Measurement of Radon Concentration and Estimation of Its Emission of Coal-Fired Power Plants

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Abstract. In the experiment, RAD7 multi-functional electronic radon meter was used to measure the radon (²²²Rn) concentration at key points in the production process of three typical coal-fired power plants in Hunan, Hubei and Jiangxi provinces along the Yangtze river in China. The results show that after the combustion of raw coal, the radon was released, which caused radon concentration after the boiler's gas system to be significantly higher than the air intake. Combined with the measurement and survey data, two different estimation methods of radon emission were established. First, based on radon concentration measured on site and the air volume at each inlet and outlet, the normalized radon emission of three coal fired power plants are calculated to be 10² GBq/GWa order of magnitude. The second one is using activity concentration of ²²⁶Ra in combination with actual coal use to calculate the emission, in this way, that typical value of normalize radon emission 151.3 GBq/GWa is obtained. This study provides data support for the construction of nuclear power plants in inland China.

1. Introduction
China's nuclear power has entered a rapid development period, the site selection of nuclear power rapidly increased, from the coast gradually extended to the inland. Studies have shown that under the safe operation, the increase of environmental radiation impact from inland nuclear power plants is much lower than the background radiation level, and it will not affect the environment and public health. However, the current data on the environmental impact of radiation caused by non-nuclear enterprises are very imperfect, which requires further investigation and monitoring.

Coal has long been used as raw material, fuel and domestic energy in China's power, metallurgy, chemical and other industries. From 1990 to 2016, in China's coal consumption constitution, the coal consumption for coal-fired power generation increased year by year, reaching 182.66 million tons in 2016 [1]. Data from China energy development report show that by 2020, 1.96~2.587 billion tons of raw coal will be needed for power generation, accounting for about 80% of China's total coal production [2].

While coal brings great contribution to social economy, it also brings environmental impact that can't be ignored. Coal contains trace natural radionuclides such as 238U, 232Th, 226Ra and their progeny, etc. With the combustion of coal, the emission of radionuclides and the gaseous progeny ²²²Rn of ²²⁶Ra to the environment will increase. The radioactive material will be released into the environment in gaseous or solid form, and spread over a large area around coal-fired power plants, producing a certain dose to the public. The project has measured ²²²Rn concentration of coal-fired power plants in three provinces along the Yangtze river, estimated ²²²Rn emission of the coal-fired power plant, providing data support for the construction of inland nuclear power plants in China. It is should be noted that radon, as mentioned below, specifically refers to ²²²Rn.
2. Methodology

2.1. Introductions of three coal-fired power plants

There are many coal-fired power plants along the Yangtze river. According to the data of the first national survey of pollution sources organized by the ministry of ecology and environment of China, there are 37 coal-fired power plants in Hunan, Hubei and Jiangxi. A typical coal-fired power plant was selected as the measurement object from each of the three provinces. The operating unit parameters of each coal-fired power plant during the measurement period are shown in table 1:

| Coal-fired power Plants | Unit | Power (MW) | Annual load efficiency | Annual operating time (h) | Power generation (KWh) | Annual coal consumption (t) | Coal consumption per unit time (t/h) |
|------------------------|------|------------|------------------------|--------------------------|------------------------|-----------------------------|-----------------------------------|
| Hunan                  | 1#   | 300        | 67%                    | 3500                     | 6.5×10⁸                | 45.52×10⁴                   | 129.77                            |
|                        | 4#   | 630        | 67%                    | 3500                     | 26.5×10⁴               | 95.38×10⁴                   | 272.52                            |
| Hubei                  | 1#   | 330        | 77.4%                  | 7861                     | 20.05×10⁸              | 150×10⁴                     | 45.38×10⁴                         |
|                        | 3#   | 680        | 78.7%                  | 7314                     | 39.19×10⁸              | 160×10⁴                     | 272.52                            |
| Jiangxi                | 5#   | 350        | 81.5%                  | 4591                     | 13.10×10⁸              | 64.27×10⁴                   | 140                               |
|                        | 7#   | 660        | 84.92%                 | 6883                     | 38.58×10⁸              | 172.07×10⁴                  | 250                               |

Figure 1. shows the common smoke and dust treatment process in China's coal-fired power plants. Raw coal is burned in a boiler to generate heat for power generation. The flue gas produced by combustion in boilers including various gases and particulates, enters into the flue gas treatment system, passes through the economizer first, then goes into the denitrification system, dust removal device (electrostatic precipitation technology, bag dust removal technology, etc.) and the desulfurization tower, finally is discharged into the atmosphere through the chimney.

Pulverized coal combustion requires a great deal of air, which is supplied by an induced draft fan (also called a primary fan) and a supply fan (also called a secondary fan). A boiler is usually equipped with one or two secondary fans each. The primary air is to send coal powder into the burner combustion. Secondary air plays a combustion supporting role, which can supplement the air needed for pulverized
coal combustion, ensure the full combustion of pulverized coal, and promote the full mixing of pulverized coal and air.

2.2. Measurement methods and experimental equipment

The RAD7 multi-function electronic radon measuring instrument produced by American company DURRIDG was used in this experiment. Four instruments used in the experiment were calibrated in the Standard Radon Laboratory of Nanhua University (IAEA Regional coordinating laboratory for radon metrology, Asia). The calibration coefficients of four RAD7 instruments are shown in Table 2.

| Serial number | RAD7-1410 | RAD7-3399 | RAD7-3583 | RAD7-3584 |
|---------------|-----------|-----------|-----------|-----------|
| Calibration coefficients | 1         | 0.97      | 1.04      | 0.98      |

Based on preliminary field research, the experimental measurement scheme is designed according to the requirements of the instrument. The specific method is shown in Figure 2 (taking the measurement of radon concentration in chimneys as an example). The measurement method of radon concentration in air of coal-fired power plant is based on the requirements of GB/T 14582-1993 (Standard Measurement Method for Radon in Ambient Air) and GBZT 182-2006 (Standard Code for Measuring Radon and Its Decay Products in Indoor).

![Figure 2. Method of radon concentration measurement in chimneys](image)

3. Results and discussion

3.1. Radon concentration

As can be seen from Figure 1, in the production process of coal-fired power plants, there are four air inlets: the primary and secondary air inlets, oxidized air inlet under desulfurization tower and positive pressure air inlet at dust removal place. The air outlets are chimneys, dust removal bags on the top of ash depots and slag discharge outlets. According to the distribution of air inlets and outlets, radon concentration was measured at the key points of the three selected coal-fired power plants. Among them, although the denitrification device is set up in Hunan coal-fired power plant, it is not used all year round, so the denitrification inlet is not considered in the measurement. The radon concentration of plant
environment, pulverized coal separator and other devices do not contribute to the calculation of emission. Therefore, specific measurements are only made at these points in Hunan coal-fired power plant for comparison. Compared with Hunan coal-fired power plant, Hubei coal-fired power plant has added denitrification equipment in the flue gas treatment process, therefore, the radon concentration at the outlet was measured. The measurement results are shown in Table 3, 4.

**Table 3. The radon concentration of Hunan coal-fired power plant, Bq/m³**

| Area                        | location                                           | Value |
|-----------------------------|----------------------------------------------------|-------|
| External environment comparison point | The upwind of the power plant chimney, 5km away and 1.2m high | 19.8  |
|                             | Pulverized coal separator-out of operation         | 10.5  |
| 1#300MW                     | Pulverized coal separator-in operation              | 34.0  |
|                             | Chimney                                             | 17.8  |
|                             | The primary and secondary air inlets, 3m high       | 15.4  |
|                             | The south side of boiler, 13m high                  | 11.8  |
|                             | Coal mill                                           | 14.5  |
|                             | Flue gas system                                     | 24.2  |
|                             | Pulverized coal separator                            |       |
|                             | Flue after boiler                                   | 20.7  |
| 4#630MW                     | Desulfurization oxidized air inlet                  | 18.8  |
|                             | Chimney                                             | 17.3  |
| Ash blowing system          | Fine dust removal bags on the top of ash depot      | 35.2  |
|                             | Raw dust removal bags on the top of ash depot       | 18.2  |
| Slag discharge system       | Boiler slag discharge port                          | 8.6   |
|                             | Near the coal pile                                  | 15.3  |

**Table 4. The radon concentration of Hubei and Jiangxi coal-fired power plant, Bq/m³**

| Plants       | Area          | Value                          |
|--------------|---------------|-------------------------------|
|              | The primary and secondary air inlets | Denitrification inlet | Desulfurization oxidized air inlet | Chimney | Ash removal air compressor | Top of ash depots |
| Hubei        | 1#330MW       | 12.6                          | 19.9                          | 12.3    | 17.6                         | 7.8               | 11.4              |
|              | 3#680MW       | 8.00                          | 28.7                          | 12.3    | 22.1                         |                   |                   |
| Jiangxi      | 5#350MW       | 6.6                           | 9.5                           | -       | 13.6                         | 7.6               | 7.0               |
|              | 7#660MW       | 8.7                           | 13.5                          | -       | 11.0                         |                   |                   |

According to the measurement results, in the flue gas system, the gas after the boiler has a large amount of radon which is released by coal burning, therefore, the radon concentration at the denitrification inlet is relatively higher. While the flue gas at the chimney is diluted by the air at desulfurization and oxidation inlet, the radon concentration decreases to a certain extent, so it is lower than the denitrination inlet. The radon concentration in intakes and outlets of Jiangxi coal-fired power plant is relatively lower than that in Hunan and Hubei, it is mainly affected by the regional background radon concentration and the difference of activity concentration of $^{226}$Ra in different types of coals.

**3.2. Estimation based on actual measurement results**

In this study, two methods are used to estimate radon emission of coal-fired power plants: The first method is based on radon concentration measured on site and the air volume at each inlet and outlet; The second one is using activity concentration of $^{226}$Ra in combination with actual coal use to calculate the emission. In this section, the first method will be described.
Actual radon emission from coal-fired power plants: actual radon concentration in the chimney multiplied by the total air intake volume without considering the background value of the air at the inlet of the flue gas system.

\[ Q_i = C_i \times (V_i + V_2) \times t \]  
(1)

In the equation, \( Q_i \) is actual radon emission from chimney, Bq; \( C_i \) is radon concentration of chimney, Bq/m\(^3\); \( V_1 \) is total boiler air intake volume, m\(^3\)/h; \( V_2 \) is desulfurization oxidized air intake volume, m\(^3\)/h; \( t \) is annual operating time, h. Therefore, the actual normalized radon emission from the chimney is the ratio of the actual radon emission from chimney to power generation, that is

\[ Q_0 = \frac{Q_i}{W} \]  
(2)

In the equation, \( Q_0 \) is actual normalized radon emission from chimney, GBq/GWa; \( W \) is radon concentration of chimney, GW.

Radon emission from the flue gas system: the difference between the measured radon concentration in the chimney and the representative value of the total intake radon concentration, multiplied by the total annual air intake volume of the flue gas system, that is:

\[ Q_2 = (C_1 - C_2) \times (V_1 + V_2) \times t \]  
(3)

In the equation: \( Q_2 \) is radon emission from flue gas system after boiler, Bq; \( C_1 \) is radon concentration of chimney, Bq/m\(^3\); \( C_2 \) is the total radon concentration, it is composed of radon concentration at the primary and secondary fan inlets, radon concentration at the oxidative desulfurization air inlet in air volume weighted, Bq/m\(^3\); \( V_1 \) is the total air intake volume of boiler, m\(^3\)/h; \( V_2 \) is the desulfurized oxidized air intake volume, m\(^3\)/h; \( t \) is annual operating time, h.

Radon emission from positive pressure dust removal system of one unit: the difference between radon concentration at the outlet and inlet of the positive pressure dust removal system, multiply by the air volume of the positive pressure dust removal of a unit, that is:

\[ Q_3 = (C_3 - C_4) \times V_3 \times t \]  
(4)

In the equation: \( Q_3 \) is radon emission from positive pressure dust removal system, Bq; \( C_3 \) is radon concentration at the outlet, Bq/m\(^3\); \( C_4 \) is radon concentration at the inlet, Bq/m\(^3\); \( V_3 \) is air volume of the positive pressure dust removal, m\(^3\)/h; \( t \) is annual operating time, h.

Additional normalized radon emission from coal combustion: the ratio of the sum of radon emission of each system to the generation of electricity, that is:

\[ Q = \frac{Q_0 + Q_2}{W} \]  
(5)

In the equation: \( Q \) is additional normalized radon emission from coal combustion, GWa; \( Q_2 \) is radon emission from flue gas system after boiler, Bq; \( Q_3 \) is radon emission from positive pressure dust removal system, Bq; \( W \) is radon concentration of chimney, GW.

The radon emission estimation results of three coal-fired power plants in Hunan, Hubei and Jiangxi are shown in Table 5. Jiangxi coal fired plant did not carry out the monitoring and estimating of radon in dust removal system. However, the radon emission from the positive pressure dust removal system is several orders of magnitude lower than that from the chimney, so the impact on the total emissions can be ignored. According to the estimation results, it can be known that:

i ) The actual annual radon emission from the chimney is at the level of \( 10^{11} \), which is about one order of magnitude higher than that from the flue gas system due to the influence of background radon concentration. The normalized radon emission is basically on \( 10^2 \) GBq/GWa order of magnitude.

ii ) The emission of different units in coal-fired power plants are different, which are mainly affected by raw coal varieties (\(^{226}\)Ra activity concentration varies greatly among different types of coal), coal consumption and background concentration level in coal-fired power plants.

| Table 5. The radon emission estimation results based on actual measurement |
|-----------------------------|-----------------------------|-----------------------------|-----------------------------|
| Plants                  | Installed capacity | Emission | Actual radon emission | Additional radon emission from coal combustion |
|-----------------------------|-----------------------------|-----------------------------|-----------------------------|
| ...                        | ...                        | ...                        | ...                        |
| ...                        | ...                        | ...                        | ...                        |
| ...                        | ...                        | ...                        | ...                        |
| ...                        | ...                        | ...                        | ...                        |
| ...                        | ...                        | ...                        | ...                        |

5
Flue gas system Positive pressure dust removal system

|        | Annual emission | Normalized emission | Normalized emission |
|--------|-----------------|---------------------|---------------------|
| Hunan  | 2.1 × 10^{11} Bq/a | 6.7 × 10^{10} Bq/a | 3.5 × 10^{9} Bq/a |
|        | 711 GBq/GWa | 223 GBq/GWa | |
| 330MW  | 1.5 × 10^{11} Bq/a | 4.2 × 10^{10} Bq/a | 5.1 × 10^{9} Bq/a |
|        | 641 GBq/GWa | 182 GBq/GWa | |
| Hubei  | 3.4 × 10^{11} Bq/a | 2.2 × 10^{11} Bq/a | 6.2 × 10^{9} Bq/a |
| 680MW  | 761 GBq/GWa | 494 GBq/GWa | |
|        | 1.0 × 10^{11} Bq/a | 5.3 × 10^{10} Bq/a | - |
| 350MW  | 667 GBq/GWa | 355 GBq/GWa | |
| Jiangxi| 1.9 × 10^{11} Bq/a | 4.0 × 10^{10} Bq/a | - |
| 680MW  | 431 GBq/GWa | 90.9 GBq/GWa | |

3.3. Theoretical estimation based on $^{226}$Ra activity concentration

In this section, the third method will be described. Based on the equilibrium of $^{226}$Ra →$^{222}$Rn, and assuming that all $^{222}$Rn in coal is released after combustion, the maximum annual radon emission of coal-fired power plants is:

$$A_{Rn} = A_{Ra} \times M$$

In the equation: $A_{Rn}$ is annual radon emission, Bq; $A_{Ra}$ is $^{226}$Ra activity concentration, Bq/kg; $M$ is annual coal consumption of the power plant.

Research has shown that China’s coal-fired power plant has a large and wide distribution of raw coal [3]. In some reference papers, the $^{226}$Ra activity concentration of raw coal used in coal-fired power plants of different regions in China are listed, according to these data, the arithmetic mean value of $^{226}$Ra in raw coal of China's coal-fired power plants is 32.5 Bq/kg (gangue coal is not taken into account). China institute of atomic energy has established the "National radionuclides database in coal mines". The radionuclides activity concentrations in coal samples from different areas of China were collected in detail in the database. Through statistical analysis of the database, the average $^{226}$Ra activity concentration in China's coal samples was obtained, as shown in table 6.

According to the survey data in the past years, the electric energy production and coal consumption of coal fired power plants varies from time to time. Therefore, the latest available query data, 4437.1 bill. KWh electric energy production and 1826.66 mill. tons of thermal coal, is selected for estimation.

| Calculation method of $^{226}$Ra | $^{226}$Ra activity concentration (Bq/kg) | Normalized coal consumption (x10^7 kg/GWa) | Normalized radon emission (GBq/GWa) |
|---------------------------------|------------------------------------------|------------------------------------------|-----------------------------------|
| Values of this survey           | 32.5                                     | 360.6                                    | 117.2                             |
| Thermal coal                    | 33                                       | 360.6                                    | 119.0                             |
| Weighted values by number of samples | 53                                       | 360.6                                    | 190.8                             |

Table 6. The radon emission estimation results based on $^{226}$Ra activity concentration
Weighted values by production

|          |        |        |        |
|----------|--------|--------|--------|
|          | 49.4   | 360.6  | 178.1  |
| Typical  |        |        | 151.3  |

4. References

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