^3$ 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 SO$_2$ RFs with the removal rate as 30.0%–88.9%. Owning to the small enterprise size, none of these factories installed the devices to remove the NO$_x$, and VOCs. 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 SO$_2$ 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 SO$_2$ formation, which could weaken the correlation between coal containing sulfur contents with SO$_2$ 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 SO$_2$ REs. Significant difference existed between companies with and without SO$_2$ RFs, the mean value of SO$_2$ EF$_{II}$ for both companies were 3.73 and 9.29 kg t$^{-1}$, respectively (Fig. 4). SO$_2$ EF$_{II}$ 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 SO$_2$ RFs and the highest sulfur content of 3.5% could be the reason. EF$_{II}$ 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 SO$_2$ 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...
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3.24 and 0.06 to 4.52 for \(SO_2\), \(NO_x\), 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 \(SO_2\) RFs showed a lower EF_{III} values than the other 21 factories without \(SO_2\) 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 \(SO_2\), \(NO_x\) 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 \(SO_2\), 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 \(SO_2\) 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 \(SO_2\), 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 \(SO_2\), 0.59 ± 0.80 for \(NO_x\), 0.19 ± 0.27 for PM and 0.27 ± 0.45 for VOCs.

The obvious fluctuation of EFs 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 \(SO_2\) 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). \(SO_2\) EF_{I} values of brick factories were higher than those of cement ones due to the installation of effective \(SO_2\) 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, \(SO_2\), PM, and \(NO_x\), were on-site measured, and VOCs were sampled and analyzed in laboratory aimed to obtain the pollutant EFs associated with coal consumptions, annual output, and product yield.

Overall the \(SO_2\) EFs dominated all the other three pollutants among all the three kinds of companies. The key influencing factors on the \(SO_2\) emissions were sulfur content of coal and \(SO_2\) RF 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 \(SO_2\) completely originated from burning of sulfur in coal, \(NO_x\) also could be formed.
Fig. 5. EFII values for 34 brick production factories.

Fig. 6. EFIII values for 34 brick production factories.
by oxidation of air N₂ at high combustion temperature, NOₓ EFᵢ values experienced minor fluctuations within the same industries compared to the other air pollutants. EFᵢ values for 4 air pollutants for brick factories dominated over the other two kinds of industries due to its low output. 3 of 8 pharmaceutical factories with SO₂ RFs (P2, P4, and P8) possessed SO₂ EFᵢ values of 3.00, 1.91 and 2.51 kg t⁻¹, and SO₂ EFᵢ values of 16.1, 86.3 and 5.01 kg MY⁻¹. The values of EFᵢ and EFᵢ in food 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 EFᵢ and EFᵢ 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 EFᵢ and EFᵢ 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. 715 kg MY⁻¹ without SO₂ RFs. In brick factories, the EFI and EFII values experienced minor fluctuations within the EFII 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 NOₓ EFᵢ in food factories was 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 EFᵢ and EFᵢ 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 NOₓ EFᵢ in food factories was 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 EFᵢ and EFᵢ 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 NOₓ EFᵢ in food factories was 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 EFᵢ and EFᵢ 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 NOₓ EFᵢ in food factories was 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 EFI and EFII values were 5.01 ± 2.36 kg t⁻¹ vs. 162 ± 194 kg MY⁻¹ in cement factories, NOₓ EFs were at the same level with those of LSIs such as CFPPs and cement factories, while their average value was 3.87 ± 2.35 kg t⁻¹. The corresponding PM EFᵢ and EFᵢ in food factories 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. The 1st food factories, and 0.55 ± 1.12 kg t⁻¹ vs. 0.84 ± 0.83 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 EFᵢ 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 EFᵢ and EFᵢ in food factories 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. 2nd and 6th pharmaceuticals factories possessed a prominent VOCs EFᵢ 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 EFᵢ in food and brick factories (in kg t⁻¹) were 4.83 ± 10.9 and 0.52 ± 0.92, and EFᵢ were 93.2 ± 184 kg MY⁻¹, 1.15 ± 0.89 t MY⁻¹ for pharmaceuticals, food, and brick companies, and the corresponding NOₓ EFᵢ values were 56.6 ± 34.3 kg MY⁻¹, 151 ± 223 kg MY⁻¹, and 2.74 ± 2.08 t MY⁻¹. In general, compared with LSIs, SO₂ EFs for these SSIs were much higher than those of LSIs such as CFPPs and cement factories, NOₓ 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

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