



Mercury isotope signatures of seawater discharged from a coal-fired power plant equipped with a seawater flue gas desulfurization system[☆]



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ABSTRACT

Seawater flue gas desulfurization (SFGD) systems are commonly used to remove acidic SO₂ from the flue gas with alkaline seawater in many coastal coal-fired power plants in China. However, large amount of mercury (Hg) originated from coal is also transferred into seawater during the desulfurization (De-SO₂) process. This research investigated Hg isotopes in seawater discharged from a coastal plant equipped with a SFGD system for the first time. Suspended particles of inorganic minerals, carbon residuals and sulfides are enriched in heavy Hg isotopes during the De-SO₂ process. $\delta^{202}\text{Hg}$ of particulate mercury (PHg) gradually decreased from -0.30‰ to -1.53‰ in study sea area as the distance from the point of discharge increased. The results revealed that physical mixing of contaminated De-SO₂ seawater and uncontaminated fresh seawater caused a change in isotopic composition of PHg isotopes in the discharging area; and suggested that both De-SO₂ seawater and local background contributed to PHg. The impacted sea area predicted with isotopic tracing technique was much larger than that resulted from a simple comparison of pollutant concentration. It was the first attempt to apply mercury isotopic composition signatures with two-component mixing model to trace the mercury pollution and its influence in seawater. The results could be beneficial to the coal-fired plants with SFGD systems to assess and control Hg pollution in sea area.

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1. Introduction

Human activities, such as coal combustion and other industrial emission sources (Foucher and Hintelmann, 2006, 2009; Sherman et al., 2012; Sun et al., 2013a, 2013b, 2013c), have accelerated the emission and geochemical cycling of mercury (Hg) into the environment (Choe and Gill, 2003; Morel et al., 1998). For instance, coal combustion as one of the largest emitters of Hg in the world, has contributed more than 35,000 tons into the atmosphere (Sun et al., 2016). Coal usually contains high levels of sulfur and trace amount of Hg. In China and other countries in the world, many coal-fired power plants along/near the coast are equipped with the seawater flue gas desulfurization (SFGD) system, which use the

alkaline seawater to neutralize acidic sulfur dioxide (SO₂) in the flue gas (Feng et al., 2014; Yu et al., 2011). During desulfurization (De-SO₂), the flush of sea water also washes Hg off the flue gas, resulting in high Hg content in the seawater (Liang et al., 2010; Liu et al., 2011). A schematic diagram showing the process and Hg transfer from the coal combustion and the SFGD system into the estuary is provided in Fig. S1 of Supplementary. The De-SO₂ seawater is mixed with fresh seawater and treated with simple aeration to further neutralize and oxidize SO₃²⁻, increase pH and dissolved oxygen (DO) (Liu et al., 2011), and then discharged into the adjacent sea. The technique utilizes local seawater resource and avoids producing huge amount of solid waste that is usually generated with common dry desulfurization techniques. The De-SO₂ seawater mainly contains relatively high levels of suspended particles and SO₄²⁻, which are considered not to be the pollutants for coastal environment. However, heavy metals, especially Hg, at high concentration in the De-SO₂ seawater could become a potential threat to the ecosystem.

A typical air purification system of coal-fired flue gas is

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composed of a selective catalytic reduction unit, a particulate control unit and a flue gas desulfurization system in series (Hower et al., 2010), installing after the boiler (Fig. S1 in the Supplementary). During the coal combustion ($>1200\text{ }^{\circ}\text{C}$) in the boiler, Hg in coal is released as Hg^0 into the flue gas (Galbreath and Zygarlicke, 2000b). The selective catalytic reduction unit is used for removal of nitrogen oxides, and at the same time it transfers 30–98% Hg^0 into Hg^{2+} , which is easily to adsorb onto particles and form PHg in the flue gas (Hower et al., 2010). Part of particles and PHg are removed in the particulate control unit. When the flue gas flows through the flue gas desulfurization system, 30–50% PHg, most of Hg^{2+} and part of Hg^0 are transferred into by-products such as gypsum or seawater (Hower et al., 2010).

The SY power plant, located in the Xiamen city (southeast coast of China, Fig. 1), is one typically example of coal-fired power plant equipped with SFDG that becomes a Hg emission source (Liang et al., 2010). For instance, the De- SO_2 seawater at the outlet of the De- SO_2 tower contains total Hg (THg) up to 385.0 ng/L, much higher than China's Class II seawater quality standard (200.0 ng/L). According to our previous studies, the De- SO_2 seawater can bring 1.03 kg/day of Hg into the adjacent sea (Liu et al., 2011). The result of regular monitoring indicated seawater within approximately 300 m of the SY power plant has shown slightly higher Hg content than that of that in other coastal areas in Xiamen. However, the environmental impact of Hg from the De- SO_2 seawater is still a question because of the mismatch of the Hg in the De- SO_2 seawater and the discharging sea area; based on the simply Hg mass balance calculations. It is thought that the mixing with local Hg background can result in decreased THg in seawaters of the discharging area, whereas there is a lack of effective tracer to quantify the contribution of Hg from De- SO_2 seawater and background. It is not yet clear how large a sea area is influenced by the Hg-contaminated De- SO_2 seawater.

During the past 15 years, mercury isotope geochemistry has been proved to be a powerful tool to trace sources and geochemical processes of Hg in the environment (Bergquist and Blum, 2009;

Blum et al., 2014; Wiederhold et al., 2015). Previous studies have investigated Hg isotopic compositions of coal from different regions of the world (Biswas et al., 2008; Lefticariu et al., 2011; Sun et al., 2014, 2016; Yin et al., 2014), Hg isotope fractionation during coal combustion process (Sun et al., 2013c), and isotopic signature of Hg in atmospheric samples near a coal power plant (Sherman et al., 2012). These studies have facilitated our understanding on Hg transformation, transportation, and environmental impacts during Hg emission by coal combustion. However, Hg isotope fractionation during Hg emission by SFDG is still not well understood. A main technological gap for this is that Hg in seawater is generally too low for Hg isotope analysis. Analytical methods for water Hg isotope determination have been developed recently (Lin et al., 2015; Štrok et al., 2014, 2015). To date, only a few studies have reported Hg isotopic compositions in seawaters (Štrok et al., 2015); whereas isotopic compositions in the De- SO_2 seawater have not been studied.

Particulate Hg (PHg) is the main species in water systems and its concentration in natural water varies in different regions around the world. Concentrations of PHg in fresh water of the Isonzo River mouth range from 9.0 to 25.0 ng/L, contributing 51–84% to THg (Stefano et al., 2006). In the seawater of discharging sea area near the SY power plant, 50–97% of THg is in PHg form (Liang et al., 2010; Lin et al., 2015) due to the fact that De- SO_2 seawater contains large amount suspended particles, mainly inorganic minerals, carbon residuals and sulfides, generated during the coal combustion process and not removed by the particulate control unit. The particles containing carbon, acid and sulfides have a high affinity to bond Hg (Galbreath and Zygarlicke, 2000a). Therefore, high concentration of PHg was observed in the De- SO_2 seawater.

This study investigated the variations of Hg isotope compositions in feed coal used in the plant, PHg and DHg in De- SO_2 seawater discharged; discussed the transfer and transformation of PHg and DHg in the seawater, and evaluated the source impact of Hg by SFDG. The results of this study could provide information for better understanding environmental behavior of Hg discharged

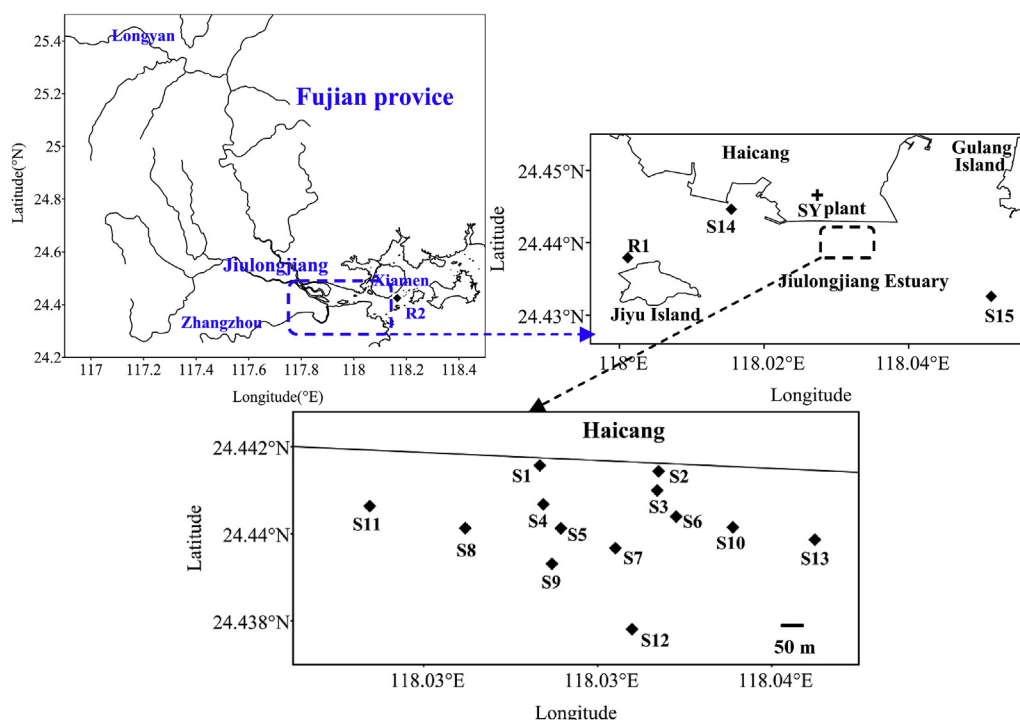


Fig. 1. Location of the sampling sites.

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