Research on Advanced Available Techniques of Air pollution Prevention and Control in Building Ceramics Industry in China

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Abstract. The flue gas emission of the building ceramics industry accounts for 85% of the whole ceramic industry in China, and it is the critical sector of air pollution control in the ceramic industry. The advanced available techniques (AAT) of air pollution prevention and control are of considerable significance to promote the upgrade of pollution prevention measures and technological progress. The paper studied and established the method of screening and evaluating the AAT for the prevention and control of air pollution in the building ceramics industry. Through collection and analysis of three normal pollutant emission concentrations of ten available techniques (AT), to select the AAT that meet at least one pollutant of all the plants was lower than 50% of the national standard emission concentration threshold in China. Based on the experience of the European Union and the comprehensive evaluation of industry associations, research institutions, and pollution control experts, five AAT were selected. The findings will provide an essential reference for the building ceramics industry to select advanced techniques, and provide significant technical support for the revision of the ceramic industrial pollutant emission standard for and selection of pollution prevention and control techniques (PPT and PCT) for pollutant discharge permission management.

1. Introduction
In recent years, China's building ceramics industry has developed rapidly. In 2016, the output reached 110.76 billion m^2, accounting for two-thirds of the world's total output [1]. While it has made significant contributions to the economy, it has also brought pressure on environmental protection and energy consumption. China's ceramic tile fume emissions account for about 85% of China's ceramic industry, and it is a crucial sector in the ceramic industry for air pollution control. Ceramic tile flue gas is generated in kiln and spray drying tower, and its primary pollutants are particulate matter, sulfur dioxide, and nitrogen oxides. Advanced available techniques (AAT) for the prevention and control of air pollution in ceramic tile have an essential role in mitigating the generation and discharge of pollutants from ceramic tile industrial plants and thereby reducing their impacts on the environment. In 2006, the European Union issued the "Reference Document on BAT in the Ceramic Manufacturing Industry" [2], which made high-level environmental protection of the ceramic industry in the most effective way. In 2018, the Ministry of Ecology and Environment of China issued the "Guideline on available techniques of pollution prevention and control for ceramics manufacturing industry" (HJ 2304-2018) [3], and recommended 10 available technique (AT) combinations, but did not recommend AAT which can
achieve higher level of environmental economic benefits. With China’s "13th Five-Year Plan for Ecological Environment Protection" [4], pollution prevention battle, pollutant permit system and other stricter requirements for air pollution emissions, screening a series of AAT for the protection and prevention of atmospheric pollution in the ceramics tile industry with higher environmental-economic benefits are of great significance to promote the ceramic tile plants upgrading pollution prevention and control measures and technological progress.

2. Overview of available techniques for air pollution control in the building ceramics industry

During the compilation process of HJ 2304-2018, through literature and research, expert discussions, field investigations, on-site tests, etc., ten technique combinations including prevention techniques, kiln gas treatment techniques, and spray drying tower gas treatment techniques were selected and specified [5]. Table 1 shows the AT for pollution Prevention and Control in building ceramics industry recommended in HJ 2304-2018. Among them, 1 to 7 are the AT for the treatment of kiln flue gas and spray drying tower gas respectively, and 8 to 10 are the combining treatment techniques for kiln flue gas and spray drying tower flue gas. These ten recommended techniques have continuous and stable operation cases in China.

3. Methods for screening and evaluation advanced available techniques

3.1. Research on screening methods

The AAT in HJ 2300-2018 is defined as “Among the available techniques for pollution prevention and control, the technique that makes the discharge of at least one major pollutant stably below 70% of the national pollutant emission standard limit” [6]. According to the survey, most of the ceramic tile plants in China can achieve this requirement by adopting AT. Therefore, the requirement of at least one pollutant below the standard limit of 70% is loose for Chinese building ceramics industry. According to this requirement, it is also impossible to screen out AAT for the building ceramics industry.

IPPC 96/61/CE [7] and Directive 2010/75/EC [8] issued by The European Union all mentioned Best Available Technique (BAT). "Best" means that the technique considered is the "most effective in achieving a generally high level of protection of the environment as a whole" [7], and introduced the BAT evaluation criteria, including the reduction of waste generation, the use of low-harm raw materials, scale applications, representing advanced technique and development direction. The selection and evaluation of BAT have strict procedures. The determination method is mainly through technical information exchange of government departments, non-government departments, industry associations, research institutions, etc., rather than quantitative screening methods.

Table 1. AT for pollution Prevention and Control in building ceramics industry [3].

| AT | Prevention Techniques | Kiln Gas Control Techniques | Spray drying tower Gas Control Techniques |
|----|-----------------------|-----------------------------|------------------------------------------|
| 1  | ①Raw material PCT (low sulfur materials) + ②Kiln gas PPT + ③Spray drying tower coal-water slurry hot blast stovel coa-based fuel high-temperature gypsum or sodium-alkali sulfur fixation technique | Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal | SNCR in hot air stovel + ② bag-type dust removal + ③ Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal |
| 2  | ①Raw material PCT (low sulfur materials) + ②Kiln gas PPT + ③Spray drying tower coal-water slurry hot blast stovel coa-based fuel high-temperature gypsum or sodium-alkali sulfur fixation + low nitrogen combustion) or generator gas collaborative dust removal hot blast stovel low nitrogen combustion technique | Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal | ① Bag-type dust removal + ② Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal |
| 3  | ①Raw material PCT (low sulfur materials) + ②Kiln gas PPT + ③Low-nitrogen combustion technique of gypsum or sodium-alkali methane hot blast stovel in spray drying tower | Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal | ① Bag-type dust removal + ② Spray-type dust removal |
### AT Prevention Techniques

| AT | Prevention Techniques | Kiln Gas Techniques | Control spray drying tower gas control techniques |
|----|-----------------------|--------------------|-----------------------------------------------|
| 4  | ① Raw material PCT (low sulfur materials) + ② Kiln gas PPT + ③ Spray drying tower pulverized coal chain type hot blast stove low sulfur fuel technique (low sulfur coal) + ④ Spray drying tower pulverized coal chain type hot blast stove coal-based fuel high-temperature sulfur fixation + ⑤ Spray drying tower pulverized coal chain type hot blast stove low nitrogen combustion | Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal | ① Bag-type dust removal + ② Spray-type dust removal |
| 5  | ① Raw material PCT (low sulfur materials) + ② Kiln gas PPT + ③ High-temperature sulfur-fixing technique for coal-based fuel in the coal-fired hot-blast stove of spray drying tower | Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal | ① Cyclone-type dust removal + ② Spray-type dust removal + ③ Wet electric-type dust removal |
| 6  | ① Raw material PCT (low sulfur materials) + ② Kiln gas PPT + ③ High-temperature sulfur-fixing technique for coal-based fuel in the coal-fired hot-blast stove of spray drying tower + Low-nitrogen combustion technique for the coal-fired hot-blast stove of spray drying tower | Kiln gas wet method for multi-pollutant collaborative control technique | ① Bag-type dust removal + ② Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal |
| 7  | ① Raw material PCT (low sulfur materials) + ② Kiln gas PPT + ③ High-temperature sulfur-fixing technique for coal-based fuel in the coal-fired hot-blast stove of spray drying tower + Low-nitrogen combustion technique for the coal-fired hot-blast stove of spray drying tower | Kiln gas wet method for multi-pollutant collaborative control technique | ① SNCR in hot air stove + ② bag-type dust removal + ③ Wet desulfurization (lime-gypsum or sodium-alkali) collaborative dust removal |
| 8  | ① Raw material PCT (low sulfur materials) + ② Kiln gas PPT + ③ Low-sulfur fuel technique for hot blast stove of spray drying tower (low-sulfur coal) + ④ High-temperature sulfur-fixing technique for high-temperature sulfur-fixing technique for the coal-fired hot-blast stove of spray drying tower for coal-based fuel | ① SNCR denitration in hot air stove of spray drying tower + ② Bag-type dust removal of spray drying tower + ③ Cooperative dust removal of kiln gas and spray drying tower gas wet desulfurization (lime-gypsum or sodium-alkali) | |
| 9  | ① Raw material PCT (low sulfur materials) + ② Kiln gas PPT + ③ Low-sulfur fuel technique for hot blast stove of spray drying tower (low-sulfur coal) + ④ High-temperature sulfur-fixing technique for high-temperature sulfur-fixing technique for the coal-fired hot-blast stove of spray drying tower for coal-based fuel | ① SNCR denitration in hot air stove of spray drying tower + ② Bag-type dust removal of spray drying tower + ③ Cooperative dust removal of kiln gas and spray drying tower gas wet desulfurization (lime-gypsum or sodium-alkali) + ④ Wet electric-type dust removal | |
| 10 | ① SNCR denitration in hot air stove of spray drying tower + ② Cyclone dust removal of spray drying tower gas + ③ Semi-dry desulfurization and desulfurization of kiln gas and spray drying tower gas circulating fluidized bed | ① SNCR denitration in hot air stove of spray drying tower + ② Cyclone dust removal of spray drying tower gas + ③ Semi-dry desulfurization and desulfurization of kiln gas and spray drying tower gas circulating fluidized bed | |

The Clean Air Act of 1990 issued by the United States stipulated that New Source Performance Standards (NSPS) should be established based on Best Demonstrated Technique (BDT) [8], and National Emissions Standard for Hazardous Air Pollutants (NESHAP) should be based on maximum available control technique (MACT) [9]. Among them, the screening of BDT is determined through a comprehensive assessment of reduction levels, implementation costs, and demonstration verification, and the screening of MACT is determined by counting the emission levels that can be achieved by various control techniques. The MACT of the new source is the technique with the lowest emission level that can be achieved by the same source. The MACT of the existing source is the technique that can reach the control level of the best 12% pollution source (more than 30 pollution sources) or the average pollution control techniques that the top 5 of similar pollution sources can reach for pollution sources less than 30.

At present, there are few studies on screening methods of AAT for pollution prevention in China, and there are no scientific, reasonable, objective, and fair evaluation index system and evaluation methods. According to HJ 2300-2018, determining the definition of AAT refers to the "Environmental Protection Tax Law of the People’s Republic of China", it said " If the concentration value is lower than 30% of the national and local pollutant emission standards, the environmental protection tax shall be reduced by 75%", indicating that emissions stably below 70% of emission standards. The "Environmental Protection Tax Law" aims to promote plants to reduce the concentration of pollutant emissions effectively. To truly reflect the most economical and practical level that AAT can achieve, the screening
research is still mainly based on the emission concentration levels that can be achieved by each technique. As the requirement of at least one pollutant lower than the standard limit of 70% is obviously loose for the building ceramics industry, this study refers to the "Environmental Protection Tax Law" which is "If the concentration value is lower than 50% of the national and local pollutant emission standards, the environmental protection tax shall be reduced by 50%.". 50% below the emission limit can theoretically effectively to reduce the emission concentration and has a prominent green leading role. Therefore, this study takes "the emission concentration of at least one major pollutant below 50% of the emission standard" as the AAT screening standards. At the same time, they are combined with the experience of the European Union, the comprehensive evaluation of industry associations, research institutions, and pollution control experts to select AAT better.

3.2. Evaluation Methodology
At present, China has not established a sound technical and economic evaluation system for AAT. Descriptive indicators for BAT assessment in the European Union and the United States mainly include indicators such as resource consumption, energy consumption, pollution emissions, economic costs, environmental effects, social impacts, and case analysis, etc.. The United States technical assessment is mainly based on whether the pollutant emissions meet the emission limit standards, and the EU’s indicator settings aim to be able to describe BAT parameters in detail. At present, China has carried out AT assessment studies for some key industries, including the Chinese thermal power industry [10], lead smelting industry [11], Vitamin C industry [12] and textile dyeing and finishing industry [13]. The technical evaluation of the study focuses on the four indicators of pollutant emissions, investment costs, economic costs, and technical-economic parameters. The operating costs include resource consumption and energy consumption. At the same time, the method of an expert-assisted comprehensive evaluation is adopted to compare and evaluate AAT comprehensively, and finally, screen out AAT that meet the status of industry development and the actual situation of the ceramic tile industry.

4. Selection of advanced available techniques

4.1. Analysis of Atmospheric Emission Data of Chinese Ceramic Tile plants
By collecting and analyzing the compliance monitoring data and technical cases of typical ceramic tile plants corresponding to ten available ceramic air PPT and PCT recommended by HJ 2304-2018. The concentration levels of particulate matter, sulfur dioxide and nitrogen oxides in the exhaust gas using various techniques are shown in Figure 1.

4.2. Selection of advanced available techniques
According to the screening method in 3.2, the number of plants and the proportions of particulate matter, sulfur dioxide, and nitrogen oxides for each AT, which are lower than 70% and 50% of the standard limits, can be counted. Table 2 shows the comparative analysis results of the emission concentrations of 10 AT and the standard emission concentration limits [14, 15]. All these ten techniques can meet the advanced available technical requirements of HJ 2300-2018, which "at least one major pollutant stably below 70% of the national pollutant emission standard limit". AT 3 and 9 can meet the emission concentration of the three pollutants of all plants below 50% of the standard emission limit. However, as for available technique 3, there are only two companies have collected emission monitoring data, and combined with a comprehensive evaluation by ceramics experts, whether this technique can stably fall below 50% of the standard emission limit requires further research and verification. Therefore, the available technique 9 screened out in this study has technologically advanced feasibility at the level of emission concentration and is the most advanced available technique. The AT 4, 5, 6, 7, 8, and 10 can meet the emission concentration of one pollutant of all plants below 50% of the standard emission limit. Combined with a comprehensive assessment of experts in the ceramic industry, the stability of AT 4, 6, and 7 also needs further study. Therefore, AT 5, 8, and 10 are selected as AAT. Besides, due to limited application plants, the SCR de-nitration technique is not included in the list of AT for pollution prevention and control of the ceramic manufacturing industry recommended by HJ 2300-2018. Still, this technique is mature in theory, and there are handful successful and stably cases in China. There are
at least 3 plants in China adopted "raw material control technique (low sulfur materials) + kiln gas pollution prevention technique + dry powder milling technique + SCR denitration technique". The desulfurization rate is greater than 98%, the de-nitration rate is greater than 90%, and the dust removal rate is greater than 99%. Kiln gas emission monitoring report shows that particulate matter emission concentration is lower than 10mg / m³, SO₂ emission concentration is lower than 20mg / m³, NOₓ emission concentration is lower than 50mg / m³, and the emission concentration of all three pollutants is lower than 50% of the standard emission limit. Also, the SCR technique worked stably, and it is worth promoting in the building ceramics industry. Therefore, this technique should be added as a "complementary available technique" in the list of AAT.
Figure 1. Plant Emission concentration level of ten available Techniques.

Table 2. Comparison of 10 AT emission concentration and the standard limit (SL).

| AT | Type of Pollution | SL (mg/m³) | Number of companies below 70% SL | Proportion (%) | Number of companies below 50% SL | Proportion (%) |
|----|-------------------|------------|----------------------------------|----------------|----------------------------------|----------------|
| 1  | PM                | 30         | 15                               | 78.95          | 12                               | 63.16          |
|    | SO₂               | 50         | 18                               | 94.74          | 18                               | 94.74          |
|    | NOₓ               | 180        | 19                               | 100            | 18                               | 94.74          |
|    | PM                | 30         | 19                               | 65.52          | 8                                | 27.59          |
| 2  | SO₂               | 50         | 28                               | 96.55          | 28                               | 96.55          |
|    | NOₓ               | 180        | 29                               | 100            | 26                               | 89.66          |
|    | PM                | 30         | 2                                 | 100            | 2                                | 100            |
| 3  | SO₂               | 50         | 2                                 | 100            | 2                                | 100            |
|    | NOₓ               | 180        | 2                                 | 100            | 2                                | 100            |
|    | PM                | 30         | 2                                 | 33.33          | 0                                | 0.00           |
| 4  | SO₂               | 50         | 5                                 | 83.33          | 4                                | 66.67          |
|    | NOₓ               | 180        | 6                                 | 100.00         | 6                                | 100.00         |
|    | PM                | 30         | 3                                 | 42.86          | 1                                | 14.29          |
| 5  | SO₂               | 50         | 6                                 | 85.71          | 3                                | 42.86          |
|    | NOₓ               | 180        | 7                                 | 100.00         | 7                                | 100.00         |
|    | PM                | 30         | 6                                 | 100.00         | 6                                | 100.00         |
| 6 and 7 | SO₂         | 50         | 4                                 | 83.33          | 4                                | 66.67          |
|    | NOₓ               | 180        | 5                                 | 83.33          | 4                                | 66.67          |
|    | PM                | 30         | 7                                 | 87.50          | 6                                | 75.00          |
| 8  | SO₂               | 50         | 8                                 | 100.00         | 6                                | 75.00          |

Note: The X-axis of the graph (a) to (i) is different from each other because the number of plants applied to available technology (AT) 1 to 10 is different.
5. Technical and economic feasibility assessment

Through the collection of data on investment costs, operating costs, technical-economic parameters of AAT selected previously, and combined with relevant expert consultation and comprehensive evaluation, the technical-economic feasibility of AAT are evaluated. The evaluation results are shown in the table 3. As can be seen from Table 3, the investment cost and operating cost of the available technique 9 with the lowest emission concentration level is significantly higher than other techniques, while the second-AAT 5 has the lowest investment cost and more economical operating cost although the emission level is higher than other techniques. Also, "complementary AT" have the most economical operating costs. Plants can make reasonable choices based on local environmental management needs, pollutant discharge targets, and affordable economic costs.

Table 3. Statistical table of economic evaluation of AAT.

| AT    | Investment costs (million RMB) | Operating costs (RMB/day) | Technical-economic parameters (m³/h) |
|-------|-------------------------------|--------------------------|-------------------------------------|
| 9     | 13                            | 10600                    | 580000                              |
| 8     | 8                             | 6000                     | 580000                              |
| 10    | 12                            | 6000                     | 580000                              |
| 5     | 2.81                          | 3253.7                   | 129882                              |
| Complementary | 4.20                          | 1400                     | 250000                              |

6. Conclusions

a) Based on the corresponding emission concentration level requirements of the "Environmental Protection Tax Law" for corporate tax reduction and exemption, from the 10 AT recommended by HJ2304-2018, select at least one pollutant that meets all plants' stable emission concentrations below 50 % of the standard limits, and draw on the EU experience combined with the comprehensive evaluation of industry associations, research institutions, and pollution control experts to screen out five AAT.

b) Based on the indicators of pollutant discharge, investment cost, economic cost, and technical-economic parameters, the technical-economic feasibility of 5 AAT are evaluated. The results show that the investment and operating costs of available technique 9 with the most substantial reduction in emissions are significantly higher than other techniques, while available technique 5 has the lowest investment costs and lower operating costs despite higher emissions levels than other techniques.

c) Plants could choose the appropriate technique according to their own needs. However, legal monitoring data is limited, and it still needs further research and verification as for long-term stability.

d) While technical upgrading, ceramic tile plants also need to improve operation and management measures, improve technical operation capabilities, and make the treatment of air pollutant emissions more effective.
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