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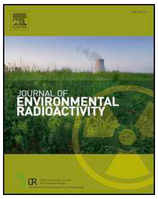

Stack releases of radionuclides from an integrated steel plant in China

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ABSTRACT

Crude steel production in China made up the majority of the global output, at 49.5% in 2014. High temperature smelting processes result in the release of natural radionuclides, including radon gas and other air pollutants into the atmosphere. This paper conducts an analysis of the raw materials, end products and flue gas sampled from an integrated steel plant from within China's Jiangxi Province, with annual production of 8.50 Mt of crude steel. Normalized stack emissions factors of radionuclides from steel production were first reported in China. The results showed that sintering was the main process that released natural radionuclides, and the main radionuclides released into the atmosphere were ^{222}Rn (86.4 GBq/Mt), ^{210}Pb (13.4 GBq/Mt), and ^{210}Po (1.71 GBq/Mt). The results provided essential basic data for radiological impact assessment of steel production, as well as that of nuclear energy chain, coal chain and other electricity sources.

1. Introduction

Industrial activities such as mining and mineral processing enhance the concentrations of naturally occurring radioactive materials (NORM). Industries contributing to the increase of natural radiation are called NORM industries. Steel production is one of the major NORM industries (Crockett et al., 2003; Leenhouts et al., 1996).

Radioactive emissions from the steel industry are the results of high temperatures used in coking (~1000 °C), sintering (~1400 °C), iron smelting (~1500 °C), and steelmaking (1500–1700 °C). This drives ^{210}Pb and ^{210}Po into gas phase, where they condense on particles (mainly $PM_{2.5}$) and release into the atmosphere along with other air pollutants (Li et al., 2015). High temperatures destroy the physical structure, so ^{222}Rn releases.

Research in the UK (Crockett et al., 2003) and the Netherlands (Leenhouts et al., 1996) showed that airborne radioactive emissions from sintering and iron smelting were the largest. Iron and steel production plants mainly emit natural radionuclides such as ^{222}Rn, ^{210}Pb and ^{210}Po. The transfer of radionuclides in particles emitted from steel production may cause the increase of the radioactivity of natural radionuclides in the atmosphere, hydrosphere, biosphere and geosphere.

Stack emissions from Redcar, Scunthorpe, and Port Talbot steel works of the UK Corus resulted in annual individual doses of 3.5, 5.8, 9.4 and 30.6 μSv/a (Crockett et al., 2003), respectively. The individual dose caused by stack emissions from the ILVA steel production plant in Italy was 27.2 μSv/a (Jia, 2013). The maximum individual dose caused by stack emissions from primary iron and steel production plant at IJmuiden in the Netherlands was 40 μSv/a (Leenhouts et al., 1996).

Some countries and organizations have already developed regulations on the NORM exposure (Li et al., 2015). The clearance levels of ^{210}Pb and ^{210}Po in fly ashes were set as 1.7×10^4 Bq/kg and 6×10^3 Bq/kg in the Basic Safety Standards of European Commission (Council Directive 96/29/EURATOM) in 1996 (European Commission, 1996). The activity concentrations of ^{210}Pb and ^{210}Po were 1.13×10^4 Bq/kg and 9.98×10^4 Bq/kg in the dust emitted from sintering of Scunthorpe steel works, so radiological controls are confined to the authorisation of atmospheric releases from sinter plant stacks (Crockett et al., 2003). Regulators in the UK set the maximum emission permit for ^{210}Pb and ^{210}Po from every integrated steel plant and that for Scunthorpe steel works were 29 GBq/a and 53 GBq/a. Airborne radioactive emissions of ^{210}Pb and ^{210}Po from this plant were 6.22 GBq/a and 39.6 GBq/a in 1990 which all met the emission requirements (Crockett et al., 2003).

Steel production and consumption has increased rapidly in China. Iron ore production reached 1500 Mt accounting for 46.6% of the global output in 2014 (Tuck, 2015). Crude steel production reached 822.8 Mt accounting for 49.5% of the global output in 2014 (Worldsteel Association, 2015). Steel production emits a large amount of waste gas, waste water and residue. In 2014, the total emissions of dust from iron and steel industry in China reached 1.015 Mt (Ministry of

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Environmental Protection of People's Republic of China, 2015). The emissions factor of dust was 0.36 kg/t at Scunthorpe steel works in the UK in 1990 (Crockett et al., 2003), while the emissions factor in China in 2014 was as high as 1.23 kg/t, more than three times of that in the UK. Therefore, steel production in China is likely to emit a large amount of natural radionuclides which may increase public exposure. There are very few reports on the emissions of natural radionuclides from steel production in China, thus the research on the emissions factors of radionuclides in steel production is of remarkable scientific significance.

This study collected samples of aerosols and measured radon concentrations from the stacks and ambient air as well as samples of raw materials and products in a major integrated steel plant in Jiangxi Province, China between 17 and 22 of July 2014. This plant consumed 21.08 Mt of iron ore and produced 8.66 Mt of pig iron and 8.50 Mt of crude steel in 2013. It ranked the 39th in the world, applied a full process flow (coking, sintering, iron smelting, steelmaking, continuous casting and steel rolling) and utilized the mainstream production processes in China (band-sintering, blast furnace and steel converter), a typical integrated steel plant, therefore an ideal model for the current situation.

Iron and steel are essential fundamental building materials, so the research on the radiological emissions of steel production will provide basic data for the comparison of the radiological impact of different energy chains (i.e., coal power, hydro power, nuclear power, wind power and solar power). This study provided essential fundamental data for the radiological impact evaluation of emissions from nuclear energy and coal chains supported by the Consulting Project of Chinese Academy of Engineering (2015-XY-15).

2. Material and methods

2.1. Basic parameters

Steel production processes mainly include coking, sintering (pelletizing), iron smelting, steelmaking and rolling. Coke, the product of coking, provides carbon in blast furnaces for iron ore to be reduced into iron. For ideal blast furnace conditions, the raw materials fed into the blast furnace must confirm to a strict size, sintering therefore refines those raw materials to form sinter. Devices for steelmaking include converters and electric furnaces, which utilize molten iron and electric energy as heat sources to oxidize impurities in the furnace.

This integrated steel plant utilizes six 192-hole coke ovens, one 115 m^2 sintering machine, one 180 m^2 sintering machine, two 360 m^2 sintering machines, one∮5 × L3 meter rotary kiln for pelletizing, three 1050 m^3 blast furnaces, two 2500 m^3 blast furnaces, three 100 t converters, two 210 t converters and two 40 t electric arc furnaces in this integrated steel plant. This study took samples from coke oven number four, sintering machine (115 m^2 with desulphurization facilities under construction) number four, blast furnace (2500 m^3) number four, converters (210 t) number one and two. Their annual productions and stack gas flows are shown in Table 1. Converters number one and two shared a common set of flue gas treatment facility, so the parameters in Table 1 combined the total amount of these two converters.

2.2. Sample collection

Sampling and measurement sites are shown in Fig. 1. Radon measurement sites included the coke oven stack, sintering machine head and end stacks, the blast furnace stack and the converter stack, as well as the ambient air in the coke plant, on the platform of sintering machine and in the steelmaking plant. Sampling sites of aerosols included the coke oven stack, sintering machine head stack, the blast furnace stack, as well as the ambient air around the platform of sintering machine and in the countryside upwind of this integrated steel plant.

The sampling system of airborne radioactive materials in the stack was designed by Wang et al. (2017). The sampling sites in stacks were set at the places with well mixed exhaust gas. Exhaust gas in stacks were sampled with a uniform flow stack sampling device and a three-stage absorption apparatus (Wang et al., 2017). Glass fiber filters were changed every two to three hours. Absorbent in the three-stage absorption apparatus was 2 mol/L HCl. The volumes of aerosols collected in the sintering machine head stack, coke oven stack and blast furnace stack were 92.5 m^3, 175.8 m^3 and 733.9 m^3. Due to the limitations in experiments, the sintering machine end stack and converter stack were not sampled.

A high volume air sampler (Staplex-TFIA-Z) was used to sample ambient air in the countryside 11 km upwind of the plant and at the platform of the sintering machine. Glass fiber filters were changed every 1–2 days. The sampling sites were set at places 1.5 m higher than the plane of reference. The actual volumes of aerosols collected in the countryside and at the platform of the sintering machine were 2422.7 m^3 and 2005.2 m^3.

Solid samples included raw materials and products from main processes. In the process of stack sampling, these solid samples were collected simultaneously.

2.3. Radon measurement

Radon concentrations in the stacks and in ambient air inside the factory were measured with RAD7 radon detector from DURRIDGE Company Inc. (USA). It used planar silicon α particle detector with a radon concentration range as 4.0–10^5 Bq/m^3.

The measuring period was set at two hours. Measuring time at every measurement site reached over 24 h. In the process of data analysis, the first measured data was discarded and the rest data was used to calculate the average value. In order to assure 0–40 °C of measurement temperature in RAD7, a curved copper pipe for cooling was used to connect the sampling outlet in the stack and the sampling tube of RAD7. In the outdoor measurements on the platform of the sintering machine, in the coke plant and the steelmaking plant, the sampling outlet was kept at least one meter away from the building and 1.2–1.5 m from the ground. This is to prevent the interference of radon emanation from walls and the ground. The weather was sunny during the sampling period, so it was not influenced by rainwater.

Table 1
Basic parameters in the integrated steel plant in Jiangxi Province.

Process	Product	Annual production (t/a)	Stack type	Stack gas flow (m^3/h)
Coke oven	Coke	337625	Coke oven stack	86400
Sintering machine	Sinter	1241000	Sintering machine head stack	720000
			Sintering machine end stack	250000
Blast furnace	Pig iron	2196899	Blast furnace stack (Cast house dedusting stack)	963800
Steel converters	Crude steel	4133676	Converter stack (Secondary dedusting stack)	1241000