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Measurement of activity concentrations of ^{40}K, ^{232}Th and ^{238}U in TSP aerosols and the associated inhalation annual effective radiation dose to the public in Gosan site, Jeju

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1. Introduction

Gamma radiation emitted from naturally occurring radioisotopes, such as ^{40}K and the radionuclides from the ^{232}Th and ^{238}U series and their decay products, which exist at trace levels in all ground formations, represents the main external source of irradiation to the human body [1-3]. Radioactivity of aerosol particles depends on their sources. Therefore, it is expected that naturally occurring radionuclides are the main contributor to TSP radioactivity. The objective of the current study is to determine the activity concentrations of ^{40}K, ^{232}Th and ^{238}U in airborne TSP and the associated internal radiation dose to the public due to inhalation in Gosan site, Jeju Island, Korea.

2. Materials and methods

2.1. Air sampling locations

Air sampling for TSP aerosols was conducted at the Gosan site (33°17′N, 126°10′E), which is located on the seashore hill of 72 m above sea level at the western edge of Jeju Island, located approximately 100 km south of the Korean peninsula.

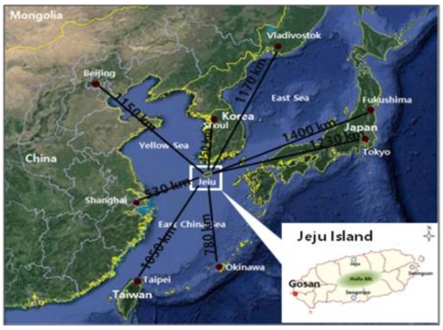

Fig. 1. Location of air sampling site in Jeju Island.

2. 2. Collection of atmospheric aerosols

The TSP samples have been collected using high volume tape sampler (KIMOTO Electric Co., 195A, Japan), which is an automatic system with roll type PTFE filters (Sumitomo Electric Co., 100 mm×10 m) at 24 hour basis with every 3 day intervals from January to April of 2013 (Asian dust generation period). On collecting samples, the air flow rate was kept to about 170 L/min, and total air flow was calculated from the flow rate and running time.

2. 3. Sample Analysis

For the analysis of TSP elemental species, the aerosol samples were decomposed with acids using a microwave digestion system (Milestone, START D, Italy), where aerosol filters were put in a PFA (perfluoroalkoxy) teflon vessel of the microwave oven together with 10 mL acid solution (5.55% HNO_3/16.75% HCl). This vessel was heated at 180℃ for 15 minutes with 1000 W microwave radiation to digest the TSP aerosols. The decomposed solution was transferred through syringe filter (Whatman, PVDF, 0.45 μm) into a 25 mL volumetric flask, and the volume was adjusted to 25 mL with 5 mL acid solution (3% HNO_3/8% HCl) and ultrapure water [4]. The number of elements determined by ICP-DRC-MS (Perkin Elmer, ELAN DRC-II, USA) instruments was 3 species such as ^{39}K, ^{238}U and ^{232}Th. DRC gas was used as the ammonia. For the elemental analysis, the instrumental detection limits (IDL) of ^{39}K, ^{238}U and ^{232}Th were 27.12 μg/L, 11.38 ng/L and 11.60 ng/L, respectively.

Table 1. Instrumental Detection Limit (IDL) and Variation of Coefficient (CV) for ICP-DRC-MS (n=7).

	^{39}K (μg/L)	^{238}U (ng/L)	^{232}Th (ng/L)
IDL	27.12	11.38	11.60
CV(%)	2.24	1.38	1.48

2.4. Isotope activity concentration

Potassium, thorium and uranium were measured as trace elements in TSP aerosol by ICP-DRC-MS. The radioactivity concentrations of the isotopes ^{40}K, ^{232}Th and ^{238}U in the collected TSP were calculated using the following equation (1) [5]:

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

where A_i, $T_{1/2}$, ρ_i, m_e, M_i, and N are the radioactivity concentration (Bq/m^3), half-life time (s) of isotope i (^{40}K; 1.28×10^9 yr, ^{238}U; 4.468×10^9 yr, ^{232}Th; 1.405×10^{10} yr), isotopic ratio (natural abundance) of isotope i, the mass concentration of element e corresponding to isotope i (g/m^3)), atomic mass (g/mol), and the Avogadro's number (6.022×10^{23} mol^{-1}), respectively. The isotopic ratios for ^{40}K, ^{238}U and ^{232}Th were 0.000117, 0.99275 and 1.0, respectively, based on the International Union of Pure and Applied Chemistry (IUPAC) report 2009 [6].

2.6. Inhalation annual effective dose

The inhalation annual effective radiation dose ($E_{h,i}$) due to TSP was calculated using the following equation (2) adapted from UNSCEAR (2000) [3]:

$$E_{h,i} = A_i \times B \times d_{h,i} (1 - F_0 + F_0F_r) \qquad (2)$$

where A_i is the integrated activity concentration of radionuclide i associated with TSP in outdoor air (Bq/m^3), B is the breathing rate (m^3/y), $d_{h,i}$ is the committed dose per unit intake from inhalation or effective dose coefficient (Sv/Bq), F_o is the indoor occupancy factor and F_r is the ratio of indoor to outdoor air concentration.

3. Results and discussion

3.1. ^{40}K , ^{238}U and ^{232}Th activity concentration in TSP

During the study period, the concentrations of ^{40}K, ^{238}U and ^{232}Th were 5.188 ~ 179.866 pg/m^3 (mean: 54.899±44.311 pg/m^3), 0.496 ~ 83.810 pg/m^3 (mean: 21.372±21.568 pg/m^3) and 0.264 ~ 285.071 pg/m^3 (mean: 55.777±69.592 pg/m^3), respectively. As a result, it was observed during this study that the radioactive concentrations of ^{40}K, ^{238}U and ^{232}Th were within the scope of 1.341 ~ 46.483 $\mu Bq/m^3$ (mean: 14.188± 11.451 $\mu Bq/m^3$), 0.006 ~ 1.032 $\mu Bq/m^3$ (mean: 0.263 ±0.226 $\mu Bq/m^3$), 0.001 ~ 2.579 $\mu Bq/m^3$ (mean: 0.226 ±0.282 $\mu Bq/m^3$), respectively. Furthermore, they are lower than the world averages for the mean atmospheric activity concentrations of ^{232}Th and ^{238}U associated with TSP, which are 0.5 and 1.0 $\mu Bq/m^3$, respectively [3]. The low atmospheric activity concentration of ^{232}Th and ^{238}U might be attributed to the low atmospheric TSP in Gosan site as compared to many of the world cities.

3.2. Activity concentrations by atmospheric phenomenon

Table 2 presents the radioactivity concentrations of the isotopes ^{40}K, ^{238}U and ^{232}Th by atmospheric phenomenon (Asian Dust, Haze, Fog-Mist and Non-Event) at Gosan site in Jeju Island, 2013. During Asian dust periods, the concentrations of ^{40}K (33.221±12.333 $\mu Bq/m^3$), ^{238}U (0.788±0.260 $\mu Bq/m^3$), and ^{232}Th (0.865±0.252 $\mu Bq/m^3$) were highly as 3.59, 5.51 and 7.86 times, respectively, compared to the non-event periods. For the haze event days, ^{40}K (34.729±10.792 $\mu Bq/m^3$), ^{238}U (0.730±0.261 $\mu Bq/m^3$), and ^{232}Th (0.608±0.475 $\mu Bq/m^3$) were highly as 3.75, 5.10 and 5.53 times, respectively, compared to the non-event days. For the fog-mist event days, ^{40}K (19.970±15.824 $\mu Bq/m^3$), ^{238}U (0.371±0.374 $\mu Bq/m^3$), and ^{232}Th (0.329±0.451 $\mu Bq/m^3$) were highly as 2.16, 2.59 and 2.99 times, respectively, compared to the non-event days.

Table 2. Airborne TSP concentrations, the associated ^{40}K, ^{238}U and ^{232}Th activity concentrations by atmospheric phenomenon at Gosan site in Jeju Island, 2013.

Atmospheric Phenomenon	^{40}K (μBq/m^3)	^{238}U (μBq/m^3)	^{232}Th (μBq/m^3)
Asian Dust	33.22±12.33	0.79±0.26	0.87±0.25
Haze	34.72±10.79	0.73±0.26	0.61±0.48
Fog-Mist	19.97±15.82	0.37±0.37	0.33±0.45
Non-Event	9.25±5.96	0.14±0.10	0.11±0.08

The ^{238}U/^{40}K ratio in Asian Dust, Haze, Fog-Mist, and Non-Event were 0.024, 0.021, 0.019, 0.015, respectively, showing a high value in Asian Dust and low in Non-Event. The ^{232}Th/^{40}K ratio of Asian Dust was 0.026, which was about 2.17 times higher than the Non-Event value (0.012). The ^{232}Th/^{238}U ratio of Asian Dust was 1.10, which was higher than those of other atmospheric phenomenon.

3. 3. Inflow Pathways of Air Mass

The five-day backward trajectories have been performed to investigate the inflow pathway of the air mass into the Gosan Site, and it was ascertained using the HYSPLIT4 (HYbrid Single Particle Lagrangian Integrated Trajectory) model of National Oceanic and Atmospheric Administration (NOAA) [7]. The air mass inflow pathways have been classified by four sectors as shown in Fig. 2; China continent (Sector I), Korean peninsula (Sector II) and Japan & North Pacific Ocean (Sector III), The sectional frequencies of air mass inflow into the Gosan area were 48.3 % in Sector I, 10.3 % in Sector II, 3.4 % in Sector III and 37.9 % in the other overlap zone during the study, showing a significant influence from China continent. This explains that most of the air masses are moved from China continent and seriously affect the air quality of Jeju Island.

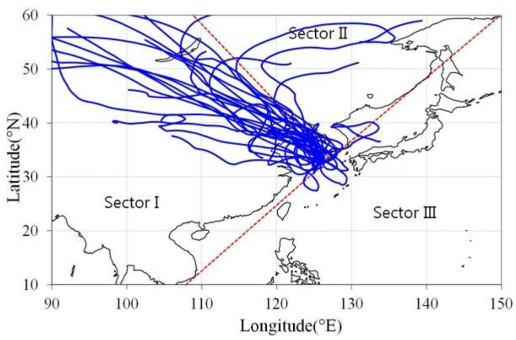

Fig. 2. 5-Day backward trajectories at the Gosan Site of Jeju Island during the study period.

3.4. Inhalation annual effective dose

The inhalation annual effective radiation dose (default mode F) to the public due to natural isotopes of the airborne TSP was in the range 16.195 ~ 77.051 nSv/y, depending on the age group. Because of the variations in air breathing rate, the total annual dose due to the natural radioactivity in airborne TSP increases for the older age groups. For instance, $E_{h,Total}$ was 16.195 nSv/y to the infants (<1 year), while it was 72.178 nSv/y to the adults (male). These values are higher than the corresponding values calculated from the data presented in [3]. It is obvious that ^{232}Th is the main contributor to the inhalation annual effective dose. ^{232}Th was found to be responsible for 98.4 ~ 99.4% of the total dose. On the other hand, ^{40}K was found to slightly contribute to the total dose (0.12 ~ 0.64%).

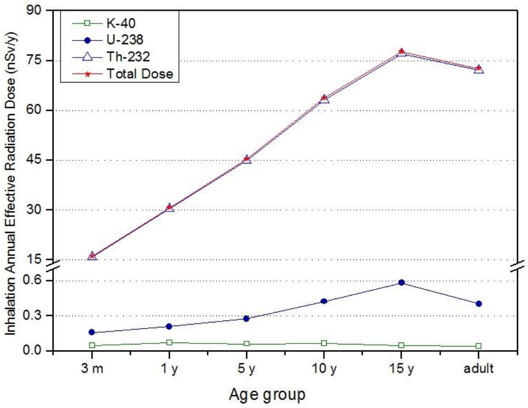

Fig. 3. Annual effective dose to various age groups in Gosan site from inhalation ^{40}K, ^{238}U, and ^{232}Th in TSP aerosols (F type, Male).

4. Conclusions

The atmospheric total suspended particulates (TSP) aerosols were collected at Gosan site of Jeju Island, which is one of the background sites of Korea, during January to April 2013. This study analyzed using ICP-DRC-MS the concentrations of potassium, uranium and thorium, and evaluated the annual effective dose by breathing from the results. During the study period, the mean concentrations of ^{40}K, ^{238}U and ^{232}Th were 54.90±44.31, 21.37±21.57 and 55.78±69.59 pg/m^3, respectively. As a result, the mean radioactive concentrations of ^{40}K, ^{238}U and ^{232}Th were 14.19±11.45, 0.26±0.23 and 0.23± 0.28 μBq/m^3, respectively. During Asian dust periods, the concentrations of ^{40}K, ^{238}U and ^{232}Th were highly as 3.59, 5.51 and 7.86 times, respectively, compared to the non-event periods. The ^{232}Th/^{40}K ratio of Asian Dust was 0.026, which was about 2.17 times higher than the Non-Event value (0.012). The ^{232}Th/^{238}U ratio of Asian Dust was 1.10, which was higher than those of other atmospheric phenomenon. The correlations between the studied natural isotopes is a good positive correlation between ^{232}Th and ^{238}U, supporting the conclusion that they originated from the same source, mostly the crust. The backward trajectory analysis has confirmed that the ^{40}K, ^{238}U and ^{232}Th are delivered as the air masses have moved from the China continent. The inhalation annual effective radiation dose (default mode F) to the public due to natural isotopes of the airborne TSP was in the range 16.195 ~ 77.051 nSv/y, depending on the age group. Jeju Island with less pollution source and low population density is also one of the best places as a background area in Asia. It is judged that the results become a preliminary data on the impact of fine dust from China, which has been recently intensified, on the Korean Peninsula. Furthermore, unlike the previous weather in which the Asian dust blows throughout the peninsula only during the spring season, but now frequently occur even in the autumn. Therefore, it is judged as necessary to observe such Asian dust phenomenon in the long term through collecting a large number of the TSP aerosol filters.

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