Radioactivity Originating from the Chinese Nuclear Test Explosions Observed in Seoul District in 1964-1967

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中共 核實驗에 의한 서울地區의 放射能 汚染度의 評價

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摘要

1963年에서 1967年에 걸쳐 서울地區의 人工放射能과 自然放射能을 全放射能의 測定과 放射性核種의 分析을 통하여 研究하였다.

核分裂生成物의 濃度가 적을때는 짧은 半減期를 갖는 라듐이나 토리움의 崩壞生成物이 浮游塵의 大部分을 차지하고 있었으며 核分裂生成物에 依한 放射能은 試料 採取후 며칠 지나야 正確히 評價할수 있었다.

7次에 걸친 中共의 核實驗의 結果 두차례의 强放射能이 爆發후 30時間을 前後하여 서울地區에 나타났으며 이들은 1956年에서 1962年사이에 美國과 蘇聯에서 행한 實驗과 比較할때 높은 比放射能을 보였으나 持續時間은 아주 짧아서 1週內에 急激히 減少하였다.

이로 보아서 서울地區의 放射能汚染은 中共의 核實驗인 境遇 核實驗의 規模와 實驗高度 및 爆發前後의 氣象條件, 特히 高層對流圈의 제트氣流에 依해서 많은 影響을 받음을 알았다.

INTRODUCTION

Since 1945 there has been a steady increase in the number of nuclear explosions tested. From 1945 through 1951 the United States and the USSR performed nuclear detonations with a total yield of about 0.8 megatons, whereas in 1957 and 1958 alone the total fission yield from the same source was 40 megatons (Anonymous, 1959).

In 1964 the Chinese exploded their first nuclear device at Lop Nor, Sinkiang Province and since then they have contributed to the distribution of radioactive fallout for the nearby countries including Korea. The relationship between the amount of fallout produced depends on the height above the earth at which the nuclear explosion is tested and the type of nuclear device.

The first counting equipment for radioactivity introduced in Korea was set at the Scientific Research Institute, Ministry of National Defence (revised to the Army Research & Testing Laboratory) in 1956. Since the installation of the equipment the survey of radioactive environmental contamination has been performed for the artificial radioactivity such as in airborne, rain-out and fallout dusts (Park, 1957, 1959; Park *et al.*, 1960; Kim *et al.*, 1962). Because of this systematic and continuous assay, it was possible to evaluate and compare the radioactivities originating from various nuclear detonations with regard to the radioactive environmental contamination in Seoul district.

The purpose of this study is to make a comparison of radioactivity originating from the huge tests, which were performed by the US and the USSR from 1956 to 1963, with the small ones by the Chinese from 1964 to 1967, and to predict the levels of radioactivity to be originated from various sources of nuclear tests.

MATERIALS AND METHODS

Airborne Dust Airborne dust was collected using a Staplex air sampler at a flow rate of 200 *l*/min about 1 meter above the ground. The filter paper used was Whatman No. 4. Collection usually was performed for 4 hours from 10 a.m. to 2 p.m. throughout 1963.

The collected dust was ashed by gradual heating to about 250°C for 2 hours and at 600°C for 3 hours in an electric muffle furnace and was conveyed into the stainless steel planchet followed by the addition of a few drops of anhydrous ethanol and heating under an infrared lamp with continuous rotation of mounted preparation. The gross-activity was measured in a GM-tube (window thickness, 1.4 mg/cm²) connected to 192A Ultrascaler. γ-Spectra were obtained with RLP-5 continuous scanning spectrometer. The calibration sources used for a threshold-energy curve were ^{226}Ra (.118 Mev), ^{137}Cs (.662 Mev), ^{54}Mn (.835 Mev), ^{214}Pb (.325 Mev) and ^{214}Bi (.609 Mev).

Fallout Dust Sampling was made using gummed paper (1 feet²) on three and four day basis on the roof about 10 meters above the ground from 1964 through 1967. Procedures similar to those just described for airborne dust have been employed for the preparation and counting of radioactive samples.

Rain-out Dust Rain-out dust was collected at every rainfall using a rain collecting device and was subjected to evaporation followed by the procedures for mounting and counting of radioactive samples similar to those used for airborne dust.

RESULTS

Radioactivity of airborne dust measured 6 hours after collection is shown in Fig. 1 along with rainfall for the comparison. The lowest activity observed was 0.68 pCi/m³ and the highest 15.95 pCi/m³. Radioactivity of airborne dust exhibited an average of about 6.5 pCi/m³ throughout the year except the rainy season of April through August, in which extremely low levels of activity appeared in airborne dust samples. Decay patterns for the airborne dust indicated that the samples consisted of mixed radionuclides of short-lived and long-lived. The half-life of short-lived nuclides was determined to be about 10 hours and that of long-lived nuclides about 1 month from the decay patterns as shown in Fig. 2.

In Fig. 3 are shown the γ-spectra obtained with a sample of high activity. From these spectra ^{214}Pb and ^{214}Bi of radium decay products and ^{212}Pb and ^{208}Tl of thorium decay products were identified. Among the low energy γ-emitters, ^{214}Pb exhibited the highest activity. Radionuclides of fallout origin were not detected in the present identification.

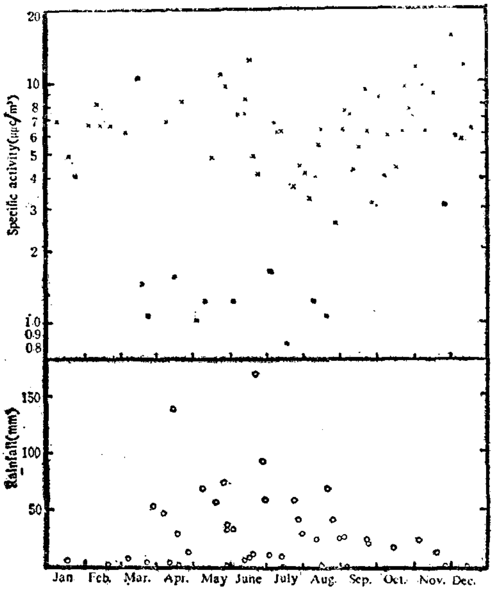

Fig. 1. Radioactivity of airborne dust samples and the amount of rainfall in 1963.

Radioactivity of rain-out and fallout dusts from 1964 through 1967 are shown in Fig. 4. The normal levels of activity were in a range of 10-100 pCi/*l* for rain-out and 100-1,000 pCi/m²-d for fallout dusts. Exceptionally high peaks in 1964 and 1966 were believed to be caused by the first and fifth Chinese nuclear tests, respectively. The increase in the magnitude of activity was of the order of 3 and 5 in comparison with that of normal levels. It was followed by a sudden decrease in activity in less than a week. The second, third, fourth, sixth and seventh Chinese nuclear tests affected not much influence to the levels of environmental radioactivity in Seoul district. The small peaks appeared were believed to be due to the fallout originating from corresponding tests.

In Table 1 is shown the comparative data on the activity of fission products from the Chinese nuclear test explosions and that of other nations so far recorded as revealing high level in the period of 1956-1962. In the period mentioned above, the highest radioactivity observed was 10,300 pCi/l and 27,200 pCi/m^2-d for rain-out and fallout dusts respectively, which was ascribed to be due to a series of tests performed at Eniwetok by the United States in 1958. On the other hand, the levels of radioactivity of fission products originating from the fifth Chinese test were 53,000 pCi/l for rain-out and 15,300,000 pCi/m^2-d for fallout dusts.

Decay patterns for fission products originating from the first Chinese nuclear explosion were obtained to ascertain if these follow the Way and Wigner's β-estimate and are shown in Fig. 5.

Also analyzed are the γ-spectra for the dust samples collected from the fallout originating from the first and fifth Chinese tests and are shown in Figs. 6 and 7. Analyzed nuclides of fallout origin are shown in Table 2.

DISCUSSION

The γ-ray spectra of airborne dusts collected in 1963 have revealed the existence of decay products of radium and thorium, in which the major activity was found to be in ^{214}Pb. This finding seems to be quite reasonable to refer the published data. Damon *et al.* (1954) estimated the radioactivity of Ra B and Ra C to be 10^{-9}-10^{-7}

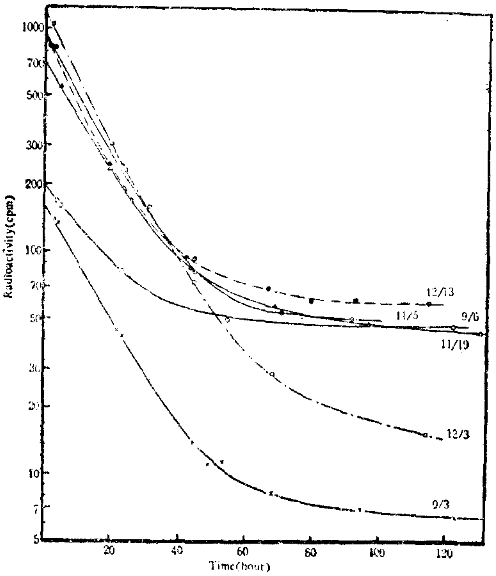

Fig. 2. Decay patterns of some airborne dust samples, which were collected and sampled on the day indicated in 1963.

Table 1. Comparison of fallout particles originating from the Chinese nuclear test explosions with those of other nations observed in Seoul district

Date of explosion	Test site	Nation	Radioactivity		Reference
			Rain-out dust(pCi/l)	Fallout dust(pCi/m^2-d)	
Jul. 19, 1956	Monte Bello	England	2,700(6/22)*	——	
Apr. 16, 1957	Siberia	USSR	3,500(4/22)	1,000(5/1)	C.D. Park
Apr. 8-	Eniwetok	USA	10,300(7/8)	27,200(7/12)	
Jul. 27, 1958	(34 tests)		3,500(7/25)		
Sep. 1-	North pole	USSR	3,900(9/25)	4,500(9/30)	
Oct. 31, 1961	(27 tests)		3,800(12/10)	5,200(11/30)	T.S. Kim
Aug. 5-	North pole	USSR	6,300(9/1)	11,400(8/30)	
Sep. 27, 1962	(12 tests)		9,400(11/10)	9,200(11/11)	
Oct. 16, 1964	Lop Nor	China	56,000(10/20)	345,000(10/21)	
May 14, 1965	Lop Nor	China	800(5/20)	3,600(5/16)	
May 9, 1966	Lop Nor	China	200(5/10)	1,000(5/30)	M.S. Kang
Oct. 27, 1966	Lop Nor	China	——	4,100(11/5)	
Dec. 28, 1966	Lop Nor	China	53,000(12/31)	15,300,000(12/30)	
Jun. 17, 1967	Lop Nor	China	100(6/19)	800(6/19)	

* Date of measurement.

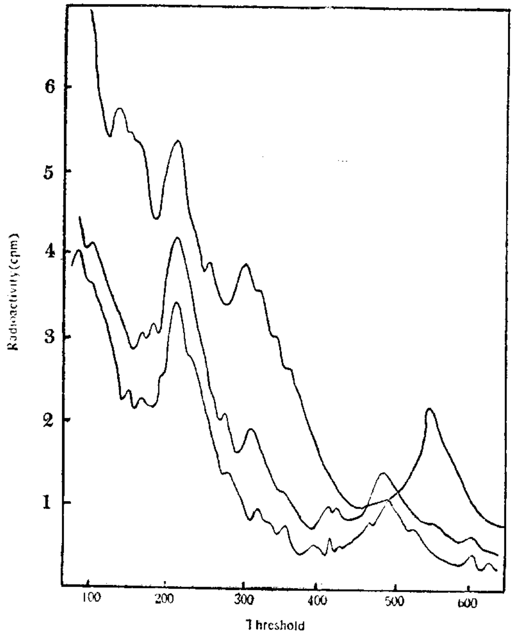

Fig. 3. A typical spectral transformation of an airborne dust sample with change in time.

Ci/*l* and that of Th B 10^{-13}-10^{-10} Ci/*l*. Also reported by Reita (1956) is that the decay products of radium and thorium were detected in the high level air of Alps. In the study of atmospheric radioactivity at Washington, D.C. in 1950 through 1961, Lockhart, Jr. (1961) observed variations in the short-lived radon and thoron decay products in the air. From the data he obtained, he concluded that the short-term variations of these products are sufficiently great to make impractical the direct determination of gross fission products in the associated air masses without removal of the short-lived natural products through decay, unless the fission product concentration is extremely high. In view of this fact and the findings I have made, it is likely to be said activity remaining after a few days is attributable to fission products.

From the present identification, in which samples were collected in 1963, no artificial radionuclides originating from the fallout were observed. The reason for this is thought to be due to the fact that ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Rh, ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce and ^{144}Pr except ^{106}Ru, originating from a series of nuclear tests held by the USSR (Tisijar-Lentulis, 1963 and Sakagishi *et al.*, 1963) have relatively so short half lives that the level of radiation intensity at the time of collection was unable to detect, whereas radioactivity of decay products of radium predominated.

General patterns of radioactivity of rain-out and fallout dusts in 1964 through 1967 exhibited several peaks owing to the increase and decrease in the activity following nuclear detonations performed by the Chinese in

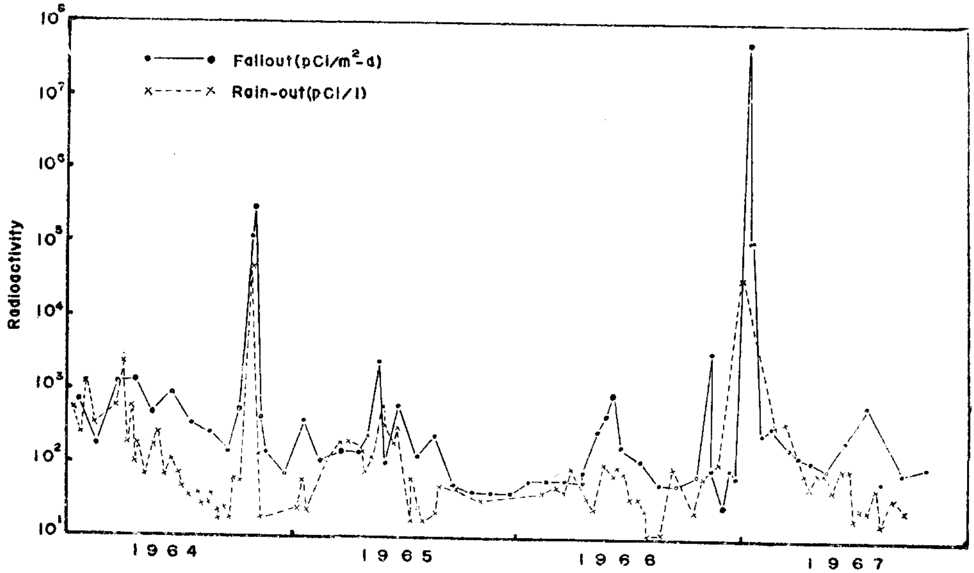

Fig. 4. Radioactivity originating from the Chinese nuclear test explosions observed in Seoul district in the period of 1964-1967.

the interim period of time.

Results of observation on radioactivities both of rain-out and fallout dusts during 1964 indicated that concentrations of radioactivity decreased from the beginning of the year towards autumn exhibiting the spring maximum, but it increased abruptly in October under the influence of the first Chinese nuclear explosion. The first Chinese nuclear test explosion was carried out on 16 October 1964 at Lop Nor, Sinkiang Province, where six successive nuclear explosions were performed thereafter. The abrupt rise in the levels of activity of fallout was observed on 18 October and a much higher level of activity was measured on 21 October. The activity observed on 18 and 21 October was 139,000 pCi/m^2-d and 345,000 pCi/m^2-d. We had a rainfall on 20 October, which precipitated the fission debris, and the activity measured was 56,000 pCi/*l*. It was concluded from the measurements of a continual search for radioactivity that the fresh fission debris reached Seoul district about 30 hours after the nuclear explosion and the influence lasted for about 4 days. It seems to be plausible to refer that a rough estimate of the flow rate of dust particles was found to be 1,000–2,000 km/day on an average. Reported by Mamuro *et al.* (1964) was that many highly radioactive fallout particles were found and collected on 19 and 20 October in Osaka, Japan. Their publication showed that they had rainfalls on 17, 20 and 21 and no fresh nuclear debris was found in the 17th-rain water, but the 20th-rain water and the 21st-rain water had activity concentrations of 16,600 pCi/*l* and 3,300 pCi/*l*, respectively, indicating clearly the presence of fresh nuclear debris. A comparatively large amount of fallout formation was predicted by learning the test was of land surface burst and the test site was close to Seoul district.

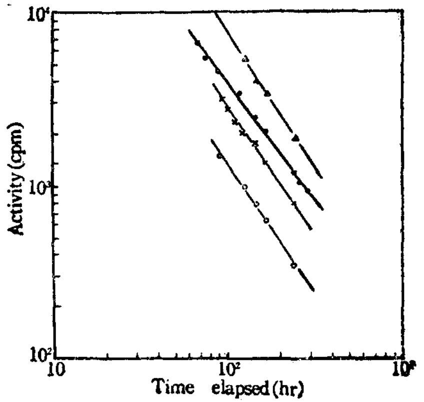

Fig.5. Decay patterns of fission products originated from the first Chinese nuclear explosion.
—●— fallout dust(10/19) —△— fallout dust(10/21)
—○— fallout dust(10/20) —×— rainwater(10/20)

Results of observation during 1965 on rain-out and fallout dusts indicated no fresh nuclear debris reached Seoul district except somewhat high values were observed in January and May. The rise in the levels of activity was attributable to a venting underground nuclear explosion which was carried out at Semiparatinck on 15 January by the USSR and to the second Chinese nuclear test explosion carried out on 14 May. The activity of fallout concentration following the tests was about 10 times those obtained for the samples collected prior to the tests. The rise in the level of activity was believed to be due to the second Chinese nuclear test explosion although not much change in the activity was observed. Referring to the results obtained by Mamuro *et al.* (1965), the concentration of fallout activity in Osaka in 1965 usually was lower than 5 pCi/m^3 except in January and May and increased to a high value of 105 pCi/m^3 on 20 January and 48 pCi/m^3 on 21 May resulting from the venting underground explosion by the USSR and the second Chinese nuclear explosion, respectively. They also detected the fresh nuclear debris under the influence of the second Chinese nuclear explosion. The low activity observed in Seoul district might be due to the weather system and especially the jet stream in middle latitude in the upper troposphere immediately after the explosion.

In 1966 three nuclear test explosions of the Chinese origin were performed; the third and fourth ones were said to be air bursts and fifth one land surface burst. As is evident from Fig. 4 not much rise of activity was observed for both third and fourth nuclear explosions. The outcome of this measurement might be ascribed to the similar reason as for the second Chinese test. Presented by Mamuro *et al.* (1966) and Fujita *et al.* (1967) was that shortly after the third Chinese nuclear test explosion of May 9, 1966, many hot particles were collected in Osaka district. In connection with this nuclear test, Baugh *et al.* (1967) have reported that the fallout particles reached Arkansas about 1 week after the nuclear explosion. It was followed by a sudden decrease of particles about 25 May.

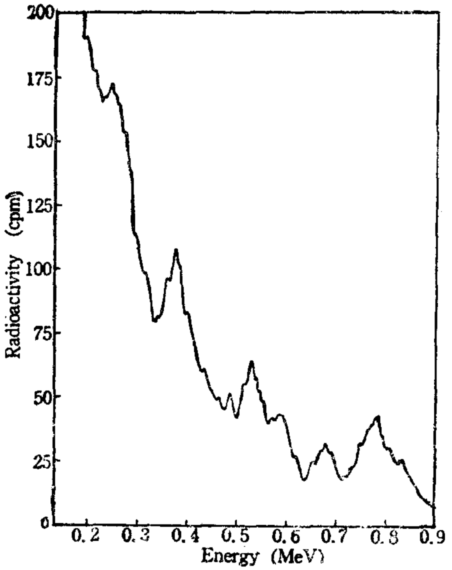

Fig.6. A γ-ray spectrum of a highly radioactive fallout dust sample originating from the first Chinese nuclear explosion.

After a small peak was observed on 30 May, the second big wave reached early in June. The low activity observed in Seoul district might be owing to the fact that there were some differences between the weather conditions of high latitude of Japan and the United States and those of Korea. The rise in the level of activity was detectable enough to reasoning the fallout, although measured activity before and after the tests exhibited not much difference.

It is generally accepted that debris produced in an atom bomb (kiloton) explosion is swept up only into the lower atmosphere (troposphere) where it is subject to a different form and rate of dispersion than is debris from a hydrogen bomb (megaton) explosion which is propelled to greater heights and initially disperses in the stratosphere. The larger particles of debris from both types of explosion return to the earth's surface within a few hours and within a few hundred miles of the explosion. The particles are usually fragments of the substratum, sand, coral, earth, on which the bomb was exploded and contain a high proportion of shorter-lived radionuclides which give rise to the great hazards of close-in fallout. This material tends to be deposited in areas downwind from the explosion. Finer particles from a kiloton explosion tend to be dispersed in a band, east and westwards, round the earth by the weather system and especially by the jet stream in middle latitude in the upper troposphere (Mauchline *et al.*, 1964).

The fifth Chinese nuclear explosion of December 28, 1966 is believed to have been a land surface burst whose yield was some hundreds of kiloton. On 30 December about 30 hours after the detonation the first visit of fresh nuclear debris was observed in Seoul district. The activity of fallout dusts collected on 30 December was 15,300,000 pCi/m^2-d and that of rain-out measured on 31 December was 53,000 pCi/*l*. The activity of fallout dusts was found to be at least 50 times that of the first Chinese nuclear explosion and that of rain-out was of the same order. A great difference in rain-out and fallout dusts activities might be caused by the weather conditions on the days collection was made.

The γ-ray spectrometry of these samples revealed the existence of ^{133}Xe-^{133m}Xe, ^{143}Ce, ^{103}Ru-^{103m}Rh, ^{132}Te-^{132}I, ^{95}Zr-^{95}Nb, ^{141}Ce, ^{131}I-^{131m}Xe, ^{115}Cd-^{115m}In and ^{99}Mo-^{99}Tc as are shown in Fig. 7. This finding is in good agreement with those obtained by Mamuro *et al.* (1967) and by Kuroda *et al.* (1965), indicating the successful operation of nuclear test.

It is generally believed that the sixth Chinese nuclear explosion of June 17, 1967, which was reported to be the first Chinese thermonuclear explosion, was carried out at a very high altitude and the influence on environ-

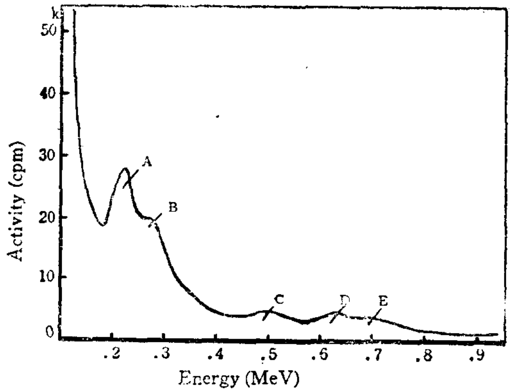

Fig.7. A γ-ray spectrum of a highly radioactive fallout dust sample originating from the fifth Chinese nuclear explosion (Scanned 2 days after the explosion). For the radionuclide analysis each photopeaks were scanned with change in time and their half-lives were also determined.

Table 2. Radionuclides identified using a continuous scanning spectrometer with their half-life and γ-energy.

Nuclide	Half-life	Gamma energy (Mev)	
^{95}Zr–^{95m}Nb	65 d, 35 d	0.03, 0.23	
^{131}I	8.05 d	0.364	
^{140}Ba–^{140}La	12.8 d, 40.2 h	0.54, 0.49	1st test
^{144}Ce–^{144}Pr	285 d, 17.3 m	0.69, 0.69	
^{95}Zr–^{95}Nb	65 d, 35 d,	0.75, 0.76	
^{133}Xe–^{133m}Xe	5.3 d, 2.3 d	0.08, 0.23	
^{143}Ce	33 h	0.29	
^{103}Ru–^{103m}Rh	40 d, 24 m	0.50, 0.04	
^{132}Te–^{132}I	77 h, 2.3 h	0.67, 0.67	5 th test
^{95}Zr–^{95}Nb	65 d, 35 d	0.75, 0.76	
^{141}Ce	32 d	0.15	
^{131}I–^{131m}Xe	8.1 d, 12 d	0.36, 0.08	
^{115}Cd–^{115m}In	54 h, 4.5 h	0.58, 0.33	
^{99}Mo–^{99}Tc	67 h, 2×10^{5}y	0.74, 0.29	

mental contamination of this explosion was unexpectedly little and the seventh Chinese nuclear explosion of 1967 was carried out in failure. No activity originating from the fallout of these two nuclear explosions was detected. In connection with this, Mamuro *et al.* (1967) reported that they could not detect neither any hot particles nor any marked increase in the activity concentration in the atmosphere shortly after the explosion. The fact that I could not detect any activity due to these tests might be ascribed to the fact that the tests were carried out at a very high altitude so that debris produced was propelled to greater height and dispersed in the stratosphere, constituting a world-wide fallout which falls little by little for a long time.

In conclusion, the radioactive environmental contamination in Seoul district largely depends on the height above the earth at which the nuclear explosion is performed and the type of nuclear device as well as the weather system at the time and immediately after the explosion, especially the jet stream in middle latitude in the upper troposphere.

SUMMARY

Artificial and natural radioactivity in airborne, rain-out and fallout dusts in Seoul district in the period of 1963–1967 were studied by measuring gross-activity and by analyzing nuclides by means of γ-spectrometry.

Short-lived radium and thorium decay products give rise to most of the airborne activity unless the fission product concentration is extremely high and it is likely to be said activity remaining after a few days is attributable to fission products.

Of seven Chinese nuclear explosions performed at Lop Nor, Sinkiang Province, two exhibited the activity of extremely high concentration of fission product and reached Seoul district around 30 hours after the explosion. The activity was followed by a sudden decrease in less than a week, in contrast to the long-lasted activity of low concentration originating from the huge tests performed by the United States and the USSR in 1956–1962.

The radioactive environmental contamination in Seoul district, due to the Chinese nuclear test explosions, largely depends on the height above the earth at which the nuclear explosion is performed and the type of nuclear device as well as the weather system at the time and immediately after the explosion, especially the jet stream in middle latitude in the upper troposphere.

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