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Analysis of Air Discharge and Disused Air Filters in Radioisotope Production Facility

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When air discharged from a radioisotope production facility is contaminated with radiation, the public may be exposed to radiation. The objective of this study is to manage such radiation exposure. We measured the airborne radioactivity concentration at a 30 MeV cyclotron radioisotope production facility to assess whether the exhaust gas was contaminated. Additionally, we investigted the radioactive contamination of the air filter for efficient air purification and radiation safety control. To measure the airborne radiation concentration, specimens were collected weekly for 4 h after the beginning of the radioisotope production. Regarding the air purifier, five specimens were collected at different positions of each filter—pre-filter, high-efficiency particulate air filter, and charcoal filter—installed in the cyclotron production room. The concentrations of F-18, I-123, I-131, and Tl-201 generated in the radioiodine production room were 13.5 Bq/m^3, 27.0 Bq/m^3, 0.10 Bq/m^3, and 11.5 Bq/m^3, respectively; the concentrations of F-18, I-123, and I-131 produced in the radioisotope production room were 0.05 Bq/m^3, 16.1 Bq/m^3, and 0.45 Bq/m^3, correspondingly; and those of F-18, I-123, I-131, and Tl-201 generated in the accelerator room were 2.07 Bq/m^3, 53.0 Bq/m^3, 0.37 Bq/m^3, and 0.15 Bq/m^3, respectively. The maximum radiation concentration of I-123 generated in the radioiodine production room was 1,820 Bq/g, which can be disposed after 2 days. The maximum radiation concentration of Tl-202 generated in the radioisotope production room was 205 Bq/g, and this isotope must be stored for 53 days. The I-123 generated in the radioiodine production room had a maximum concentration of 1,530 Bq/g and must be stored for 2 days. The maximum radiation concentration of Na-22 generated in the radioisotope production room was 0.18 Bq/g and this isotope must be disposed after 827 days. To manage the exhaust, the efficiency of air purification must be enhanced by selecting an air purifier with a long life and determining the appropriate replacement time by examining the differential pressure through systematic measurements of the airborne radiation contamination level.

Key Words: Airborne radiation concentration, Air purifier, Multi-channel analyzer

Introduction

Radioisotope contribute greatly to the advancement of modern medicine and they are gradually implemented in various other fields.[1] The retained radioactivity of sealed radioisotopes increased from approximately 3 billion MBq in 2000 to nearly 11 billion MBq in 2015 and that of unsealed radioisotopes increased from approximately 160 million MBq in 2000 to 640 million MBq in 2015.[2]

However, large-scale radioactive material leakage incidents, such as the Japanese Fukushima nuclear power plant accident on March 11, 2011, are increasing the fear of people for radioactivity.[3] As the exposure to radiation that results from the use of medical radioisotopes is an issue directly related to the health of patients and radiation workers, various studies on exposure doses are continuously performed.[4-6] As the implementation of radioisotopes may cause different forms of ra-

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dioactive contamination depending on the application, appropriate management is required according to the type of occurrence.[7)]

Although in the case of radiation workers, the domestic annual dose limit for radiation exposure is 50 mSv and the accumulated dose for 5 years is limited to 100 mSv or less (as specified in Subparagraph 4, Article 2 of the Enforcement Ordinance of the Nuclear Safety Act), it cannot exceed 1 mSv a year in the case of the general public; thus, people inevitably become more sensitive to exposure.[8)] In particular, as the contaminated air that is discharged from a radioisotope work area or production facility may affect the surrounding environment and expose the general public to radiation, it must be properly managed.

For this reason, in this study, we analyzed the possibility of external contamination by measuring the radioactivity concentration in the air discharged from a medical radioisotope production facility. Furthermore, we obtained the data for efficient air purification and radiation safety control by analyzing the radioactivity concentration in the waste of the air purifier filter that is directly connected to the exhaust.

Materials and Methods

We analyzed the radioactivity concentration in the air discharged from the radioisotope production facility of Korea Institute of Radiological & Medical Sciences for various nuclides produced for research and treatment, such as F-18, I-123, I-131, Tl-201, and Tl-202. Specifically, we measured the radioactivity concentrations accumulated in the air and in different types of filters used for air discharge and evaluated their contamination levels. Additionally, we calculated the clearance time for the air purifier waste and reviewed the subsequent management process.

1. Airborne radioactivity concentration

From January until December 2015, a study was performed to analyze the radioactivity concentration in the air discharged from the 30 MeV cyclotron production facility, which is the largest producer of radioisotopes in the Korea Institute of Radiological & Medical Sciences. The target location was the exhaust system installed on the roof above the fifth floor of the cyclotron facility. The exhaust system purifies the contaminated air that is generated in the radioiodine production room, cyclotron accelerator room, and the production room of other radioisotopes and discharges it to the surrounding environment. The radioactive particles contained in this air were collected with a charcoal filter after they passed through the exhaust filters. An air sampler (a portable digital flow meter with a low-volume air sampler, F&J Specialty, USA) and a charcoal filter were installed on the roof above the fifth floor of the cyclotron facility to filter the air from each sector of the cyclotron facility (Fig. 1). Measurements were conducted once a week and specimens were collected by sampling approximately 9 m^3 of air for 4 h at the beginning of radioisotope production (between 9 and 10 a.m.).

2. Radioactive waste of air purifier

For the analysis of the radioactive contamination level of the air purifier, the waste of the following filters was collected on January 14, 2016, when all the filters were replaced: the pre-filter, high-efficiency particulate (HEPA) filter, and charcoal filter installed in the radioiodine production room on August 13, 2014; the pre-filter and HEPA filter installed in the cyclotron accelerator room on April 13, 2015; and the pre-filter and HEPA filter installed in the radioisotope production room on February 22, 2013. Five specimens were collected at

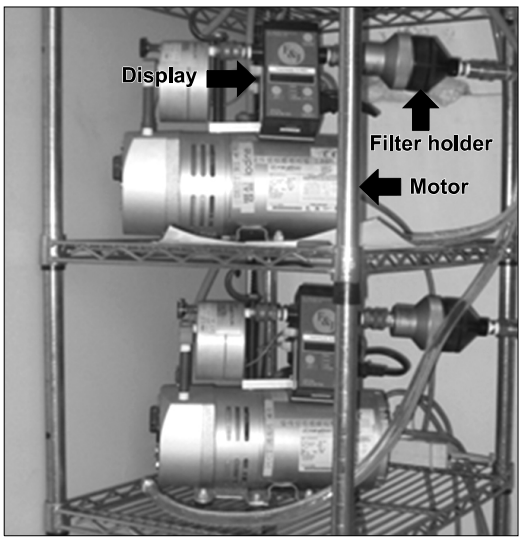

Fig. 1. Setup of air samplers used for collecting the exhausted air from the radioisotope production facility.

different positions in each filter and all the specimens were stored in 90 ml plastic containers (Fig. 2).

3. Radioactivity measurement method

As the radioactivity concentrations of the filter specimens were not high compared to the natural radioactivity level, a high-precision measurement instrument was required. Additionally, a certified reference material was used to determine the measurement efficiency of the instrument and the geometric shape of the measurement specimen. The measurements were conducted as follows:

1) Measurement of radioactivity concentration: A high-purity Ge (Li) semiconductor detector (HPGe, Canberra, USA) were used to detect the nuclides and measure the radioactivity concentration of the collected filter specimens. The results were analyzed in accordance with the Genie-2000 (Canberra, USA) operation procedure. The preparation of the specimens and analysis process were as follows:

(1) The collected specimens were stored in 90 ml plastic containers and their weights were measured thrice using electronic scales (AJH-2200 ED, ViBRA Shinko Denshi, Japan).

(2) The contamination level of the discharged air was analyzed by applying the measurement efficiency of the charcoal filter and the filter specimen data were analyzed using the measurement efficiency of the 90 ml plastic container.

(3) If the dead time of the MCA was above 3%, the measurement was performed after it had decreased to less than 3% or with the specimen positioned 5 cm or 10 cm away from the detector.

2) Setup efficiency calibration using a certified reference material: A certified reference material (CRM) is typically required to accurately measure the radioactivity that corresponds to the geometric configuration of the specimen on the measuring instrument. Thus, the measurement efficiency is determined using the shape and size of the containers that are measured. In this study, a CRM with the same shape and size as the 90 ml plastic containers was produced by the Korea Research Institute of Standards and Science and the efficiency calibration of the charcoal sampler and the 90 ml plastic container was performed on July 11, 2015 using a standard source (Fig. 3).

3) Calculation of clearance time for air purifier waste: The clearance time for radioactive waste was calculated in accordance with Article 107 (Procedures and Methods for Self-disposal of Radioactive Waste)[9] and the permitted concentration was determined according to [attached Table 1] 'Concentration of Each Radioactive Nuclide allowed to be self-disposed' of the Nuclear Safety and Security Commission announcement no. 2014-003 (related to Subparagraph 2 of Article 2 and Paragraph 1 of Article 3).[10] The clearance time was estimated using the average life, which is proportional to the measured radioactivity concentration.

Clearance time:

$$=-1.443\times\text{half life (D)}\times\ln\left[\frac{permitted\,activity\ (Bq/g)}{maximum\ activity\ (Bq/g)}\right] \quad (1)$$

Fig. 2. Samples collected from the charcoal filter (left), HEPA filter (middle), and pre-filter (right), stored in 90 ml plastic containers.

Fig. 3. Photograph of CRMs used to determine the measurement efficiency of the MCA.

Table 1. Monthly nuclide activity concentrations at different air sampling locations.

Air sampling position	Nuclide	Activity (Bq/m^3)												
		Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec	Aug
Iodine production room	F-18	20.0	9.19	-	0.08	12.7	13.0	82.7	1.62	4.47	1.50	1.00	2.30	13.5
	I-123	14.4	40.6	9.15	48.7	0.30	8.55	124	61.1	7.67	3.35	3.40	2.89	27.0
	I-131	-	0.07	-	-	-	0.12	-	-	-	-	-	-	0.10
	Tl-201	-	11.5	-	-	-	-	-	-	-	-	-	-	11.5
Radioisotope production room	F-18	0.29	1.38	-	-	-	0.20	-	0.34	-	-	-	-	0.55
	I-123	65.6	8.88	2.51	2.02	3.60	2.46	27.9	61.5	5.34	3.66	2.46	7.51	16.1
	I-131	0.16	0.07	-	-	0.05	0.29	1.59	0.64	0.29	0.71	0.21	-	0.45
Accelerator room	F-18	-	-	-	4.79	1.53	1.62	2.80	1.63	0.02	-	-	-	2.07
	I-123	53.2	80.5	97.0	-	17.6	54.4	25.9	87.6	65.0	37.1	54.5	10.5	53.0
	I-131	0.09	0.97	0.20	0.27	0.07	0.30	0.60	0.29	0.36	0.49	0.46	0.30	0.37
	Tl-202	-	-	-	0.30	0.04	0.12	-	-	-	-	-	-	0.15

-: None detectable.

Results

1. Measurement of airborne radioactivity concentration

The nuclides and the radioactivity concentrations measured in each medical radioisotope working area for 1 year are shown in Table 1. In the case of the radioiodine production room, four types of nuclides were detected. Additionally, the airborne radioactivity concentration of I-123 was relatively high compared to that of the other nuclides, with an average value of 27.0 Bq/m^3. Regarding the highest monthly radioactivity concentration, the airborne radioactivity concentration of F-18 in July was 82.7 Bq/m^3 and the average airborne radioactivity concentration of I-123 in July and August were measured to be 124 Bq/m^3 and 61.1 Bq/m^3, respectively. Overall, the concentration of I-131 was low compared to that of the other nuclides, with an average concentration of 0.10 Bq/m^3. The concentration of Tl-201 was 11.5 Bq/m^3 only in February, while no Tl-201 was detected in the other months. The radioactivity concentration of I-123 in the radioisotope production room was highest in January with a value of 65.6 Bq/m^3. Similar to I-131, the concentration of F-18 was less than 2 Bq/m^3 and the average concentrations of F-18 and I-131 were 0.55 Bq/m^3 and 0.45 Bq/m^3, respectively. In the accelerator room, the average concentrations of F-18, I-131, and Tl-202 were 2.07 Bq/m^3, 0.37 Bq/m^3, and 0.15 Bq/m^3, respectively and I-123 had the highest concentration in March with a value of 97.0 Bq/m^3. The results show that the discharged air contained less than 200 Bq/m^3 of F-18, 300 Bq/m^3 of I-123, 3 Bq/m^3 of I-131, 1,000 Bq/m^3 of Tl-101, and less than 300 Bq/m^3 of Tl-202, which correspond to the discharge control standards established by the Nuclear Safety and Security Commission.

2. Measurement of radioactivity concentration in air purifier waste and calculation of clearance time

Tables 2, 3, and 4 show the average and maximum radioactivity concentrations, the uncertainty of the measurement, and the clearance time of the filter waste collected from the air purifier at each position. The clearance time was calculated based on the maximum radioactivity concentration for safety control. Table 2 shows the results for the charcoal filter specimen; two nuclides could be identified in the charcoal filter, which was used for the discharge of air from the radioiodine production room. I-123 and I-131 showed average radioactivity concentrations of 1,270 Bq/g and 1.13 Bq/g, respectively. In accordance with the allowable concentration standard, which considers the half-life of the isotope, the clearance time for I-123 was calculated to be 2 days; therefore, I-131 can be disposed immediately.

Table 3 shows the analysis results for the HEPA filter, through which air was discharged from the radioisotope production room. The nuclide Tl-202 was detected in the HEPA filter with average and maximum concentrations 130 Bq/g and

Table 2. Nuclide activity concentrations and corresponding clearance times at the sampling position of the charcoal filter.

Air filter sampling position	Nuclide	Activity (Bq/g)		(Mean) Uncertainty (%)	(Max.) Clearance time (day)
		Average	Maximum		
Radioiodine production room	I-123	1270	1820	6.51	2
	I-131	1.13	1.48	2.91	Direct (0)

Table 3. Nuclide activity concentrations and clearance times at the sampling of the HEPA filter.

Air filter sampling position	Nuclide	Activity (Bq/g)		(Mean) Uncertainty (%)	(Max.) Clearance time (day)
		Average	Maximum		
Radioisotope production room	Tl-202	130	205	1.33	53

Table 4. Nuclide activity concentrations and the clearance times at the sampling of the pre-filters.

Air filter sampling position	Nuclide	Activity (Bq/g)		(Mean) Uncertainty (%)	(Max.) Clearance time (day)
		Average	Maximum		
Radioiodine production room	I-123	1240	1530	7.00	2
Radioisotope production room	Na-22	0.18	0.18	9.57	827
	Tl-202	167	339	1.26	62
Accelerator room	Co-57	0.33	0.61	6.81	Direct (0)
	Tl-202	0.76	0.76	6.81	Direct (0)

205 Bq/g, respectively. The clearance time was calculated to be 53 days based on the maximum radioactivity concentration.

Table 4 shows the results for the pre-filter, which was used for the discharge of air from the radioiodine production room. I-123 was detected in the pre-filter specimen with average and maximum concentrations of 1,240 Bq/g and 1,530 Bq/g, respectively. Based on these findings, the pre-filter must be stored for 2 days. Two nuclides, Na-22 and Tl-202, were detected in the radioisotope production room; their maximum concentrations were 0.18 Bq/g and 339 Bq/g, respectively, and their clearance times were calculated to be 827 days and 62 days. Two nuclides were detected in the 30 MeV cyclotron accelerator room, Co-57 and Tl-202, with average radioactivity concentrations of 0.33 Bq/g and 0.76 Bq/g, respectively. Both nuclides were evaluated to be immediately dischargeable.

Discussion

Most medical institutes aim to maintain the exposure dose below the standard value using shields and personal exposure dosimeters for the exposure control of radiation workers during the use of radioisotopes. However, the discharge of radioactive materials to the environment is ignored compared to internal control. In medical institutes that handle unsealed radioisotopes, radioactive contamination of the air is unavoidable. If this is not properly controlled within the medical institutes, secondary proliferation to the external environment may occur; therefore, thorough radiation safety control is required. For this reason, this study evaluated whether external radioactive contamination was appropriately controlled by analyzing the airborne radioactivity concentration that is directly related to the exhaust of medical radioisotope work areas and that of air purifier waste.

Although the doses measured on the roof of the 30 MeV cyclotron production facility immediately before the air discharge generally showed concentrations lower than the limit, relatively high values were measured in July for the radioiodine production room, such as those of F-18 and I-123.

These results appeared to be temporarily high owing to the multiple works performed, as there was no change in the structure. However, as these concentrations were not above the standard value, they seemed to have no significant effect on human health. Tl nuclides were detected less frequently than other nuclides because of difficulties in the data collection, as the applied measurement times differed significantly from the production times of Tl family nuclides. Nevertheless, considering the very low radioactivity concentrations in the examined months, the degree of external contamination caused by Tl family nuclides is considered to be negligible.

The analysis results of the air purifier specimens showed that the use of only one type of filter involves difficulties in evaluating the radiation safety control conditions owing to the physical properties of the different nuclides. Additionally, the replacement cycle of the air purifier must be determined depending on the nuclides used in each production facility. In particular, it is considered to be appropriate to calculate the clearance time based on the maximum radioactivity concentration to conservatively control radiation safety issues in accordance with the radiation exposure control principle ALARA (As Low As Reasonably Achievable).

A past study on an efficient exhaust control plan for medical institutes that use radioisotopes reported that the air purifier management method must be changed from artificial replacement to periodic replacement; in the latter approach, the replacement is performed when the initial differential pressure is doubled in order to determine an appropriate point of compromise between exhaust control regulations and the air purifier replacement cycle.[11] Moreover, a paper on an efficient control plan for HEPA filters described comparative research on the life and differential pressure of HEPA filters; the authors stated that the filter replacement cost and the generation of radioactive waste can be reduced by selecting a suitable filter.[12] Therefore, as the air purifier is a key factor for the discharge of air to the environment, the selection of an efficient air purifier in accordance with the financial situation of the institute and its periodic replacement are considered to be helpful for radiation safety control.

Conclusion

The analysis of the radioactivity concentrations measured in the air and the air purifier when radioisotopes were handled in the 30 MeV production facility showed differences in the contamination level depending on the time of measurement and the type of air purifier. To control the exhaust, the efficiency of air filter must be enhanced by selecting an air purifier with a long life. Additionally, an appropriate replacement cycle must be selected by assessing the differential pressure through systematic measurements of the airborne contamination level. This study is expected to contribute to efficient air purification and radiation safety control.

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