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Occurrence of uranium in Chinese coals and its emissions from coal-fired power plants

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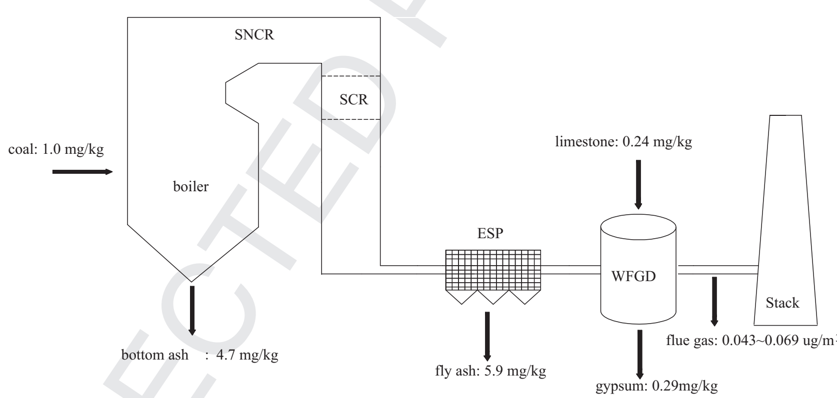
HIGHLIGHTS

- Uranium content for most of Chinese coals is in the range from 1 to 3 mg/kg.
- Uranium release from coal increases directly with combustion temperature.
- Distribution of uranium in coal-fired power plant boiler effluents is majority in the ash.
- For coal-fired power plants, less than 1/1000 of input uranium in coal results in flue gas.

GRAPHICAL ABSTRACT

Uranium concentrations in samples of a power plant.



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ABSTRACT

Uranium is a trace elemental in coal. The uranium content from coal mines in China and its distribution in coal-fired power plant effluents was studied and reported in this paper. Chinese coal samples from eighteen coal mines were collected and analyzed for uranium. It was found that the uranium content for most of the coals was in the range from 1 to 3 mg/kg. Laboratory experiments with a tubular furnace indicate that the amount of uranium released from heated coal increases with temperature and with the retention time, when it combust. Most of the uranium (about 90%) in the coal can be transferred to the ash. Full-scale field tests were carried out at a coal-fired power plant to investigate the uranium distribution (and mass balance) in the coal, limestone, fly ash, bottom ash, gypsum and the flue gas. Almost all the uranium in the coal concentrated in the fly ash (about 80%) and the bottom ash (about 10%). The uranium content in the flue gas was less than 1/1000 of the total elemental input, between 0.043 and 0.069 μg/m^3, which is much lower than the typical concentration of mercury or arsenic in flue gas of coal-fired power plant. The uranium content in the desulfurization gypsum product was found to be slightly larger than the uranium content of the limestone.

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1. Introduction

Coal is China's major energy source. Coal is used as the most important form of electricity generation in China. According to China statistics, about 3.526 billion tons of coal were consumed in China in 2012, from which, 1.785 billion tons were used for power generation [1]. However, as the concern for environmental protection increases in China, the occurrence, distribution and impact of trace elements in coal needs to be studied [2]. At the beginning of 2014, there was an unconfirmed rumor in China indicating that the haze in different parts of China was caused by uranium, which was released from coal combustion from coal-fired power plants, a so-called "nuclear haze" that sparked widespread concern in the society [3]. Uranium is one of the natural radioactive elements and its concentration in coal has an important impact on coal mining and utilization [4]. Yang surveyed more than 1500 types of Chinese coal samples, concluded that the mean value of uranium in Chinese coals is 2.31 mg/kg [5]. Huang et al. [6] reported the mean value of uranium in Chinese coals at 3 mg/kg, which is near the global average. Research results from the U.S. Geological Survey indicate that the uranium content in most of American coals is less than 4 mg/kg and, furthermore, that uranium concentrates in solid combustion wastes after coal combustion in power plants [7].

Studies have been reported in the literature on analysis of material samples from coal-fired power plants, in terms of uranium content in coal, fly ash, and bottom ash [7–13]. For example, in Parami' study [9], the range of uranium concentrations in fly ash ranges from 1.1 to 21.7 mg/kg for Philippine's coal-fired thermal power plants. In another study by Flues [10], in Brazil, the range of uranium concentrations in the fly ash ranges from 117 to 1190 mg/kg.

However, those analyses do not account for direct measurements for uranium content in the flue gas. There is a need to document uranium flue gas emission from coal-fired units, since that compared with studies of other trace elements in coal, only very limited data are available. This makes very difficult to confirm how much uranium is released to air from coal-fired power plants in China. Moreover, reports on the release characteristics of uranium from coal combustion and the occurrence and distribution of uranium from the different effluents in coal-fired power plants is also limited.

To fill this void, this paper reports measurements performed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) on uranium concentration in a variety of Chinese coal samples from eighteen coal mines in the Inner Mongolia, Xinjiang, Ningxia and Yunnan Provinces. The paper also reports the release characteristics of uranium from coal combustion in a tube furnace and uranium mass balance characteristics of coal combustion in a drop tube furnace. A round of field measurements was performed of coal, fly ash, bottom ash, limestone, gypsum and flue gas samples of a 200 MW coal-fired power plant. Material samples were collected, measured and analyzed, and the distribution and mass balance characteristics of uranium is discussed.

2. Analytical methods – sample collection and preparation

The coal samples analyzed in this study were collected from different coal mines. These samples were prepared by grinding and screening to an 80-mesh screen size.

The sampling coal-fired power plant is equipped with low NO_x burner, a hybrid selective catalytic reduction (SCR) + selective non-catalytic reduction (SNCR) denitrification system, an electrostatic precipitator (ESP) for particulate matter (PM) control and wet flue gas desulfurization (WFGD) equipment. Samples were collected from the flue gas at the stack. Solid samples include samples collected from the feed coal, bottom ash, fly ash, limestone and desulfurization gypsum. The sampling locations were located at the coal conveyor belt, the slag scraper, bottom hoppers of the ESP, the entrance to the desulphurization tower and the gypsum discharge belt. Flue gas samples were collected from the flue after the desulphurization unit using the U.S. Environmental Protection Agency (EPA)-Method 29, with an Apex analyzer (Apex, USA), which is applicable to the determination of metal emissions from stationary sources, and it was also used for gas sampling of the drop tube furnace in the laboratory.

Measurement methods of uranium includes spectrophotometric method, neutron activation analysis, atomic absorption spectrometry, X-ray fluorescence analysis, ICP-AES (Inductively Coupled Plasma Atomic Emission Spectrometry), ICP-MS, among other [14,15]. Inductively coupled plasma mass spectrometry (ICP-MS) is a means for simultaneous multi-element analysis with characteristics of high sensitivity, low detection limit and high detection speed, It's suitable for the analysis of uranium of complex samples. The detection limit of the ICP-MS (NEXION 300D, USA) for U-238 is 0.01 ng/g. The ICP-MS was used in this study to analyze the concentration of uranium in the samples. For this technique, digestion is necessary for solid sample analysis. In order to digest the samples completely, the coal samples were ashed in the muffle furnace. Then, solid samples were processed into liquid samples after digestion for the analysis by the ICP-MS. All these procedures could contribute to loss of uranium in the samples and induced errors. Additionally, readings from the ICP-MS technique could have been affected by fixation by other elements in the sample.

After sampling, coal and other solid samples were prepared by grinding and screening to an 80-mesh screen size. Then the solid samples were digested in a microwave digestion system (Speed-Wave MWS-4, German) with a combination of high pure acids HNO_3 and HF and measured by ICP-MS. A uranium standard solution (SPEXertificate PLU2-2Y, USA) was used to derive calibration curves.

3. Uranium content in different coal mine samples

The data obtained from the uranium measurements of mined coal samples is shown in Fig. 2. Coals from eighteen mines in China were used. Those mines are located in the Inner Mongolia, Xinjiang, Ningxia and Yunnan Provinces. It can be determined from Fig. 1 the range of uranium content from 1 to 3 mg/kg for the majority of the coals. The uranium content of coal P from Inner Mongolia is 5.8 mg/kg. The highest uranium content of

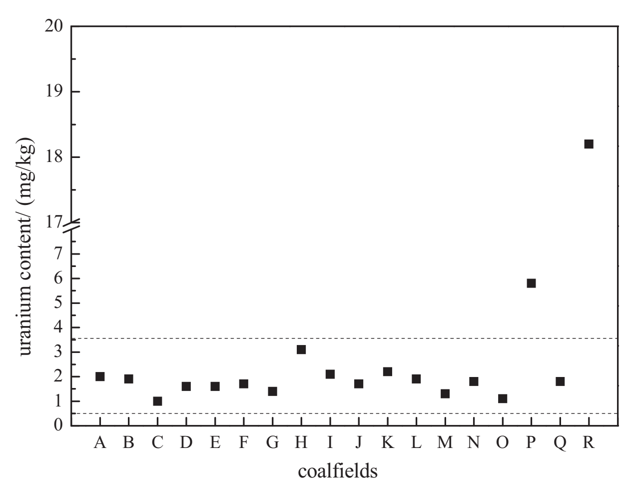

Fig. 1. Uranium content of different Chinese coalfields.

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