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Improving Flue Gas Mercury Removal in Waste Incinerators by Optimization of Carbon Injection Rate

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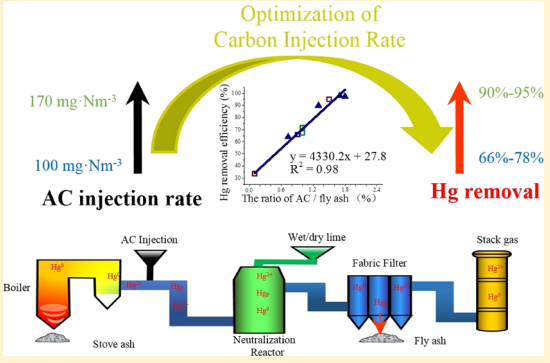
Supporting Information

ABSTRACT: This study tested the mercury emission characteristics of six municipal solid waste incinerators (MSWIs) and recommended future mercury control via adjusting operational parameters. The results indicated that over 99% of the mercury in solid wastes ended in fly ash and flue gas, of which 3.3−66.3% was emitted to air through stack gas. Mercury in the stack gas was mainly in the form of oxidized mercury (Hg^{2+}), the proportion (65.4−89.0%) of which was far higher than previous estimation (15%). Mercury removal efficiencies (MRE) of the tested incinerators were in the range of 33.6−95.2%. The impact of waste incineration capacity, gas flow, fly ash yield, and activated carbon (AC) injection on MRE were analyzed. We found that the MRE was significantly linearly correlated to the ratio of AC injection and fly ash yield (correlation coefficient = 0.98, significance <0.01). AC injection value is determined based on the control of dioxin emissions without considering mercury control in traditional design. To increase MRE of MSWIs, the AC injection should increase from around 100 mg·Nm^{-3} to 135 mg·Nm^{-3} for grate furnace combustor and 170 mg·Nm^{-3} for circulation fluidized bed combustor, so as to reach a MRE of 90%.



1. INTRODUCTION

Mercury, because of its high toxicity to humans and the environment, has attracted a great deal of attention in recent years.[1] The municipal solid wastes incineration is an important anthropogenic mercury emission source in the world and also one of the five key controlled mercury emission sources in the *Minamata Convention on Mercury*.[2,3] Therefore, mercury emissions characteristics and reduction measures of MSWIs have been a research hotspot.

China is the largest anthropogenic mercury emission country in the world[4] and the emissions from municipal solid waste incinerators (MSWIs) gradually catch attention due to the dramatic increase of wastes incineration amounts.[4−6] The incineration amount of municipal solid waste has increased from 11.4 million tons in 2006 to 61.76 million tons in 2015 with the rate of 21%. It was expected that the waste incineration capacity will increase to 231.35 million tons in 2020 based on *The 13th Five-Year Plan for the Facility Construction of Municipal Waste Nonhazardous Treatment in China*.[7] The rapid increase of waste incineration amounts may lead to the increment of mercury emission in MSWI.[3,4]

To control mercury emissions, it is important to improve the mercury removal efficiency (MRE) of air pollution control devices (APCDs) by either upgrading the APCDs or adjusting operation parameters.[8,9] Kim et al., (2010) indicated that the MRE of water spray tower and fabric filter were 54% and 41%, respectively.[9] Fumitake's study showed that the average MRE increased from 34.5% to 92.5% when the electrostatic precipitator was replaced by fabric filters for dioxin control.[8] In China, above 90% of waste incinerators install typical APCDs combination of dry/semidry scrubbing system + activated carbon injection + fabric filter. This method is one of advanced APCD combinations in the world and it will continue to be used in the coming years in China. However, this APCD combination is primarily to control the dioxin emissions and synergistic mercury removal is not one assessment index during the design of the incinerators' operation parameters. Therefore, although unintentional synergistic mercury removal were observed in the MSWIs, the MRE had obvious differences (from 5.7% to 100%) in waste incinerators.[10,11] Zhang et al. (2016) found that the

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overall mercury removal efficiency of the APCDs for MSWIs ranged from 60% to over 99%.[12,13] The large variation of mercury removal efficiencies of current APCD combinations may attribute to many factors, such as the ingredient of wastes and operating parameters. For a specific incinerator, the ingredient of wastes were relative stable so as to meet the incineration requirement. Therefore, the MRE of APCDs in MSWI is quite possible to be attributed to the operating parameters, such as burner type, waste incineration capacity, gas flow, lime injection, AC injection, etc. To better control mercury emission from waste incineration, optimizing operation parameters of APCDs for mercury control in MSWIs is worthy researching.

In this study, field tests of mercury emission characteristics were conducted in six waste incinerators. Based on the test results, potential impact factors (waste amount, gas flow, fly ash yield, and activated carbon (AC) injection) were analyzed and we established the relationship between mercury removal efficiency and AC injection. Such kind of quantitative relationship can be used to control mercury emissions in MSWIs.

2. MATERIALS AND METHODS

2.1. Testing Waste Incinerators. Detailed introduction of the six tested incinerators (denoted as A−F) was described in Supporting Information (SI) Table S1. The A and B plants used circulation fluidized bed combustors (CFBC); C−E plants used grate furnace combustor (GFC); F plant used pyrolyzing furnace (PF). The three incineration methods accounted for above 95% of waste incineration field in China. All incineration plants had the same APCDs combination: dry/semidry scrubbing system for acidic gases, activated carbon injection, and fabric filter (SI Figure S1). Other information (furnace type, waste incineration amount, temperature, AC injection quantity and fly ash/bottom ash yield, etc.) of the tested plants were collected in SI Table S1.

2.2. Sample and Analysis Techniques. *2.2.1. Solid Sampling and Analysis.* During the monitoring period, solid samples, including incinerated wastes, bottom ash and fly ash, were sampled three times per day at all incinerators. Based on the *Sampling and Physical Analysis Method of Municipal Domestic Waste* (CJ/T313−2009), the sampled wastes were classified into nine categories (food, paper, rubber/plastic, weave, wood/bamboo, brick/ceramics, glass, metal, and fine mixture). After other eight categories were screened out, the remaining waste with particle diameter less than 10 mm was defined as fine mixture. Then each category of waste was dried and weighed. The mercury content of solid samples were analyzed by Lumex 915 M + PYRO (Lumex Instruments Company, Russia). The detection limit of the system is 2 $ng \cdot g^{-1}$.

2.2.2. Flue Gas Sampling and Analysis. The flue gases before and after pollution control facilities were sampled 3 times per day with the Ontario Hydro Method (OH method).[14] The adsorption liquid was measured via mercury analyzer (F732 V, Shuangxu Electron Company, China). The detection limit of mercury analyzer is 0.05 $\mu g \cdot L^{-1}$.

The tested result was used to calculate the MRE:

$$\mathrm{MRE} = \frac{C_A - C_B}{C_A} \times 100\% \quad (1)$$

where C_A is the mercury concentration of flue gas before the APCDs, $\mu g \cdot Nm^{-3}$; C_B is the mercury concentration of flue gas after the APCDs, $\mu g \cdot Nm^{-3}$.

2.2.3. Quality Assessment and Quality Control (QA/QC). All data of samples is the average value of at least six parallel samples. Every sampling of flue gas was conducted over 500−1500 L to ensure the representative. Each sample was analyzed three times, and the error caused by the analysis procedures was less than 5%. The more information on QA/QC was in SI section S1.

3. RESULTS AND DISCUSSION

3.1. Component Analysis and Mercury Input of MSW Plants. SI Table S2 showed the dry/wet weight percentage of the nine waste categories in the tested plants. The food, paper, rubber, and plastic were the main components of the wastes, accounting for 48.1 wt %-77.6 wt % of the waste in A−D plants.. The fine mixture in A and B waste was much higher than that in other plants. This led to the more fly ash formation during incineration. Therefore, the fly ash yield of A and B plants was higher than that of other plants (SI Table S1).

As shown in Figure 1, the mercury concentrations (1.07−3.72 $mg \cdot kg^{-1}$) of the fine mixture were distinctly higher than

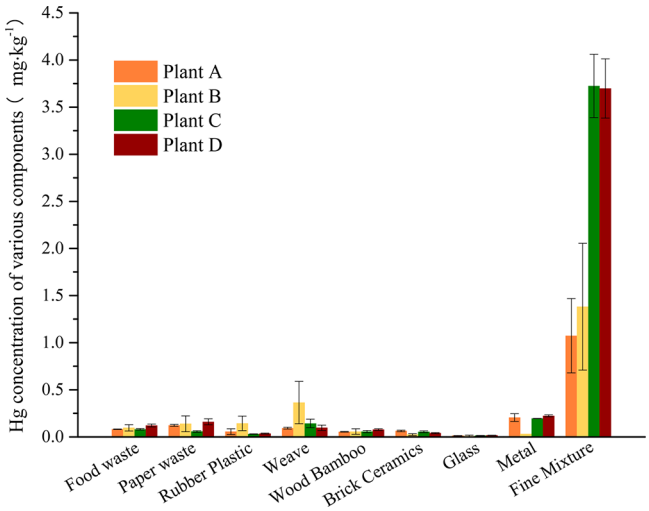

Figure 1. Mercury concentrations of nine waste categories in four plants.

that of other eight categories wastes (0.01−0.36 $mg \cdot kg^{-1}$) in the four waste plants. The other eight components showed similar mercury concentrations in the four plants. The average mercury concentrations in the waste of A, B, C, and D plants are 0.273 ± 0.084, 0.345 ± 0.177, 0.372 ± 0.036, and 0.566 ± 0.053 $mg \cdot kg^{-1}$, respectively. This result is lower than the data of previous studies that the range of average mercury concentration of wastes is from1.8 $mg \cdot kg^{-1}$ in 1995 to 0.5 $mg \cdot kg^{-1}$ in 2009.[6,15]

In 2009, batteries still predominated as the major mercury source, contributing approximately 54% of the total mercury in MSWIs, with fluorescent lamps accounting for 21%.[15] The contribution of battery and fluorescent lamp was gradually reduced in wastes. This phenomenon is attributed to two aspects: (1) The battery and fluorescent lamp are partly collected and recycled in the upstream of waste treatment in China. (2) Mercury-containing battery and fluorescent lamp are gradually replaced by nonmercury battery and light emitting

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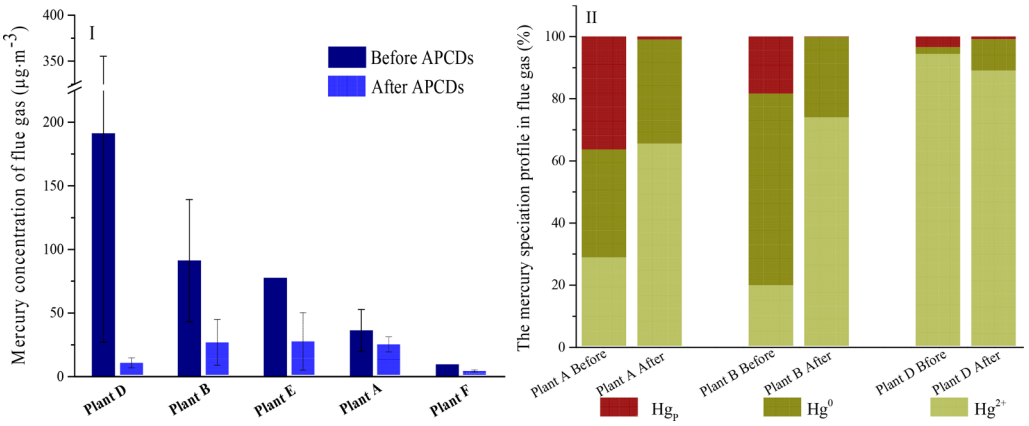

Figure 2. Mercury concentrations and speciation profile of the flue gas, (I) mercury concentrations (the data of E and F before APCDs originates from material balance calculation), (II) mercury speciation profile (before represents before APCDs, after represents after APCDs).

diode (LED) lamp for meeting implementation requirements of the *Minamata Convention on Mercury* and *Cleaner Production Program for Battery Industry*.[16]

Mercury inputs in tested waste incinerators were shown in SI Figure S2 and Table S3. In the five plants, mercury input mainly depended on mercury input from waste. The mercury input percentage reached around 100% for GFC and above 90% for CFBC (10% mercury from coal). In the wastes, the percentage of Hg input from fine mixture, serving as largest mercury contributor, reached 61.4−84.3% (SI Figure S2). The food, paper, rubber, and plastic were also relative big mercury contributors since they were the major components of wastes (Figure 1).

3.2. Mercury Output Analysis. *3.2.1. Mercury Concentrations in Flue Gas.* As shown in Figure 2(I), the differences of mercury concentrations before and after APCDs distinctly varied among plants. The order (plant D > plant B > plant A) of mercury concentrations in the flue gas before APCD combinations is consistent with that of mercury concentrations in the wastes in Figure 1. After APCDs, the mercury content of the flue gas obviously decrease to be lower than 26.4 $\mu g \cdot m^{-3}$ in all plants. Mercury speciation profile in flue gas before and after APCDs in A, B, and D plants is shown in Figure 2(II). The percentages of particle-bound mercury (Hg_P) in the flue gas of plant A (18.5%) and plant B (36.5%) were much higher than that in plant D (2.2%) before APCDs, which was mainly attributed to higher particular content in the flue gas of CFBC in A and B plants. The higher particular content of CFBC guaranteed the exposure opportunity of mercury compound to particular matter, resulting in the form transformation from more mercury compounds into Hg_P. Thereby, the percentage of Hg_P was higher in flue gas of A and B plants. The Hg^{2+} was the dominate form in flue gas of D−F plants before APCDs. In GFC technology, the excess air coefficient reaches 90−100%, which is higher than that of CFBC technology (60−70%). The excess air coefficient offers oxygen-enriched environment, resulting in mercury form transformation from mercury compound into Hg^{2+}. Therefore, the Hg^{2+} percentage of flue gas is higher in GFC technology than that in CFBC technology.

3.2.2. Mercury Output Analysis. The mercury outputs and output percentage of different materials in the tested MSWIs were shown in SI Table S4. The mercury concentrations of the fly ash in plant A (16.0 $\mu g \cdot g^{-1}$) and plant B (17.1 $\mu g \cdot g^{-1}$) were obviously lower than that in D, E, and F plants (119−268 $\mu g \cdot g^{-1}$). According to SI Table S1, the CFBC technology produces higher fly ash yield ratio (a) in A-B plants (4.3%−10.0%) compared to the GFC technology in D−F plants (0.8−3.5%). The high yield ratio of fly ash dilute mercury content in A, B plants, giving rise to the low mercury content. The mercury contents of bottom ash were 20.0−138 $ng \cdot g^{-1}$ in five plants, which was far lower than that of fly ash.

As shown in Figure 3, the bottom ash output percentage was so small (0.1−0.8%) in all plants that it could be neglected in

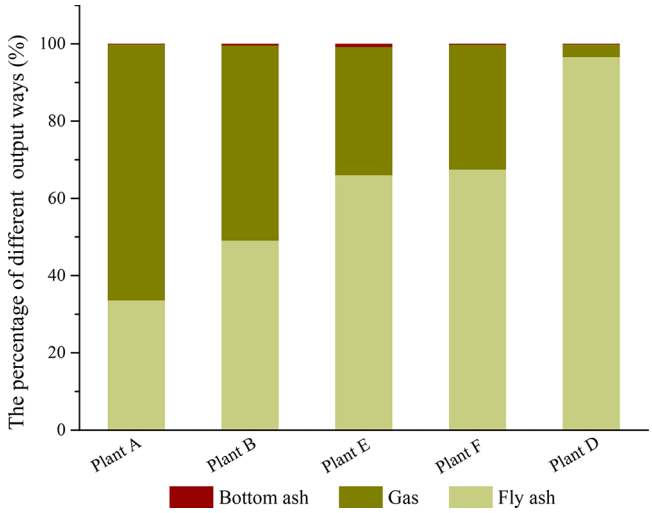

Figure 3. Percentage of different output ways in various plants.

output calculation. This indicated that the mercury compounds in wastes were almost completely released into flue gas in incineration processing. The mercury in A and B plants was mainly emitted via flue gas and their percentage was 66.3% and 50.5%, respectively. This could be explained that CFBC condition generates high percentage of Hg^0 based on Figure 2(II), which was more difficult than Hg^{2+} to adsorb on AC or fly ash.[17,18] In E, F, and D plants, the fly ash was the main output approach and the percentage was 66.0%, 67.5%, and 96.6% respectively.

3.3. Quantifying the Effect of Parameters Adjustment on MRE. Though all plants install the same APCDs, the MREs of the five plants showed obvious differences (33.6%, 71.9%, 95.2%, 67.5%, and 66.0%, respectively). The differences of MRE were possibly attributed to different operating parameters.[11,17,19,20] In the APCD combinations (dry/semidry

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scrubbing system + AC injection + fabric filter), adjustable parameters contains lime injection, AC injection, etc. The lime injection point is usually located in front of AC injection point. The acid gas (HCl, NO, SO_2, etc.) is removed before AC injection in case they influence AC adsorption property on dioxin. In previous study, the difference of MRE resulting from lime injection in dry or semidry scrubbing system was proved to be low 1% since inorganic material (lime) has poor adsorption capacity for mercury.[8,21,22] Moreover, the MRE of lime injection is relatively low (<22%), which is hard to determine the overall MRE (33.6−95.2%) of current APCD combinations. Instead, previous studies proved that the AC injection performed excellent adsorption capacity for mercury since the gas mercury could transform into Hg_P on physical and chemical adsorption sites of AC.[23−25] Then the particulate matter is captured via subsequent fabric filter. Therefore, the correlation of the MRE and AC injection amount was calculated, fitted and shown in SI Figure S3. The correlation coefficient (R^2) is 0.16, indicated that other operating parameters (e.g., waste incineration capacity, gas flow, fly ash yield) may also influence the mercury removal efficiency of APCDs. This study aimed to research the probability of quantitative control MRE via adjusting these operating parameters. The analysis of operating parameters on mercury capture via AC is shown in SI Figure S4, Figure S5 and Figure 4.

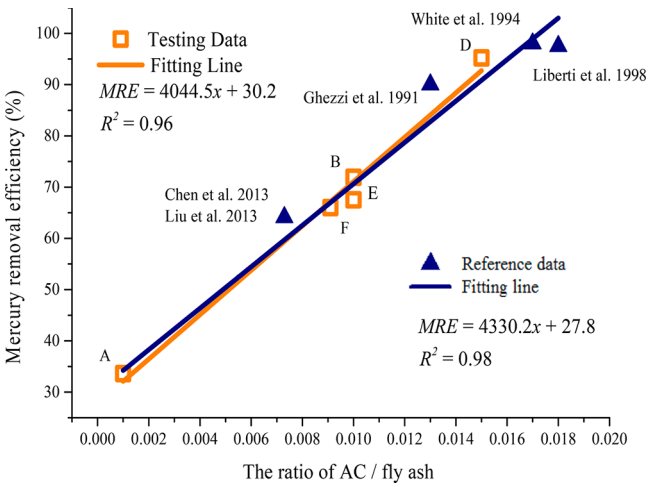

Figure 4. Correlation analysis between amount ratio of injected AC to fly ash yield and mercury removal efficiency (Orange data points of A−F origins from this study and orange line is the fitting result of A−F data; Navy data points origin from references and the navy line is the fitting result of A−F data and reference data).

First, according to SI Table S3, approximately 90.9−99.9% of mercury input originated from waste. Hence, mercury emission amount is positively correlated with waste incineration capacity if the mercury content is relatively consistent. The mercury removal mainly depends on AC capture, so AC injection capacity per unit mass of wastes possibly has positively related with the MRE. As shown in SI Figure S4, the correlation was calculated between the MRE and the ratio of AC injection amount to waste incineration capacity per day (AC/waste ratio). The result showed that the correlation coefficient was 0.19, indicating AC/waste ratio is not key factor determining mercury removal.

Second, the gas flow influences the AC concentration in flue gas. Higher AC concentration of flue gas increases the collision probability between AC and mercury, beneficial to the mercury capture. As a result, the AC concentration possibly influences the MRE. In SI Figure S5, the correlation coefficient between the MRE and ratio of AC injection amount to gas flow (AC/gas ratio) was relatively low and only 0.41. The AC/gas ratio of B and D plants was similar (146 and 156 mg·Nm^{3-}), but the MRE showed obvious gap (71.9% and 95.2%) in SI Table S5 and Figure S5. This manifested that AC/gas ratio and mercury removal have certain correlation, yet the AC/gas ratio was not the main factor influencing mercury removal.

Third, fly ash yield is also possibly an important influence factor. Fly ash can cover physical and chemical adsorption sites on surface of AC, resulting in the decrease of mercury adsorption capacity.[26] Thereby, the effect of fly ash yield is explored in this study as well. As shown in Figure 4, the MRE and the ratio of AC injection amount to fly ash yield per day (AC/fly ash ratio) exhibited very good correlation with a correlation coefficient of 0.96. The data used for fitting analysis contained the tested data inA, B, D, E, and F plants, the furnace type includes CFBC, GFC, and PF, and the waste includes MSW and medical waste. These results indicated that the correlation of MRE and AC/fly ash ratio can apply to various furnaces and wastes. The fitting formula is shown in Figure 4.

Moreover, the results with same APCDs in literature were also researched. The data are marked in navy in Figure 4.[10,27−30] These waste incineration plants have same APCDs combination (lime slurry + activated carbon + FF). Chen et al., (2013)[10,30] tested the waste incineration plant in Zhongshan and found that the MRE was 64.1% and the AC/fly ash ratio was 0.0073. Researches in Italy, the U.S., and testing results indicated that the AC/fly ash ratio was similar 0.013−0.018 and the MRE was in the range of 90−98%.[27−29] These data were added to fit correlation line in navy curve in Figure 4. The data from literature has good correlation (R^2 = 0.98) with the navy fitted curve. The result indicated that the calculation method has also good application in other plants. The fitting formula was shown as follows.

$$\mathrm{MRE} = 4330.2x + 27.8 \quad (2)$$

where the MRE represented the mercury removal efficiency, %. The x represents the ratio of AC injection amount to fly ash yield per day (AC/fly ash ratio). The slope and intercept of two formulas was very close and the average deviation of them was only 3.5% and 4.3%, respectively.

The reason for good fitting could be explained as follows. First, according to above analysis, the crucial factor of mercury removal is the form transformation from gas mercury to Hg_P in existing APCDs of China. AC injection can adsorb Hg^0 and Hg^{2+} on the surface and then be removed by subsequent fabric filter.[1,31,32] Hence, the AC injection amount is key factor influencing mercury removal efficiency. The mercury removal efficiency (MRE) is directly proportional to the AC injection amount. The ACI can be calculated based on the product of AC concentration and gas flow. The formula can be conversed to formula 3.

$$\mathrm{MRE} = 4330.2 \times \frac{\mathrm{ACI}}{\mathrm{FAY}} + 27.8 = 4330.2 \times \frac{b \times \mathrm{GF}}{\mathrm{FAY}} + 27.8 \quad (3)$$

where the ACI represents AC injection amount, t·d^{-1}; FAY represents the fly ash yield, t·d^{-1}; b represents the AC

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concentration of flow gas, mg·Nm^{-3}; the GF represents the gas flow, Nm^3·d^{-1}.

Second, fly ash can influence the mercury adsorption on sorbents. Although fly ash shows certain mercury adsorption capacity, it has negative effect on the mercury adsorption of carbon-based sorbents since the ash covers adsorption sites when the concentration of fly ash is high in flue gas. Previous studies proved that fly ash can decrease pore volume and cover the adsorption sites, resulting in the decline of mercury removal efficiency.[23,26,33] It indicates that the high concentration of fly ash hinders the adsorption capacity of AC.

Third, fly ash yield is calculated based on the product of waste incineration capacity and fly ash yield ratio (a). The formula 3 can be conversed to formula 4.

$$\mathrm{MRE} = 4330.2 \times \frac{b \times \mathrm{GC}}{a \times \mathrm{WIC}} + 27.8 \quad (4)$$

where WIC represents the waste incineration capacity, t·d^{-1}; the a represents the ratio of fly ash yield per unit mass of wastes. When the a is ascertained, the fly ash yield have positive correlation with WIC.[10,30] Meanwhile, mercury input is positively correlated with WIC since above 90% mercury input originated from wastes. Therefore, the fly ash yield is positively related with mercury input. The fly ash yield is inversely proportional to AC consumption capacity and MRE.

For specific municipal solid waste plant, the a, GF and WIC is ascertained. According to formula 4, the mercury removal efficiency (MRE) depends on the AC concentration of flow gas (b). Therefore, the MRE can be increased via increasing the carbon injection rate.

3.4. Synergic Control of Mercury and Dioxin Emission.

The formula 4 can applied to the design of new waste incinerator. The GF, WIC, a and b should be considered to obtain desirable MRE. However, the mercury control was out of the scope of design and construction of MSWIs in the past. The target of AC injection is primarily dioxin. When the design of waste incinerator is finished, the GF and WIC, serving as relative constant operating parameters, is difficult to change. If the waste components do not change significantly, the a is also usually constant. Therefore, the b, serving as only parameter, is adjusted to increase the MRE.

In previous studies, Tejima et al.(1996) found that the FF removed 97−98% dioxin and reduced to less than 0.1 ng toxic equivalency quantity (TEQ)·m^{-3} when AC was injected to the flue gas.[34] The similar result was also seen in the Chi's study that 100 mg·Nm^{-3} AC injection decreased the dioxin to 0.03 ng TEQ·m^{-3}.[35] Abad et al.(2003) proved that the 100 mg·Nm^{-3} AC injection could meet the emission limitation value (0.1 ng TEQ·m^{-3}) with the APCDs combination (Lime slurry + AC + FF).[36] Chang et al.(2009) found that the AC injection value from 50 mg·Nm^{-3} to 100 mg·Nm^{-3} increased dioxin removal efficiency from 60% to 90%.[37] Moreover, when AC injection reached 150 mg·Nm^{-3}, the dioxin removal efficiency reached 95% and tend to be stable. According to above studies, the 100 mg·Nm^{-3} AC injection can meet new *Standard for Pollution Control on the Municipal Solid Waste Incineration* in China.[38] Therefore, the 100 mg·Nm^{-3} AC injection is chosen as design value for dioxin removal. But this value does not consider the mercury removal. On the basis of this study, the mercury removal efficiency conformed to formula 4.

The fly ash yield ratio (a) and GF/WIC have crucial role on mercury removal efficiency. SI Table S6 gathered the data from 10 waste plants, including five tested data and five reference data. According to SI Table S6, the average a and GF/WIC are 0.031 and 3600 N m^3·t^{-1} for GFC and 0.072 and 6400 N m^3·t^{-1} for CFBC. When the b is 100 mg·Nm^{-3}, the MRE of CFBC and GFC was 66.3% and 78.0%, respectively. Under such circumstance, the mercury emission concentration of A-F plants was 3.1−26.4 μg·m^{-3}, most of which was higher than 0.05−4.56 μg·m^{-3} in Japan, 1.96−4.71 μg·m^{-3} in Korea, and 3.7 μg·m^{-3} in the U.S.[39−41] If China will further tighten the emission standard in future, the MRE is necessary to increase. When the MRE increases to 90%, the average AC injection value of should rise to 170 mg·Nm^{-3} for CFBC and 135 mg·Nm^{-3} for GFC, respectively on the basis of formula 4. Although average AC injection value was calculated for desired MRE, the b can be recalculated for specific MSWI based on formula 4. The fitting result can applied to the MSWI design and to reduce mercury emission from existing APCDs combination.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b05560.

S1, Quality assurance and quality control (Figure S1, Table S1); S2, Mercury content in nine categories wastes (Figure S2, Table S2−S5); S3, Correlation analysis between operating parameters and MRE (Figure S3−S7); S4, The data summary of fly ash yield ratio and FG/WIC from references (Table S6−S7) (PDF)

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Notes

The authors declare no competing financial interest.

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