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Radioactivity concentrations of natural radionuclides in fine dust of Jeju, Korea

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Abstract: Radioactivity concentrations for natural radionuclides were determined from fine dust samples collected in Jeju, Korea according to atmospheric events (Asian dust, haze, fog-mist, and non-event), and radium equivalent activity was calculated. The mean atmospheric radioactivity concentrations for ^{238}U, ^{232}Th, and ^{40}K in 127 fine dust samples were 0.49, 0.24, and 7.23 µBq m^{-3}, respectively, and the radium equivalent activity was 33.25 Bq kg^{-1}. The mean concentrations of ^{238}U and ^{232}Th in the fine dust during the Asian dust period were 1.31 and 1.60 µBq m^{-3}, respectively, above the global average, while the values for the other three atmospheric events were lower. The ratio of $^{232}Th/^{238}U$ radioactivity during the Asian dust period was 1.22, higher than the ratio for the other three atmospheric events.

Key words: natural radionuclides, fine dust, atmospheric phenomenon

1. Introduction

Humans are always exposed to natural radiation associated with non-decaying radionuclides such as ^{40}K and the daughter radionuclides of the ^{232}Th and ^{238}U decay series. We receive over 80 % of our radiation from these nuclides.[1,2] Radiation is commonly found in rocks, soils, oceans, houses, and more,[3] so, radiation levels vary by region due to different concentrations of nuclides derived from soil, fertilizers, or their equivalents.[1,4]

Many studies have been performed to measure these radioisotope activities released from rocks and soils, and the dose gamma radiation on human was estimated by calculation.[1,5-11] Besides the rocks and soils, internal radiation doses from tobacco, ingestion, building materials, fine dust, etc. have also been studied.[12-16]

In 2015, as in 2014, fine dust (PM_{10} aerosols) was collected from Gosan site on Jeju's Halla mountain in Korea, which is largely unaffected by urban air pollutants and serves as a background for estimating air pollution on the Korean Peninsula.[17] The area is centrally located between China, Taiwan, Japan, and the Korean Peninsula, making it a good place to monitor the characteristics of pollutants traveling

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long distances from neighboring countries.

In general, natural radionuclides are easily measured by mass spectrometry, because they have too low a gamma-ray emission rate or a very low branching ratio from the parent nuclide to be measured by conventional gamma spectrometry. Therefore, instead of directly measuring gamma radiation, we used inductively coupled plasma - mass spectrometry (ICP-MS) to measure the radioactivity concentrations of ^{40}K, ^{232}Th and ^{238}U in the fine dust and compare the results with those from 2014.

2. Experimental

2.1. Sample preparation and analysis

Jeju Island faces the Asian continent, and the Gosan site where the fine dust was collected is located at 33°1′N and 126°10′E on the western side of Jeju Island.

Sampling was performed using an automated system, PMS-103 (APM Engineering, Korea), equipped with a Teflon filter (47 mm/2.0 μm) for 24 hours every three days during whole year of 2015, the same as the previous collection method.[16] It maintained a flow rate of 16.7 L/min with mass flow controller (MFC) to collect fine dust.

Again, as in reference 16, the aerosol filter sample was placed in a perfluoroalkoxy Teflon container in a microwave digestion system, and 10 mL of a 5.55 % HNO_3/16.75 % HCl acid solution was added.[18] This solution was heated in a 180 °C and 1000 W microwave for 15 min to digest the particulate matter, and the digested solution was filtered through a 45 μm PVDF syringe filter. To the solution, 5 mL of 3 % HNO_3/ 8 % HCl acid solution and ultrapure water were added until the volume was 25 mL.[19] Three species, ^{39}K, ^{232}Th and ^{238}U, were measured by ICP-MS (ICP-DRC-MS, Perkin Elmer, ELAN DRC-II, USA) under the conditions of 1500 W RF power, 40 MHz RF frequency, 15.0 L min^{-1} cooling water, 0.9-1.05 L min^{-1} Ar flow rate, and assist 1.2 L min^{-1}.

2.2. Concentrations of isotopic activity

Based on ICP-MS measurements of potassium, thorium, and uranium in fine dust, the radioactivity concentrations of the isotopes ^{40}K, ^{232}Th, and ^{238}U could be calculated as follows:[20]

$$A_i = \frac{\ln 2}{T_{1/2}} \times \frac{R_i \times m_e}{M_i} \times N \quad (1)$$

Here, A_i: radioactivity concentration (Bq m^{-3})

$T_{1/2}$: half-life time (s) of isotope i (^{40}K; 1.28×10^9 y, ^{232}Th; 1.405×10^{10} y and ^{238}U; 4.468×10^9 y)

R_i: isotopic ratio (natural abundance) of isotope i (^{40}K; 0.000117, ^{232}Th; 1.0 and ^{238}U 0.99275, based on the IUPAC report 2009[21])

m_e: mass concentration of element e corresponding to isotope i (g m^{-3})

M_i: atomic mass (g mol^{-1})

N: Avogadro's number (6.022×10^{23} mol^{-1})

2.3. Radium equivalent activity

The radioactivity concentrations per unit mass for the dust above were used to calculate the radium equivalent activity (Ra_{eq}). The Ra_{eq} is an indicator of radiation hazard from dose of daughter radionuclides contained in a material, which is often expressed as a biohazard index.[22] This Ra_{eq} was set under the assumption that the γ dose rate is the same for nuclides ^{238}U = 370, ^{232}Th = 259, and ^{40}K = 4810 Bq kg^{-1} per unit mass. In this study, the concentrations of the nuclides (^{40}K, ^{232}Th, and ^{238}U) in PM_{10} samples collected in 2015 were calculated as dust masses, and the Ra_{eq} was calculated by utilizing them. The results were then checked to see if they were below the limit of 370 Bq kg^{-1}.[23]

$$Ra_{eq} = \left(\frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810}\right) \times 370 \quad (2)$$

Here, Ra_{eq}: radium equivalent activity

A_U: activity concentration (Bq kg^{-1}) of ^{238}U

A_{Th}: activity concentration (Bq kg^{-1}) of ^{232}Th

A_k: activity concentration (Bq kg^{-1}) of ^{40}K

3. Results and Discussion

3.1. Radioactivity concentration per dust mass

Total 127 fine dust samples were collected at

Table 1. Comparison of ^{40}K, ^{232}Th, and ^{238}U mass and radioactivity concentrations per dust mass of Gosan fine dust

Samples	^{40}K		^{232}Th		^{238}U		Ref.
	mg kg^{-1}-dust	Bq kg^{-1}-dust	mg kg^{-1}-dust	Bq kg^{-1}-dust	mg kg^{-1}-dust	Bq kg^{-1}-dust	
2014 (n=115)	0.56±0.38	146.90±101.84	1.03±0.73	4.16±2.98	0.53±0.60	6.48±7.43	16
2015 (n=127)	0.62±0.41	163.31±107.69	1.15±1.16	4.65±4.71	1.14±0.91	14.05±11.16	

Gosan site during 2015. Among them, 7 samples on Asian dust events, 62 samples on normal weather days (non-event days), 10 samples on haze days, and remaining 48 samples on fog-mist days were collected. All of these samples were analyzed by ICP-MS to determine the radioactivity concentrations of ^{40}K, ^{232}Th, and ^{238}U in the fine dust.

Table 1 shows the concentrations of the masses of the isotopes ^{40}K, ^{232}Th, and ^{238}U of whole fine dust samples in 2014 and 2015, as well as the radioactivity concentration per mass of dust. The mean mass concentration of whole fine dust samples in 2015 was 41.54 μBq m^{-3}, which was similar to the 2014 concentration.[16] The mean mass concentrations of ^{40}K, ^{232}Th, and ^{238}U were 0.62±0.4, 1.15±1.2, and 1.14±0.9 mg kg^{-1}-dust respectively. In addition, the mean radioactivity concentrations per dust mass of ^{40}K, ^{232}Th, and ^{238}U were 163.31±107.7 Bq kg^{-1}-dust (range: 3.16~468.23 Bq kg^{-1}-dust), 4.65±4.7 Bq kg^{-1}-dust (range: 0 ~ 42.69 Bq kg^{-1}-dust), and 14.05 ± 11.2 Bq kg^{-1}-dust (range: 2.58~78.82 Bq kg^{-1}-dust) respectively. The global average radiation concentrations of ^{232}Th and ^{238}U are 30 and 35 Bq kg^{-1}-dust, so the concentrations in the measured samples are much lower.[1]

3.2. Activity concentrations per air volume and radium equivalent activity

Table 2 and *Fig.* 1 show the activity concentrations per air volume and radium equivalent activity by month. The overall high levels in February and March are due to the effects of the 2015 Asian dust, which was concentrated on February 22-23 and March 21-22. However, the high radium equivalent activity in September is unusual, and the reasons for it are not yet fully understood. Nevertheless, all ^{232}Th and ^{238}U activity concentrations and radium equivalent activity values are below than the global average of mean atmospheric activity concentrations and radium equivalent activity which are 0.5, 1.0 μBq m^{-3}, and 310 Bq kg^{-1}, respectively.[1] This is expected due to the nature of the Gosan region, where relatively low dust levels are present, whereas the global average is obtained based on total suspended particles.

Table 2. The activity concentrations of ^{40}K, ^{232}Th, and ^{238}U per air volume and radium equivalent activity per month at Gosan

Year-month (n)	^{40}K (μBq m^{-3})	^{232}Th (μBq m^{-3})	^{238}U (μBq m^{-3})	Ra_{eq} (Bq kg^{-1})
2015-Jan (8)	7.19±7.02	0.14±0.12	0.31±0.20	22.95±10.24
2015-Feb (13)	15.59±13.83	0.77±1.10	0.92±0.75	40.68±17.04
2015-Mar (11)	14.52±13.77	0.49±0.51	0.63±0.45	30.54±14.53
2015-Apr (6)	5.73±7.11	0.20±0.21	0.40±0.38	21.78±10.61
2015-May (11)	2.56±1.78	0.08±0.07	0.30±0.27	21.88±15.53
2015-Jun (11)	6.71±7.47	0.16±0.31	0.28±0.37	22.21±14.35
2015-Jul (11)	3.04±1.54	0.10±0.04	0.45±0.14	31.43±16.39
2015-Aug (11)	4.52±4.21	0.12±0.07	0.59±0.43	31.76±23.62
2015-Sep (14)	4.95±2.25	0.23±0.42	0.50±0.22	54.28±25.52
2015-Oct (12)	10.98±5.84	0.25±0.12	0.52±0.21	44.27±15.21
2015-Nov (9)	5.21±4.29	0.09±0.05	0.33±0.10	34.08±8.46
2015-Dec (10)	3.14±1.83	0.09±0.04	0.44±0.28	26.63±13.89
Total mean (127)	7.23±8.29	0.24±0.46	0.49±0.40	33.25±19.17

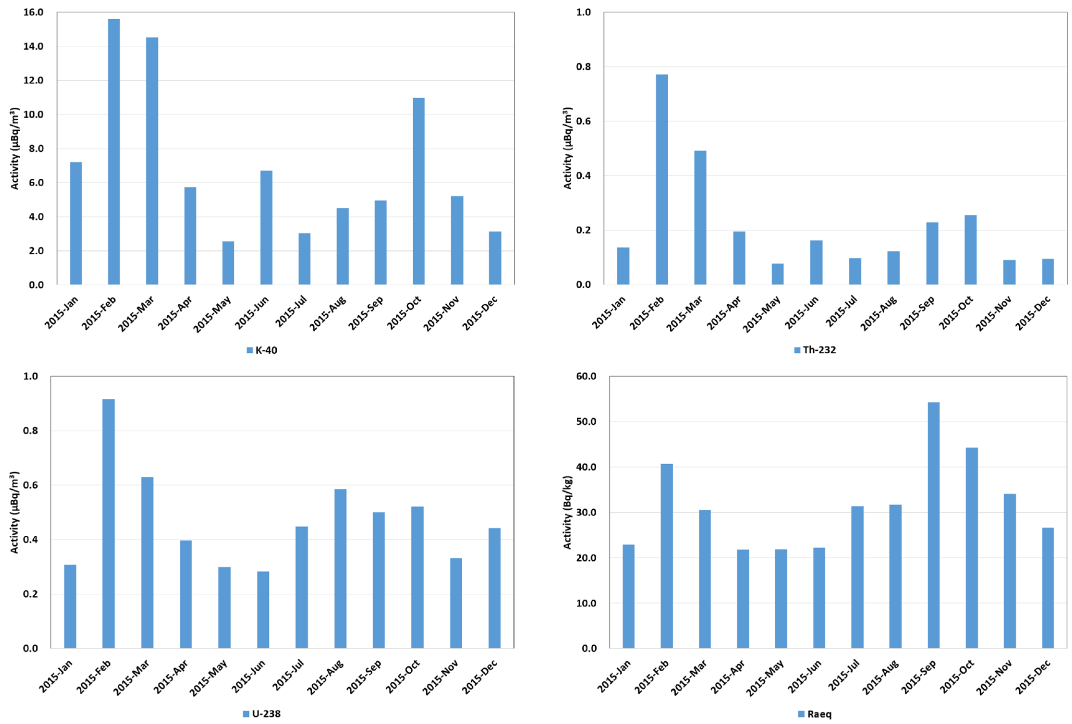

Fig. 1. Histogram of *Table* 2 (The activity concentrations of ^{40}K, ^{232}Th, and ^{238}U per air volume and radium equivalent activity per month at Gosan).

Table 3. The activity ratio of ^{40}K, ^{232}Th, and ^{238}U activity concentrations (μBq m^{-3}) by atmospheric phenomenon at Gosan site in Jeju

Atmospheric Phenomenon (*n*)	^{40}K (μBq m^{-3})	^{232}Th (μBq m^{-3})	^{238}U (μBq m^{-3})	^{232}Th/^{238}U
Asian Dust (7)	27.98±14.6	1.60±1.2	1.31±0.7	1.22
Haze (10)	18.39±8.4	0.41±0.3	0.77±0.5	0.53
Fog-Mist (48)	4.65±4.7	0.11±0.1	0.39±0.3	0.28
Non-Event (62)	5.09±3.7	0.17±0.2	0.46±0.3	0.37
Whole (127)	7.23±8.3	0.24±0.5	0.49±0.4	0.49

The radioactivity concentrations by atmospheric phenomenon are summarized in *Table* 3. The mean radioactivity concentrations of ^{40}K, ^{232}Th, and ^{238}U were 7.23±8.3 μBq m^{-3}, 0.24±0.5 μBq m^{-3}, and 0.49±0.3 μBq m^{-3}, respectively. The radium equivalent activity was 33.25 Bq kg^{-1}.

The mean atmospheric radioactivity concentrations of ^{232}Th and ^{238}U per air volume obtained from 7 fine dust samples during the Asian dust event were 1.60 ±1.2 μBq m^{-3} (range: 0.23 to 3.63 μBq m^{-3}) and 1.31±0.7 μBq m^{-3} (range: 0.37 to 2.18 μBq m^{-3}), respectively, which exceeded the global reference values. These were 5.5, 9.4, and 2.9 times higher than the mean atmospheric radioactivity concentrations of ^{40}K, ^{232}Th and ^{238}U per air volume, respectively, from 62 samples during the non-event period. The ^{232}Th/^{238}U ratio was also high for Asian dust, at 1.22, compared to other atmospheric phenomena (0.28-0.53), including haze, fog-mist, and non-event cases.

Fig. 2 shows the correlations of the investigated natural isotopes. The correlation between ^{232}Th and ^{238}U activity concentrations in total fine dust was

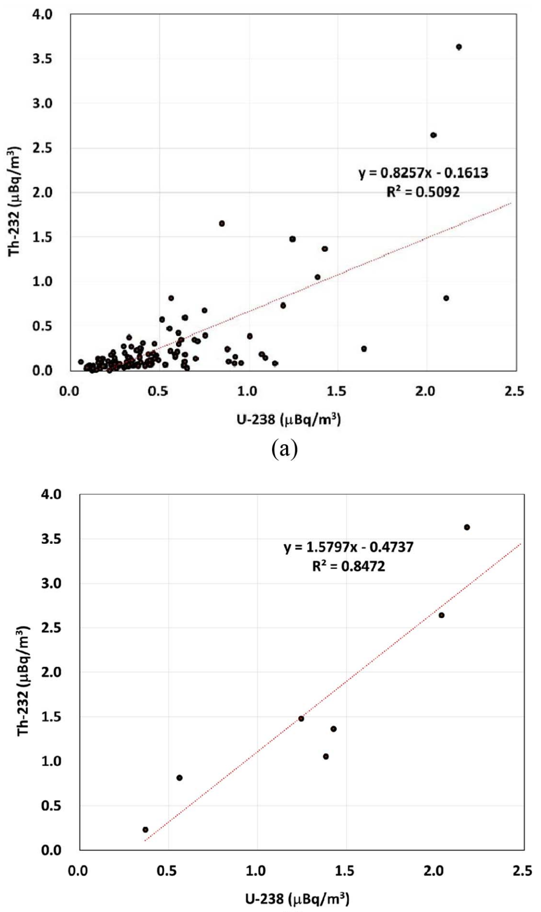

Fig. 2. Correlation between ^{232}Th and ^{238}U concentrations in (a) whole fine dust and (b) Asian dust.

positive (R^2 = 0.5092, p-value < 0.0001). This confirms that the two isotopes come from the same source, i.e., the Earth's crust (*Fig.* 2(a)). The correlation between ^{232}Th and ^{238}U activity concentrations in Asian dust is positive (R^2 = 0.8472, p-value = 0.125), suggesting that dust from China has a higher ^{232}Th content than dust from other regions (*Fig.* 2(b)).

3.3. Air mass inflow paths

Using the National Oceanic and Atmospheric Administration's (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT4) model,[24] the 5-day backward trajectories of air entering the Gosan site was analyzed. The percentage of air entering the Gosan site on the day of the measurement was

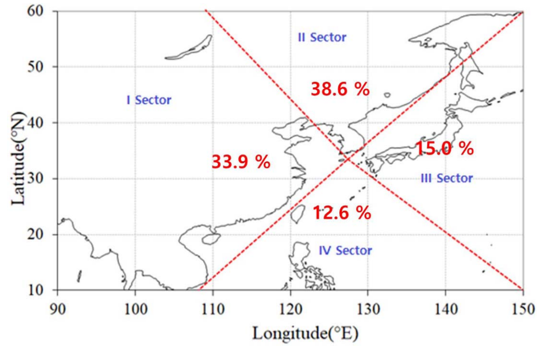

Fig. 3. Results of Gosan site backtracking for 5 days during the study period.

33.9 %, 38.6 %, 15.0 %, and 12.6 % for Sector I (Mainland China), Sector II (Korean Peninsula), Sector III (Japan), and Sector IV (North Pacific), respectively (*Fig.* 3).

The ^{40}K, ^{232}Th and ^{238}U concentrations in sector I were 11.23, 0.43, and 0.65 μBq m^{-3}, respectively, which were greater than those in other sectors (sector II: 6.28±6.9, 0.19±0.3, 0.40±0.3 μBq m^{-3}; Sector III: 4.13±3.2, 0.08±0.1, 0.46±0.4 μBq m^{-3}; Sector IV: 3.08±3.1, 0.10±0.1, 0.38±0.3 μBq m^{-3}). They are, however, still lower than the global averages of the mean atmospheric activity concentrations of ^{232}Th and ^{238}U. The ratio of ^{232}Th/^{238}U activity in Sector I was 0.66, also higher than the other sector ratios (Sector II: 0.48, Sector III: 0.17, Sector IV: 0.26). Since Sector I is the dominant source of atmospheric inputs on Asian dust days, it can be said that this sector has a higher ^{232}Th content than dust sources in other regions.

4. Conclusions

Radioactivity concentrations of natural radionuclides by atmospheric event (Asian dust, haze, fog-mist, and non-event) were measured in 127 particulate matter samples from Jeju Island collected throughout 2015.

The mean atmospheric radioactivity concentrations of ^{238}U, ^{232}Th, and ^{40}K in all samples were 0.49, 0.24, and 7.23 μBq m^{-3}, respectively, which is lower than the global average concentration in fine dust. The

$^{232}Th/^{238}U$ radioactivity ratio for the entire sample was 0.49. The average concentrations of ^{238}U and ^{232}Th in fine dust during the Asian dust period were 1.31 and 1.60 μBq m^{-3}, respectively, which were higher than the global average concentrations and the values for the other three atmospheric events. The $^{232}Th/^{238}U$ radioactivity ratio for the Asian dust period was 1.22, which was higher than the ratios for the other three atmospheric events.

After analyzing the concentrations by dividing the airborne pathways by 4, it was found that the mainland China pathway had higher concentrations than the other three pathways. The $^{232}Th/^{238}U$ radioactivity ratio of the mainland China pathway was 0.66, which was higher than the other pathway ratios.

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