Analysis and comparison of mercury-containing byproduct and estimation of mercury release from the industrial coal-fired boiler

Huang Kuangwei[1], Arizono Koji[2], Yakushiji Yuka[2], Ishibashi Yasuhiro[2]

[1]Graduate School of Environmental & Symbiotic Science, Prefectural University of Kumamoto, Kumamoto, Japan, 862-8505, hkw801103@gmail.com
[2]Faculty of Environmental & Symbiotic, Prefectural University of Kumamoto, Kumamoto, Japan, 862-8505

Introduction

Mercury (Hg) is one of the most volatile elements and environmental pollutants that has caused the world concern because of the toxicity, long distance transport, bioaccumulation, persistence and degradability in the environment. One of the most important content is to control the amount of mercury discharged into the environment caused by human factors. Coal-fired power plant is not only the largest coal consumer but also a primary pollution source of anthropogenic mercury emission [1-4]. Hg-release from coal combustion facilities depend on the coal type, boiler type, and what kind of air pollution control devices (APCDs) configuration and so on [5]. Current study has almost exclusively focused on atmospheric mercury emission from coal combustion processes [6]. Accompanied by the improvement of the APCDs efficiencies, the mercury emission can be obviously decrease in coal-fired power facilities. Therefore, more proportion of mercury emitted from coal-fired power facilities is in the recovered byproducts, such as fly ash, sludge, wastewater and desulfurization gypsum. The mercury content remained in these byproducts are usually much higher than that exhausted from flue gas. The bioavailability and leaching ability of mercury are related with its speciation and finally decide the total release amount of mercury from coal-fired power facilities. Therefore, it is important to understand the speciation of mercury in the byproducts. In this study, the fate of Hg including its removal by APCDs was quantified by collecting and analyzing solid samples such as coal, fly ash, bottom ash. The main purpose as below: (1) analysis of mercury concentrations in various byproducts, (2) estimate mercury emission from the industrial coal-fired boiler by substance flow analysis.

Materials and methods

The mercury-containing byproducts and substance flow analysis of mercury were studied at the circulating fluidized bed (CFB) boiler which is provide the electricity power for paper manufacture. The onsite tested boiler capacity power is 90 t/h. APCDs of the boiler fitted with ESP and bag dust collector to control the emission of particulate matter (PM) and NOx. The FGD or WFGD are typical devices to control SOx emission depend on limestone-gypsum but the tested boiler doesn't install the related device. The substitute way was used limestone powder as an in-furnace desulphurization agent during the combustion process. Sampling products include coal, limestone, bottom ash, electrostatic precipitator (ESP) ash, bag dust collector ash. All the samples were crushed by agate mortar and pass through the sieve (ø = 0.15mm) after the process, and mercury concentration was analysised by fully-automatic thermal-vaporization mercury analysis system (MA-3000). The repetitions were conducted at least three times for each sample. The mercury measurement uses the principle of thermal decomposition, gold amalgam collection, and cold-vapor atomic absorption spectroscopy (wave-length 253.7 nm). The samples were

collected into sampling boats to measure the Hg contents. We confirmed 0.1 ng Hg as the detection limit of MA-3000 in this experiment. Fig. 1 shows the schematic configuration of the APCDs, the CFB, and the sampling points.

Substance flow analysis (SFA) is a systematic assessment of the flows and stocks of materials within a system defined in space and time [7]. The purpose is to analyze the flow of substance related with human activities such as the resource extraction, usage and disposal of the process. Four main steps which are goal and system definition, data acquisition and inventory, substance balances and modeling, and interpretation were utilized and shown in Fig. 2. In this study, the results were obtained by the paper company, experimental data and using the following Eq. (1) to calculate:

$$F_{Hg} = F_{Hg\ coal} + F_{Hg\ limestone} = F_{Hg\ bottom\ ash} + F_{Hg\ ESP\ ash} + F_{Hg\ bag\ dust\ collector\ ash} + F_{Hg\ stack} \qquad (eq.1)$$

where F_{Hg} is the total flow in the system; $F_{Hg\ coal}$ is the total amount of mercury in coal; $F_{Hg\ limestone}$ is the total amount of mercury in limestone; $F_{Hg\ bottom\ ash}$ is the total amount of mercury in bottom ash; $F_{Hg\ ESP\ ash}$ is the total amount of mercury in ESP ash; $F_{Hg\ bag\ dust\ collector\ ash}$ is the total amount of mercury in bag dust collector ash; $F_{Hg\ stack}$ is the total amount of mercury emitted from flue gas. All the amount of mercury in items are used, produced and emitted in 2016.

Results and discussion

In the amount of mercury in ESP ash and bag dust collector ash, we can't obtain the amount of produced respectively. Because the outlet of ESP and bag dust collector are together not independent. Consequently, when the paper company records the amount of ESP ash and bag dust collector ash, the data is represented mix amount. Table.1 presents the Hg concentration in combustion byproducts and limit values for soil proposed by the State Technical Supervision Bureau and National Environmental Protection Agency of China [8]. As can be seen that the concentration of ESP ash is the highest. The ash concentration between ESP and bag dust collector have a significant decrease (497.69 µg/kg → 121.61 µg/kg). In 2016, the consumption of coal, limestone powder, bottom ash, fly ash in the industrial coal-fired boiler is 93,738 tons, 10,950 tons, 9,454 tons and 12,401 tons, and the total mercury amount of coal, limestone powder, bottom ash, fly ash, flue gas produce is 3.64 kg, 0.88 kg, 0.06 kg, 4.20 kg, 0.26 kg based on the mercury quality estimation method. Although limestone powder is an effective in-furnace desulphurization agent during the combustion process. However, itself contains more mercury than coal. This is another issue that can be discuss. The application of APCDs can significantly increase the production of combustion byproducts. During the coal combustion process, a large portion of Hg can be removed by APCDs and then be transformed into solid or liquid combustion byproducts. The paper company sold byproducts to the cement industry and cement industry use byproducts as material to make cement. Therefore, the reutilization of fly ash in applications such as cement and wall board production, can cause Hg release to the atmosphere again during any calcining process [9-10]. As a result, the disposition of combustion byproducts (e.g., landfilling) and reutilization (e.g., construction) should be paid more attention.
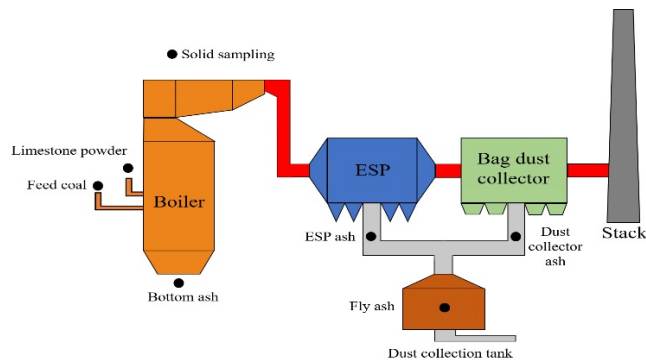
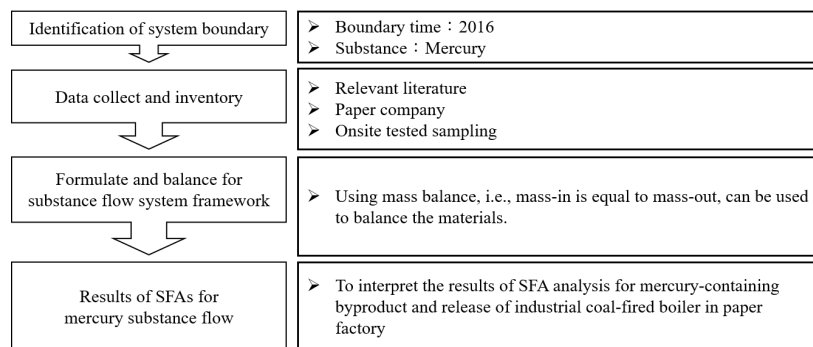Fig. 1: Schematic of sampling points across APCDs.

Fig. 2: SFA framework for mercury in industrial coal-fired boiler in 2016

Tab. 1: Hg concentration in combustion byproducts and its consumption, production volume

Flow	Input		Output			
Item	Coal	Limestone powder	Bottom ash	ESP ash	Bag dust collector ash	Fly ash
Concentration (μg/kg)	37.68	85.99	6.76	497.69	121.61	348.75
Consumption (ton)	93,738	10,950	-	-	-	-
Production volume (ton)	-	-	9,454	-	-	12,401
Limit value			500[a]			500[a]

[a]Environmental quality standard for soils (secondary-level standard), GB15618-1995
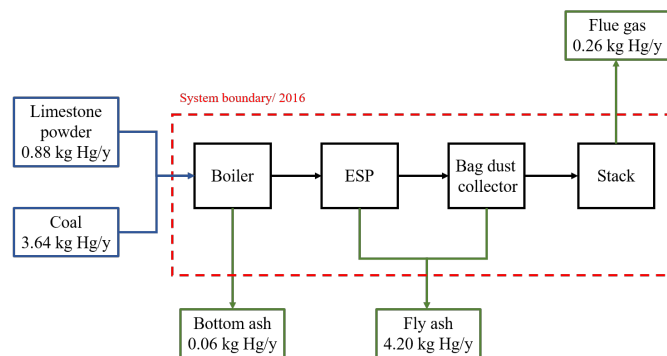
Fig. 3: SFA of mercury in the coal-fired industrial boiler in 2016

Acknowledgements

References

1. Hui ML, Zhang L, et al. (2015); China Environ Sci: 35:2241-50.
2. Wang YM, Liu GI, et al. (2016); J Environ Sci: 43: 1-3.
3. Zhu ZW, Zhuo YQ, et al. (2016); J Environ Sci: 43: 169-76.
4. Wang FY, Wang SX, et al. (2016); J Environ Sci: 43: 293-301.
5. Galbreath, K.C., Zygarlicke, C.J. (2000); Fuel Process. Technol: 65–66, 289–310.
6. Duan Y, Zhao C, et al. (2010); Energy Fuels: 24:220-4.
7. P. H. Brunner and H. Rechberger. (2002); Environ. Sci. Technol: 36, 809-816.
8. GB 15618-1995 (National Standard of P.R. China). Environmental quality standard for soils, 1995.
9. Liu X, Wang S, et al. (2013); Fuel: 111:621-7
10. Rallo M, Lopez-Anton MA, et al. (2010); Fuel: 89:2157-9.