Fuel 180 (2016) 194–204

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Actual mercury speciation and mercury discharges from coal-fired power plants in Inner Mongolia, Northern China

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HIGHLIGHTS

- Mercury emissions from coal combustion are in the dominant form of Hg^0 (>85%).
- Direct and indirect speciated Hg discharge factors of coal-fired power plants were obtained.
- The maximum of Hg emissions from coal-fired power plants of Inner Mongolia is 24 Mg/y.
- The largest direct Hg emissions may be the utilization industry of fly ash and gypsum.

ARTICLE INFO

Article history:
Received 22 November 2015
Received in revised form 7 April 2016
Accepted 8 April 2016

Keywords:
Mercury species
Direct Hg emission factors
Hg discharge factors
Coal-fired power plants
Inner Mongolia

ABSTRACT

Mercury species distribution in the flue gas from three typical coal-fired power plants have been investigated using a C-5000 sampler for Ontario Hydro Method (OHM) to evaluate specified Hg emission inventories in the largest coal-production province of China, Inner Mongolia. The feed coal, bottom ash, fly ash and gypsum have also been sampled in the field. Mercury emissions are in the dominant form of Hg^0 (>85%). The Hg^{2+} (<15%) and the Hg^p (<2%) emissions are minor fractions of Hg^t in the flue gas. The wet limestone flue gas desulfurization device (WLFGD) showed much higher removal efficiency for Hg^{2+} (87–95%) than for the other two Hg species. The input Hg/output Hg varied from 92% to 115% for the tested 3 coal-fried utility boilers during sampling time. We obtained comprehensive Hg discharge factors, including direct atmospheric Hg emission factors and indirect discharge factors of Hg associated with bottom ash, fly ash and gypsum, to the environment, designating the fates of Hg in coal. We estimated the Hg discharges from coal-fired power plants in Inner Mongolia in 2008–2014 and expanded to China. The maximum of direct Hg emissions from coal-fired power plants of Inner Mongolia was 24 Mg in 2012. The Chinese coal-fired power plants emitted directly 100 Mg Hg to the atmosphere every year at the nationwide air control actions. The maximum discharges of Hg associated with fly ash and gypsum of Inner Mongolia was 81 Mg and 11 Mg in 2012, respectively. The national Hg discharges with fly ash and gypsum every year are respectively about 300 Mg and 82 Mg, majority of which may be reemitted to the atmosphere by the utilization industry. Therefore, mercury emissions from coal-fired power plants are not the largest single source of Hg to the atmosphere in normal operation of the current efficient denitrification, particles control and desulfurization devices. Instead, the utilization industry of fly ash and gypsum would be emitting more Hg to the atmosphere.

1. Introduction

Mercury (Hg) is a global pollutant because of the long-range Hg transportation and deposition from anthropogenic sources, its bioaccumulation as methylmercury in the environment and its neurological health impacts [1]. It has been found in Arctic biota [2] at concentrations so high that it presents a threat to human health. The modern model of atmosphere Hg circulation has revealed that Hg budget had tripled since pre-industrial times. The increased part of Hg is mainly from anthropogenic sources, especially from coal-fired power plants. It has been demonstrated that burning of fossil fuels (primarily coal) is the largest Hg source of emissions accounting for about half of anthropogenic Hg emissions of the total anthropogenic emissions with an annual emission

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http://dx.doi.org/10.1016/j.fuel.2016.04.037
0016-2361/

Hg flux of 700–900 Mg to the atmosphere [3–6], and power plants are considered to be the largest single source in most countries [3].

Mercury emitted from natural sources is believed to be mainly the elemental mercury (Hg^0), whereas anthropogenically emitted Hg has a significant portion in gaseous oxidized mercury (Hg^{2+}) and particulate-phase mercury (Hg^p) depending on the specifically conditions [7]. It has been well-documented that different physicochemical forms of Hg influence its environmental transport, fates, and in some cases, impacts. Elemental mercury is volatile, relatively inert, and virtually insoluble. As a result, it can pass through conventional scrubbers and particulate control devices, such as electrostatic precipitators (ESP) and fabric dust collector (FDC) and flue gas desulfurization (FGD) and contributes to the global mercury emissions inventory [8–10]. In contrast, oxidized mercury is water-soluble, thus more apt to react with particles to form Hg^p, and part of Hg^{2+} can be captured in scrubbers while the Hg^0 may be transported a few hundred kilometers away from the source. Particulate-phase mercury is likely to deposit at an intermediate distance depending on the aerosol diameter. In the atmosphere, elemental mercury has a residence time of approximately 1 year [11,12], which is significantly longer when compared to the other forms which have atmospheric residence time of a few hours to several months. Consequently, Hg^p and Hg^{2+} are expected to be deposited closer to emission sources by dry or wet deposition. Therefore, some developed countries have been studying and using dedicated mercury-control technologies to regulate mercury emissions from coal-fired power plants [13–16].

Increasing attention has been paid on the Hg pollution problem in China because of the considerable amount of Hg emissions [3,17,18]. Streets et al. [17] estimated Hg emissions from China in 1999 where 537 (±236) Mg, and 38% of the Hg comes from coal combustion. Wu et al. [18] estimated Hg emissions from coal combustion increased from 202 Mg in 1995 to 257 Mg in 2003 at an average annual rate of 3.0%. Pacyna et al. [3] concluded that about two-thirds of global anthropogenic releases of Hg to the atmosphere appear to come from Asian sources, with China as the largest contributor worldwide for 2005. Obviously, coal-fired power plants are one of the largest anthropogenic sources of Hg emissions in China, but the above results have large uncertainties because comprehensive field investigations to characterize Hg emissions from coal-fired power plants are only available for developed countries [19–21], and have been gone for at least ten years.

However, with the increasing trend of air pollution in recent years, especially characterized by regional photochemical smog and haze in rapidly developing regions such as the North China Plain, the Yangtze River Delta and the Pearl River Delta, the Chinese government has implemented a series of national control policies to reduce the emissions of air pollutants since 2005 [22,23]. The 11th Five-Year Plan (2006–2010) for national environmental protection required the reduction of annual emissions of sulfur dioxide (SO_2) in 2010 by 10% from its 2005 level. Besides stressing on primary particles control (PPC), FGD and nitrogen oxide control ($NOxC$), the Hg control had been listed in "Emission standard of air pollutants for thermal power plants" as Chinese national standard (GB 13223-2011) and implemented in 2015. In 2013, the Chinese government has promulgated and implemented the feasible "Air Pollution Prevention and Control Actions". Therefore, with the above nationwide control actions, a large reduction in ambient SO_2 level, NOx emission and primary particles has been observed by ground monitors and satellite monitoring instruments [22,24]. In view of the above nationwide air control actions, mercury emissions from coal-fired plants in China would or will be reduced largely but lack of the actual measurements of mercury species, concentration in flue gases and capture of Hg through emission control devices in China.

The coal production of Inner Mongolia has surpassed Shanxi province and becomes the largest coal production province in China since 2007, and the coal consumption has also increased greatly. Coal resources in Inner Mongolia are divided into two areas: the first is the low metamorphic bituminous coal region in Erdos City possessing 4.3×10^{11} Mg of proven reserves, accounting for 53% of the provincial coal reserves and 13% of the national coal reserves. The second is the lignite area in eastern Inner Mongolia owning 3.7×10^{11} Mg of proven reserves, accounting for 45% of the provincial coal reserves and more than 3/4 of the national lignite reserves. Therefore, many advanced thermal power plants and coal chemical enterprises were constructed around the Ordos and the eastern Inner Mongolia.

In view of the coal distribution characteristics of Inner Mongolia, we chose three typical power plants, two of which have similar generating process, flue gas treatment process except for PPC and same generating capacity but burn bituminous coal and lignitous coal, respectively, and conducted three direct sampling and Hg analyzing campaigns. The subjects of this work aim to (1) directly determine the Hg species in flue gases from the 3 typical coal-fired plants in Inner Mongolia, Northern China, (2) study the fates of Hg in coal, the Hg discharge factors and the resulting discharges through the Hg mass balance verification, (3) estimate the Hg discharges from coal-fired power plants in Inner Mongolia in 2008–2014 and expand to China.

2. Method

2.1. Site description

Inner Mongolia has an abundance of resources especially coal, natural gas and rare earth elements, etc. It is an important coal production base, with more than a quarter of the world's coal reserves located in the province. The coal reserve has exceeded more than 8×10^{11} Mg in 2014 in Inner Mongolia.

Inner Mongolia is not only rich in bituminous coal also accounts for about 3/4 of lignite resources of China. However in the past, the exploitation and utilization of resources were rather inefficient, which resulted in poor returns from rich resources. With the implementation of series of national strategy, such as "The development campaign of the western regions", "West-East electricity transmission project" and "West-East natural gas transmission project", it has grown up mainly around coal, power generation. In 2007, the coal production in Inner Mongolia has surpassed Shanxi Province as the largest coal production province in China. The provincial coal production has increased to 1080 million Mg in 2012 from 457 million Mg in 2002 and slightly decreased from 908 million Mg in 2014 in Inner Mongolia. Consequently, many large-scale coal-fired power plants (≧300 WM) have been built with high efficiency of $NOxC$, PPC and FGD device. Therefore, the Hg investigation of the coal-fired plants in Inner Mongolia is particularly urgent for China with the nationwide air control and reduction actions situation.

In this study, three typical of coal-fired power plants are chosen to conduct three directly measuring campaigns of Hg species in flue gas and Hg in the by-products of the flue gas treatment in Inner Mongolia. The first one is Jinshan bitumite-fired power plant (JSBFPP, 2 × 300 MW) of Hohehot Municipality, the provincial capital of Inner Mongolia, which represents to the type of bituminous coal-fired power plants. The second is Xilinhot lignite-fired power plant (XLLFPP, 2 × 300 MW), the prefectural capital of Xilinguole located in the eastern Inner Mongolia, which represents to the type of lignite-fired power plants. The third is named as a gangue-fired power plant, in fact, which is a lignite-fired power plant during our