Concentration of Uranium and the Activity Ratio of $^{234}\text{U}/^{238}\text{U}$ in Surface Air: Effect of Atmospheric Burn-up of Cosmos-954

by

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Abstract

Results of study on the concentration of uranium and the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in surface air in Tokyo and Tsukuba Science City indicate that the average concentrations of uranium are $24 \pm 15 \text{ pg m}^{-3}$ and $14 \pm 10 \text{ pg m}^{-3}$ respectively, and the average activity ratio is about 1.0. The high ratio up to 1.7 observed in three spring seasons. The causes of these high isotopic ratios of $^{234}\text{U}/^{238}\text{U}$ may be due to the effect of ablated debris deriving from the atmospheric burn-up of Cosmos-954 on 24 Jan. 1978 over northern Canada.

1. Introduction

Only limited number of studies have been done on the concentration of uranium and its isotopic ratio in the air (Hamilton, 1970; McEachern et al., 1971; Krey et al., 1979). It is considered that there are two sources of atmospheric uranium: One is the natural suspended particles of terrestrial origin and the other is the anthropogenic sources, such as fly ashes from fossil fuel burning, nuclear fuel mining and processing, fissile materials from nuclear weapon testings and atmospheric burn-up of nuclear powered satellites.

The projected rapid increase of nuclear generated power plants and associated industry and also of fossil fuel power plants has raised questions regarding the possible increase of existing uranium levels in the environment in near future.

Consequently it is important to know the present level of the concentration and isotopic ratio of uranium in the airborne particles in Japan. It is also important to look, if any,

for the effect of the Russian satellite Cosmos-954 in the surface air, as was observed previously in the case of the atmospheric burn-up of the SNAP-9A (^{238}Pu loaded) satellite in 1964.

In this short report, the authors intend to present the results of study on the concentration and activity ratio of uranium isotopes in the surface air and in some of the rain water collected in Tokyo and Tsukuba Science City, Japan, from 1979 to 1981, with some discussions on the probable effect of the atmospheric burn-up of the reactor loaded satellite.

2. Samples and the method of analysis

Particulate matter in the air near the ground was collected continuously on glass fiber filter (Toyo GB-100R, its nominal efficiency in dealing with particles $0.3 \mu\text{m}$ in size is 99.99%) by using High Volume Air Sampler at a flow rate of 1000 liters per minute. Some of the air sample was collected separately

using a two-stage sampler according to their particle sizes, i.e., those larger than and those less than $10\ \mu\text{m}$ in particle size.

The particulate matter collected on the filter was measured for every month, and subjected to analysis. The amount of airborne particles was determined by gravimetric method. The suction volume of the air was equivalent to 1 to $3 \times 10^4\ \text{m}^3$.

The sample filter was thoroughly digested with conc. HCl and HNO_3 and uranium in the solution was separated by using anion exchange resin (Dowex 1×8 , NO_3 form, $6\ \text{mm}$ dia. and $10\ \text{cm}$ long) in $1\ \text{M}$ solution of HNO_3 in CH_3OH media.

In the analysis of uranium in rain water, the sample water was passed through a membrane filter (Millipore HA, $0.45\ \mu\text{m}$ of pore size), and the filtrate was concentrated on a hot plate. The residue on the filter was digested with a mixed solution of HCl and HNO_3 . Separation of uranium in the solution from the other elements which interfere with the alpha-spectrometry of uranium was carried out by the same procedure as described above.

Uranium isotopes adsorbed on the resin were eluted with $0.1\ \text{M}$ HCl and they were electroplated on a silver disk cathode at $15\ \text{V}$ and $250\ \text{mA}$ for $2\ \text{hr.}$ in $2\ \text{M}$ HCl and ethyl alcohol media ($1:20$).

The determination of uranium and the activity ratio of $^{234}\text{U}/^{238}\text{U}$ was done by alpha-spectrometry. The alpha-ray intensities of ^{234}U ($4.77, 4.72\ \text{MeV}$), ^{238}U ($4.20, 4.15\ \text{MeV}$ and ^{232}U ($5.32, 5.27\ \text{MeV}$) were measured with an alpha-ray spectrometer consisting of four solid state detectors connected with a multichannel pulse height analyzer for 1 to 3×10^4 minutes.

To obtain a yield of chemical separation, an aliquot of ^{232}U tracer was used. For the isotopic ratio standard, uranium extracted from uraninite, Blanka Iron Mine, Sweden was used, the activity ratio of which is obtained as 1.003 ± 0.002 . In converting the activity of ^{238}U into a weight, a half life of ^{238}U of $4.51 \times 10^9\ \text{y}$ and an abundance ratio of 0.9928 were used.

A blank run was done by using the same

batch of filters and the same procedure. The results indicate that the content and isotopic ratio of uranium are less than $1\ \text{ng}$ per sheet of filter and 1.00 ± 0.01 of $^{234}\text{U}/^{238}\text{U}$ activity ratio, respectively.

3. Results and discussion

The results of determination of the concentration of uranium and the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the air near the ground in Tokyo and Tsukuba Science City are shown in Table 1 and Fig. 1.

3.1 Concentration of uranium in the air: As shown in Table 1 and Fig. 1, the concentration of uranium in the surface air in Tokyo ranged from 11 to $68\ \text{pg m}^{-3}$ with an average value of $24.1 \pm 15.0\ \text{pg m}^{-3}$.

The highest value, which was as high as $68\ \text{pg m}^{-3}$, was observed in April and the lowest, which was $11\ \text{pg m}^{-3}$ was found in August of the same year.

As seen in Table 1, the amount of total suspended particles in the air in Tokyo ranged from 93 to $259\ \mu\text{g m}^{-3}$, with an average value of $169 \pm 50\ \mu\text{g m}^{-3}$. The content of uranium per unit weight of the atmospheric suspended particles ranged from 0.08 to $0.26\ \mu\text{g g}^{-1}$ with a mean value of $0.14 \pm 0.06\ \mu\text{g g}^{-1}$.

In this connection, Hamilton (1970) gave

Fig. 1. Concentration of uranium and the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in surface air.

Table 1. Concentration of uranium and the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in surface air.

Month	Total U ($\mu\text{g m}^{-3}$)	$^{234}\text{U}/^{238}\text{U}$ activity ratio	Total suspended matters ($\mu\text{g m}^{-3}$)
1979, Jan.	22.4 ± 0.7	1.10 ± 0.02	200
Feb.	22.9 ± 0.7	1.19 ± 0.02	218
Mar.	16.8 ± 0.5	1.68 ± 0.01	212
Apr.	68 ± 2.0	1.21 ± 0.01	259
May	15 ± 0.5	1.18 ± 0.02	159
June	25 ± 0.7	0.91 ± 0.02	216
July	16 ± 0.5	1.05 ± 0.01	138
Aug.	10.7 ± 0.3	0.90 ± 0.02	100
Sept.	34.5 ± 1.0	0.93 ± 0.01	160
Oct.	24.3 ± 0.8	0.93 ± 0.01	125
Nov.	14.4 ± 0.5	1.00 ± 0.01	145
Dec.	14.5 ± 0.5	1.11 ± 0.01	93
1980, Jan.	29.0 ± 0.8	0.95 ± 0.01	130
Feb.			
Mar.	—	—	—
Apr.	6.9 ± 0.3	1.46 ± 0.02	—
May			
June	6.8 ± 0.3	1.10 ± 0.03	50
July	3.7 ± 0.3	0.90 ± 0.02	30
Aug.	14.3 ± 0.5	0.90 ± 0.01	60
Sept.	9.2 ± 0.3	1.04 ± 0.02	40
Oct.	14.0 ± 0.5	0.99 ± 0.01	50
Nov.	15.8 ± 0.5	1.21 ± 0.01	75
Dec.	4.9 ± 0.3	1.03 ± 0.02	30
1981, Jan.	36 ± 1.0	1.09 ± 0.02	150
Feb.	35 ± 1.0	1.05 ± 0.01	150
Mar.	7.5 ± 0.3	1.51 ± 0.02	40
Apr.	13.7 ± 0.4	1.21 ± 0.02	66
May	14.4 ± 0.5	0.98 ± 0.01	69
June	7.2 ± 0.3	1.15 ± 0.02	50

#: Over the western North Pacific (30°N , 147°E).

the same order of magnitude of uranium in the airborne dust as $0.4 \mu\text{g g}^{-1}$ in the air in England.

From June, 1980, the sampling of atmospheric suspended matter has been continued at Tsukuba Science City, which is located about 60 km north of Tokyo.

The concentration of uranium in the air during the period from June 1980 to June 1981 ranged from 5 to $36 \mu\text{g m}^{-3}$ with an average value of $14.0 \pm 10.3 \mu\text{g m}^{-3}$. During the period of study, the high values of up to 35 to $36 \mu\text{g m}^{-3}$ were observed in January to February 1981, whereas the lowest one was observed in July, 1980.

The concentration of total uranium in the

air at Tsukuba is about half of that in Tokyo, but its content per unit weight of airborne dust shows a little higher than that in Tokyo on an average ($0.20 \pm 0.05 \mu\text{g g}^{-1}$). In general, there are two kinds of linear relationship between the concentration of uranium and the amount of atmospheric suspended particles as shown in Fig. 2.

One of the reasons of these differences may be due to the differences of the content of volatile matter in the atmospheric dust between the two cities (Dokiya et al., unpublished).

The concentration of uranium in surface air in Tokyo and Tsukuba is of the same order of magnitude as given in Sutton, Eng-

Fig. 2. The relationship between concentration of uranium and amount of total suspended matter in surface air.

○: Tokyo, △: Tsukuba

land (Hamilton, 1970), but much lower than those in New York State, USA (McEachern et al., 1971), which is located downwind of a nuclear fuel fabrication plant.

The content of uranium in airborne particles larger than $10\ \mu\text{m}$ ranged from 13 to 35% of the total uranium in Tokyo and 15 to 52% in Tsukuba. It is noticed that the average concentration of uranium in large particles in both cities showed the same value, $4.2\ \text{pg}$, with about 1.0 of the activity ratio of $^{234}\text{U}/^{238}\text{U}$.

Therefore it may be said that the large particles in surface air are mainly derived from the resuspended local soil particles or debris.

The results of examination of the water soluble part of airborne dust indicate that most of the uranium in airborne particles exists in insoluble form in distilled water.

A sample of airborne dust over the open ocean (30°N , 147°E) which is collected from the end of April to early May, 1980, indicates that the concentration of uranium in the air over the ocean is $6.9\ \text{pg m}^{-3}$. This value is in good agreement with those over the Atlantic Ocean (Hamilton, 1970).

3.2 Uranium in rain [water]: From 1980 to 1981, several determinations of the uranium content and its activity ratio of $^{234}\text{U}/^{238}\text{U}$ in rain water were carried out concerning both filtrate and particulate matter.

As seen in Table 2, the content of uranium ranged from 1 to $15\ \text{ng l}^{-1}$ and it is found that most of the uranium in rain water is contained in particulate matter.

Some of the results dealing with the monthly depositions of uranium in Tokyo are shown in the same table, where it is seen that the monthly depositions of uranium ranged from 0.3 to $4.7\ \mu\text{g m}^{-2}$ from March to October, 1980, with a mean concentration of

Table 2. Concentration of uranium and the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in rain water.

Date	Total U (ng l ⁻¹)	²³⁴ U/ ²³⁸ U activity ratio	
Aug. 22, 1980	2.3±0.2	1.02±0.02	
May 12, 1981	11.3±0.3	1.03±0.02	
May 17, 1981	14.5±0.5	1.22±0.02	
June 15, 1981	1.4±0.1	1.01±0.03	
Monthly deposition of uranium in Tokyo			
Month	Total U (μg m ⁻²)	²³⁴ U/ ²³⁸ U activity ratio	Amount of ppt (mm)
Mar, 1980	4.65±0.14	1.01±0.01	173.5
June 1980	0.34±0.05	1.35±0.02	172.5
July 1980	0.59±0.04	1.16±0.02	202.5
Aug. 1980	0.54±0.03	1.03±0.01	168.0
Sept. 1980	1.04±0.05	0.87±0.02	181.5
Oct. 1980	1.93±0.07	1.08±0.01	136.0

about 10 ng l^{-1} .

The activity ratio of $^{234}\text{U}/^{238}\text{U}$ ranged from 0.9 to 1.35, most of which is shown within the range of natural variations (Hyde, 1964), but a little higher value, 1.35, observed in June, 1980 may reflect the high activity ratio of the atmospheric uranium.

3.3 Uranium isotopic ratio in the air: As shown in Table 1 and Fig. 1, the activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the air ranged from 0.9 to 1.7 with an average value of 1.07 ± 0.21 for Tokyo and 1.11 ± 0.19 for Tsukuba.

The maximum ratios as high as 1.78 and 1.51 are observed in March 1979 and 1981. Another high ratio of 1.46 is also observed in April to May 1980 over the Pacific Ocean.

It is to be noted that the higher ratios of uranium isotopes in the air were observed only in the spring season. This is quite similar phenomenon to the "spring peak" of radioactive fallout which is derived from the introduction of the stratospheric air into the troposphere through the tropopause of middle latitudes.

The causes of such a high ratio of $^{234}\text{U}/^{238}\text{U}$ in the air must be taken into account by the addition of enriched uranium other than of natural origin. (The range of natural variation of the activity ratio of uranium isotopes 0.9 to 1.2). As pointed out by McEachern et al. (1971), nuclear fuel fabrication or reprocessing plants may be one of them in the limited area around the plants.

However, considering the distance from the plant and the wind direction prevalent over the sampling sites, especially in ocean areas, it is difficult to explain the higher isotopic ratio of uranium by the effect of these sources.

Another possible source may be the ablated radioactive debris from the atmospheric burn-up of the reactor loaded satellite, such as Cosmos-954.

It is well known that in the case of atmospheric burn-up of the SNAP-9A loaded satellite over the Southern Hemisphere on 21 April, 1964, the first fallout debris of ^{238}Pu was observed at the surface about one year

after the accident in Australia and Italy (Hardy et al., 1972). The effect of high ^{238}Pu fallout continued for several years on a global scale (Telegadas, 1968, 1969; Krey, et al., 1970; Hardy et al., 1972; Miyake et al., 1970, 1975).

High atomic ratios of uranium isotopes (0.15% for ^{234}U and 1.4% for ^{235}U) were recently reported in the stratospheric air collected after the atmospheric burn-up of the Russian satellite Cosmos-954 over northern Canada on 24 Jan. 1978 (Krey et al., 1979). The reactor on board is assumed to have been a thermal power plant of 40 kW containing as much as 50 kg of 90% enriched ^{235}U .

According to their study, the isotopic ratio of the stratospheric air was two to three times higher than that of natural uranium. The estimated amount of Cosmos ^{235}U in the stratospheric air is 3 to 4 pg m^{-3} STP assuming that 25% of ^{235}U was released into the stratosphere of high latitudes in the Northern Hemisphere. The concentration of total uranium in the stratospheric air is about 400 pg m^{-3} STP, which contains about 3 pg m^{-3} of ^{235}U except for the Cosmos-954 contamination. Addition of the Cosmos-954 uranium may increase the concentration of ^{235}U in the stratosphere up to 100% (from 0.7% to 1.4%).

On the other hand, in surface air, a modal concentration of 10 pg m^{-3} of natural uranium contains 0.07 pg m^{-3} of ^{235}U under ordinary conditions. It is well known that during the enrichment process of ^{235}U , ^{234}U is much more enriched than ^{235}U . In the 90% enriched ^{235}U (125 times more enriched than natural), ^{234}U is contained about 1% which is 182 times higher than natural abundance. The enrichment ratio of $^{235}\text{U}/^{234}\text{U}$ is 0.8.

A 70% increase of the observed activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the air may correspond to about a 50% increment (about 0.03 pg m^{-3}) of the amount of ^{235}U in the air.

Therefore, only a few percent of the stratospheric air introduced into the troposphere in the spring season may be enough to explain the observed increment of the isotopic ratio of uranium in the surface air.

It is concluded from the above that ac-

cording to our knowledge on the behavior of the stratospheric fallout derived from nuclear explosions, and also from our experiences on the fallout of the SNAP-9A satellite failure, there is every reason to believe that the anomalous high activity ratio and abrupt increase of $^{234}\text{U}/^{238}\text{U}$ ratio in the surface air observed in the spring season in Japan and over the open ocean were occasioned by the atmospheric burn-up of the Russian satellite Cosmos-954. The effect is expected to continue for several years more.

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地表大気中のウラン含量および $^{234}\text{U}/^{238}\text{U}$ 放射能比

広瀬勝己・杉村行勇

東京および筑波における地表大気中のウラン含量は、東京で平均 $24 \pm 15 \text{ pg m}^{-3}$ 、筑波では、 $14 \pm 10 \text{ pg m}^{-3}$ であり。エアロゾル 1 g 当り $0.1 \sim 0.2 \text{ } \mu\text{g}$ であった。ウラン同位体 $^{234}\text{U}/^{238}\text{U}$ の放射能比は $0.9 \sim 1.7$ を示し、天然ウランの同位体比変動巾を大きくこえるものが、1979年および1981年3月、1980年4～5月（海上）にみられた。この高い同位体比の原因としては、1978年1月24日に北部カナダに落下したソ連原子炉積載衛星コスモス 954 からの、濃縮ウラン放出によるものと推定された。