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Distribution and characteristics of radioactivity(^{232}Th, ^{226}Ra, ^{40}K, ^{137}Cs and ^{90}Sr) and radiation in Korea

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Abstracts - The concentrations of natural and artificial radionuclides in soil and gamma ray dose rate in air at 233 locations in Korea have been determined. The national mean concentrations of ^{232}Th, ^{226}Ra , ^{40}K , ^{137}Cs and ^{90}Sr in soil were 60±31, 33±14, 673±238, 35±9.3 and 5.0±3.4 Bq kg^{-1}, respectively. The mean gamma-ray dose rate at 1 m above the ground was 79 18 nGy h^{-1}. ^{137}Cs concentration had highly significant correlation with organic matter content and cation exchange capacity. ^{90}Sr concentration had slightly coherent with pH. The results have been compared with other global radioactivity and radiation measurements.

Key words : natural radionuclide; artificial radionuclide; soil; gamma ray dose rate

INTRODUCTION

The studies on radioactivity and radiation levels are very important to public health and the environment for several reasons. Measurements of natural and artificial radionuclides in soils can be used as a baseline to assess any change in background levels of radioactivity from various geological processes as well as any potential accident from nuclear facilities. These baseline levels are necessary in order to protect the public and the environment from potential radioactivity hazards. In addition, the accumulation of data on radioactivity and radiation levels is of great value in understanding the nature of the environmental radioactivity in a particular region and in establishing rules and regulations relating to radiation protections [1,2].

Nationwide surveillance on background levels of radioactivity and radiation has been carried out many countries. In Korea, a great deal of work has already been done to survey different aspects of radioactivity and radiation. Several papers discuss the distributions of various radioactive elements including ^{137}Cs and ^{40}K in soils [3] and ^{90}Sr, ^{137}Cs and Pu isotopes in soil samples of densely populated cities [4] and in coastal sediment [5].

The purpose of the present study is to establish baseline values of ^{232}Th, ^{226}Ra, ^{40}K, ^{137}Cs and ^{90}Sr concentrations in Korean soils and the gamma-ray dose rate in the areas where samples were taken. To understand relationship of the characteristics of soil and radionuclides, we analyzed pH, organic matter content, cation exchange capacity in soils.

MATERIALS AND METHODS

Fig. 1 shows 233 sampling locations and the geographical distribution of the nine administrative provinces in Korea.

Each soil sample was obtained from each grid covered in an area of 20 km x 30 km. Samples were collected from 233 locations believed not to be eroded or disturbed for at least 40 to 50 years. Each core (5 cm in diameter x 5 cm in height) was taken with a depth of 5 cm from a randomly chosen point within each site area. The

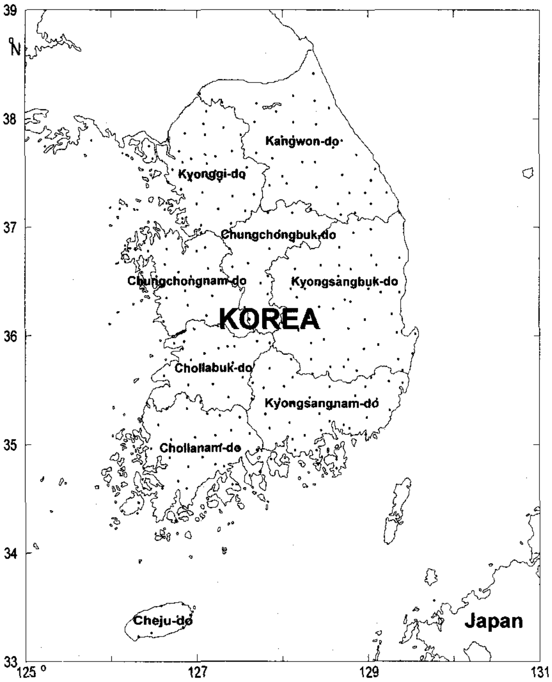

Fig. 1. Map of Korea depicting the geographical distribution of the nine administrative provinces and 233 sampling locations.

samples of 16 cores collected at each site were thoroughly mixed together in the laboratory. Each sample was dried in an oven at 50 ℃ for seven days and sieved through a 2 mm mesh to remove stones and pebbles. The prepared samples were placed in sealed plastic containers and left for at least three weeks before measurement by gamma spectrometry. The concentrations of ^{232}Th, ^{226}Ra, ^{40}K and ^{137}Cs in the samples were determined by employing a high-purity germanium (HPGe) detector with a relative efficiency of 25.0%. The measurements were performed for a counting time of 80000 sec. ^{90}Sr was measured in a 100 g soil sample by first separating out Ca by precipitation of $Sr(NO_3)_2$ with fuming HNO_3 [6]. ^{90}Sr concentration was determined using the concentration of ^{90}Y and radioactive equilibrium was measured by a low background / gas flow proportional counter (Tenelec, series 5). The minimum detectable activity (MDA) for each radionuclide was estimated to be 8.5 Bq kg^{-1} for ^{226}Ra, 7.0 Bq kg^{-1} for ^{232}Th, and 13.5 Bq kg^{-1} for ^{40}K. The MDAs for ^{137}Cs and ^{90}Sr were estimated to be 0.12 Bq kg^{-1} and 0.2 Bq kg^{-1}, respectively.

Gamma-ray dose rates in the air at a height of 1m above the ground were measured using a pressurized ionization chamber (RSS-1012/3, Reuter stokes) at the sites where soil samples were taken. Gamma-ray dose rates were measured using a counting time of 1800 sec. Exposure rates (R h^{-1}) were converted to gamma-ray dose rates (nGy h^{-1}) in air by multiplying by a factor of 8.66.

The relative frequency distribution of the concentrations of ^{232}Th, ^{226}Ra, ^{40}K, ^{137}Cs and ^{90}Sr in soils and the gamma-ray dose rate in air was computed by counting the numbers of data points lying within the intervals of 2.3 Bq kg^{-1}, 1 Bq kg^{-1}, 20 Bq kg^{-1}, 1.4 Bq kg^{-1}, 0.24 Bq kg^{-1}, and 2 nGy h^{-1}, respectively. The intervals of the nuclides and radiation took 70 divisions into equal parts of maxmum and minimum, respectively. Skewness is a lack of symmetry in a probability distribution. Positive values indicate a shift or tail to the right; negative values indicate a shift or tail to the left. Skewness is 0 for a normal distribution. Kurtosis is a measure of whether the data are peaked or flat relative to a normal distribution. That is, data sets with a high kurtosis tend to have a distinct peak near the mean, decline rather rapidly, and have heavy tails. The symmetry and peakedness in the relative frequency distributions of radionuclides and radiation were represented by the skewness and kurtosis.

The pH in water was measured using a combination pH electrode [7] and organic matter content was determined by dichromate oxidation [8]. Cation exchange capacity [9] was determined using 1N CH_3COONH_4.

RESULTS AND DISCUSSION

The concentrations of the primordial radionuclides ^{232}Th, ^{226}Ra and ^{40}K and the artificial radionuclides ^{137}Cs and ^{90}Sr in soil, the gamma-ray dose rate in air were analyzed to derive the statistical estimates in soil for each province and the country as a whole. The means and ranges

Table 1. The concentrations of radionuclides in soil and the gamma-ray dose rate in air for the nine provinces, the whole country and the world.

Region	Mean (Bq kg^{-1})						Range (Bq kg^{-1})					
	^{232}Th	^{226}Ra	^{40}K	r-ray dose*	^{137}Cs	^{90}Sr	^{232}Th	^{226}Ra	^{40}K	r-ray dose*	^{137}Cs	^{90}Sr
Kyonggi-do	84	43	772	97	37	2.1	35~160	25~91	399~1360	57~141	9.6~80	0.34~7.1
Kangwon-do	65	38	731	91	45	7.8	15~139	11~108	320~1560	33~148	7.3~100	1.9~17
Chungchongbuk-do	70	38	788	95	29	4.2	22~160	14~62	537~1200	69~145	4.7~68	1.3~12
Chungchongnam-do	65	37	780	85	30	2.7	24~129	20~56	398~1330	48~145	9.4~53	0.72~5.4
Chollabuk-do	56	29	687	75	30	3.0	24~ 93	18~40	248~ 967	49~107	12~50	<0.17~8.2
Chollanam-do	60	32	629	86	35	4.3	29~110	17~48	391~1140	46~125	15~73	<0.17~13
Kyongsangbuk-do	50	29	698	76	23	4.7	16~156	9~72	217~1240	39~135	4.5~51	1.4~12
Kyongsangnam-do	53	29	610	70	34	3.2	26~282	14~59	221~1180	23~140	9.0~82	1.2~11
Cheju-do	34	25	360	38	54	13	19~ 51	14~38	203~504	23~53	29~117	6.5~25
Korea	60	34	693	79	35	5.0	15~282	9~108	203-1560	23~148	4.5~117	<0.17~25
World	45	32	420	55								

* gamma-ray dose rate measured with a pressurized ionization chamber (nGyh^{-1})

of the concentrations of ^{232}Th, ^{226}Ra, ^{40}K, ^{137}Cs, ^{90}Sr and the gamma-ray dose rates are presented in Table 1.

^{232}Th, ^{226}Ra, ^{40}K Concentrations

The national mean concentrations of ^{232}Th, ^{226}Ra and ^{40}K in soil were 60±31, 33±14 and 673±238 Bq kg^{-1}, respectively. The concentrations of ^{232}Th, ^{226}Ra and ^{40}K for the country as a whole were in the ranges of 15 to 282, 9 to 108 and 203 to 1560 Bq kg^{-1}, respectively. The lowest level of provincial mean concentration for ^{232}Th (34 Bq kg^{-1}) was found in Cheju-do, while the highest level (84Bq kg^{-1}) was observed in Kyonggi-do. Similarly, the lowest (25 Bq kg^{-1}) provincial mean concentration of ^{226}Ra was observed at Cheju-do and the highest (43 Bq kg^{-1}) concentration was recorded at Kyonggi-do. If Cheju-do is excluded, the provincial mean concentrations of ^{232}Th and ^{226}Ra varied in the range of 50 to 84 Bq kg^{-1} and 29 to 43 Bq kg^{-1}, respectively. The provincial mean concentrations of ^{40}K varied widely between 360 and 788 Bq kg^{-1}, the lowest (360 Bq kg^{-1}) being observed in Cheju-do and the highest (788 Bq kg^{-1}) at Chungchongbuk-do.

The low concentrations of ^{232}Th, ^{226}Ra and ^{40}K observed on the island of Cheju can be attributed to the volcanic ash origin of the soil which is composed mainly of allophane [3]. The mean concentrations of ^{232}Th, ^{226}Ra and ^{40}K in soils from areas that essentially consisted of sediments were lowest, and those from areas where granite and shale formations were prevalent were higher [10]. Similarly, in Hong Kong, the natural radioactivity contents from acidic volcanic areas are less than those from granite areas [11]. Overall, the worldwide estimates of the concentrations of ^{232}Th, ^{226}Ra and ^{40}K are 45 Bq kg^{-1} for ^{232}Th, 32 Bq kg^{-1} for ^{226}Ra, and 420 Bq kg^{-1} for ^{40}K [12] . The mean concentrations of ^{232}Th and ^{40}K observed in Korean soils are thus 1.33, and 1.65 times higher, respectively than the world mean and the concentration of ^{226}Ra was 1.06 times higher [12]. However, the mean concentrations of ^{232}Th, ^{226}Ra and ^{40}K in Cheju-do were 0.76, 0.78 and 0.86 times lower, respectively than the world means, due to the presence of volcanic ash, with a low thorium, radium and potassium content over the island of Cheju.

^{137}Cs and ^{90}Sr Concentrations

The national mean ^{137}Cs concentration in the soil from 233 sampling sites was 35±9.3 Bq kg^{-1}, with values ranging between 4.5 to 117 Bq kg^{-1}. Soil samples from Kyongsangbuk-do had the lowest (23 Bq kg^{-1}) mean concentration, while those from the island of Cheju had the highest. The high concentration of ^{137}Cs on Cheju-do can be attributed to the high cation exchange capacity, the organic matter content, and the presence of allophane, in the soil which cause ^{137}Cs to be tightly bound by soil particles and organic matter [4].

The concentrations of ^{90}Sr that were found in soil samples ranged from <0.17 to 13 Bq kg^{-1}, with a mean of 5.0 Bq kg^{-1} with the exception of Cheju-do where the mean concentration of ^{90}Sr was observed to be 12.5 Bq kg^{-1}. The highest concentrations of ^{90}Sr in the nine provinces were observed in Cheju-do and varied between 6.5 and 25 Bq kg^{-1}. The concentration of ^{90}Sr in Korean soils is similar to that found in other countries located at the same latitude of the northern hemisphere [13,14]. The mean concentration of ^{90}Sr was found to be 1 to 2 orders of magnitude less than the mean concentrations of natural radionuclides (^{232}Th, ^{226}Ra and ^{40}K) and artificial radionuclides (^{137}Cs). The artificial radionuclides ^{137}Cs and ^{90}Sr were deposited in the soil of Korea as a result of atmospheric radioactive fallout following worldwide atmospheric tests of nuclear devices.

Gamma-ray dose rate

The mean gamma-ray dose rate in air from 233 sampling sites is 79 nGyh^{-1}, excluding the contribution of cosmic rays. The background contribution from cosmic rays (32.0 nGyh^{-1}), has been determined by placing a detector on a boat far from shore in the East Sea (Sea of Japan) [1]. Table 1 provides the measured gamma-ray dose rates at 1m above the ground observed in Korea as well as the world means as reported by the United Nations Scientific Committee on the Effect of Atomic Radiation [12]. The values of the gamma-ray dose rate ranged from 23 to 148 nGy h^{-1} for all of the locations. The mean outdoor gamma-ray dose rate in Korea is about 1.5 times higher than the world mean (55 nGy h^{-1}) reported by UNSCEAR[12]. The mean gamma-ray dose rate was the highest in Kyonggi-do (97 nGy h^{-1}), while the island of Cheju has the lowest rate overall (38 nGy h^{-1}) due to it is the parent material of the soil. The mean gamma-ray dose rates from Kyonggi-do, Kangwon-do and Chungchongbuk-do are higher than those from Kyongsangnam-do and Cheju-do.

Frequency Distributions

The frequency distributions of the concentrations of radionuclides and gamma-ray dose rate were investigated. The kurtosis and skewness of the frequency distributions are shown in Fig. 2. The kurtosis of the frequency distribution of ^{226}Ra and ^{137}Cs concentrations is slightly less than zero, but that kurtosis of ^{232}Th, ^{40}K, ^{90}Sr and the gamma-ray dose rate is larger than 0. From the value obtained for the kurtosis coefficient, it can be deduced that the tails of the ^{226}Ra and ^{137}Cs concentration distributions are slightly fatter than those of a corresponding normal distribution. However, the tails of the ^{232}Th, ^{40}K, ^{90}Sr and gamma-ray dose rate distributions are thinner than those of a normal distribution. Skewness of the frequency distributions of all of the radionuclides and the gamma-ray dose rate are positive. Given a skewness coefficient value of approximately +1, ^{232}Th, ^{226}Ra, ^{40}K, ^{137}Cs, ^{90}Sr and the gamma-ray dose rate are approximately normal distributions.

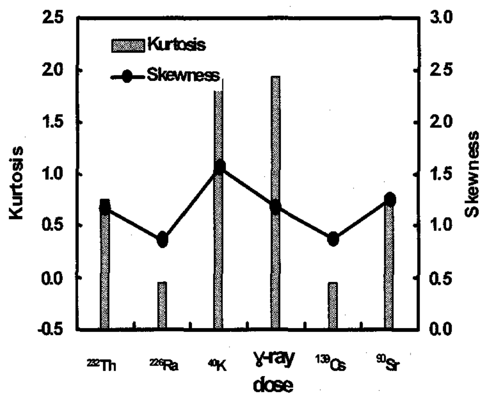

Fig. 2. The kurtosis and skewness for the concentrations of radionuclides and gamma-ray dose rates in Korea.

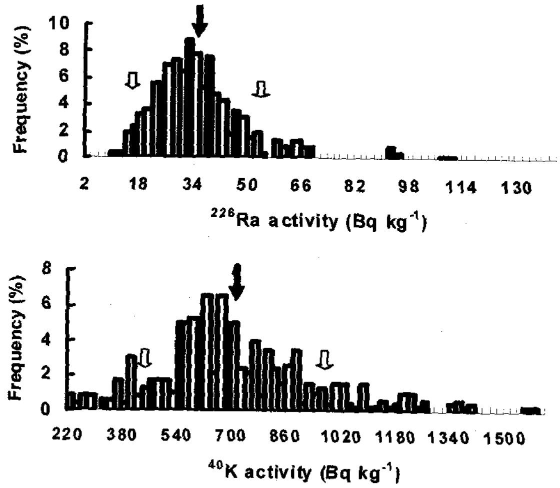

Fig. 3. Relative frequency distribution for the concentrations, (upper) of ^{226}Ra, and (down) of ^{40}K. The solid- and thin arrowed line indicate the mean and standard deviation.

That is to say, around +1 values indicated that the normal distributions are to be a little skewed to the right or positively skewed. Fig. 3 shows the frequency distributions for the concentrations of ^{226}Ra and ^{40}K. The frequency distributions for the mean concentrations of ^{226}Ra are nearly Gaussian (normal) distribution, which was almost symmetrical with respect to the mean. ^{40}K are the nearest in lognormal distributions than the other.

Effect of soil characteristics in the distribution of ^{137}Cs, ^{90}Sr

Items that analyzed the content of soil were pH, organic matter content (OMC), cation exchange capacity (CEC) in soils (Table 2). The pHs of soil samples were in the range of 3.7~7.3, with the highest at Cheju-do and the lowest at Kyonggi-do, most of which were below 5.0. The mean pH in the nine provinces was 4.8 which was 1 unit lower than that of Cheju-do. The mean OMC in the soil was 7.31±3.30%, with values ranging between 1.29 to 26.6%. Cheju-do had the highest (15.7%) mean content, while that from Kyongsangbuk-do had the lowest. The CEC that was found in soil ranged from 4.3 to 94.4 cmol kg^{-1}, with a mean of 21.3±14.1 cmol kg^{-1}. The highest CEC in the nine provinces were observed in Cheju-do and varied between 22.4 and 94.4 cmol kg^{-1}. The volcanic ash soil of Cheju-do was mostly made of allophone. The allophone had high organic matter content and CEC.

The relationship of radionuclides (^{232}Th, ^{226}Ra, ^{40}K, ^{137}Cs, ^{90}Sr), radiation and soil properties

Table 2. Characteristics of soil samples used in the study.

Region	Mean			Range		
	pH	Organic matter content (%)	Cation exchange capacity (cmol kg^{-1})	pH	Organic matter content (%)	Cation exchange capacity (cmol kg^{-1})
Kyonggi-do	4.3	6.75	20.3	3.7~4.8	2.22~13.2	7.4~39.6
Kangwon-do	5.1	8.85	23.9	4.4~7.3	1.68~13.8	4.4~45.2
Chungchongbuk-do	4.7	6.43	19.6	4.1~5.2	2.02~11.9	9.6~29.6
Chungchongnam-do	4.5	6.03	20.3	4.2~5.0	2.49~11.4	9.0~61.8
Chollabuk-do	4.7	5.11	11.4	4.0~5.9	2.42~8.50	6.6~18.3
Chollanam-do	4.9	6.36	14.0	4.5~5.8	2.28~14.7	7.2~32.3
Kyongsangbuk-do	5.0	4.68	11.5	4.5~5.9	1.76~13.5	4.6~21.4
Kyongsangnam-do	4.8	6.11	13.8	4.2~5.8	1.29~11.1	4.3~27.4
Cheju-do	5.8	15.7	57.1	4.9~7.2	8.87~26.6	22.4~94.4
Korea	4.8	7.31	21.3	3.7~7.3	1.29~26.6	4.3~94.4

Table 3. The cross-correlation coefficients of radionuclides, radiation and soil properties.

	^{232}Th	^{226}Ra	^{40}K	PIC	^{137}Cs	^{90}Sr	pH	OMC	CEC
^{232}Th	1	0.58	0.43	0.69	0.00	-0.07	-0.21	-0.13	-0.14
^{226}Ra		1	0.38	0.58	0.02	-0.02	-0.20	-0.07	-0.08
^{40}K			1	0.50	-0.28	-0.28	-0.15	-0.42	-0.36
PIC				1	-0.10	-0.07	-0.22	-0.19	-0.24
^{137}Cs					1	0.60	-0.19	0.73	0.50
^{90}Sr						1	0.21	0.64	0.52
pH							1	-0.06	0.16
OMC								1	0.81
CEC									1

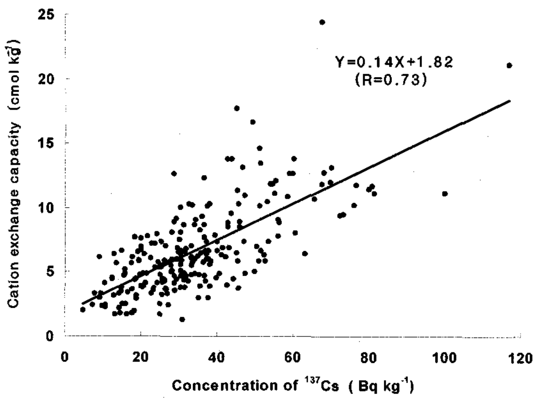

Fig. 4. Cross-correlation between ^{137}Cs concentration and cation exchange capacity(CEC).

can be inferred by means of cross-correlation analysis. The positive or negative cross-correlation among radionuclide, radiation and soil properties at 233 locations indicates that their variations at each pair vary in phase or out of phase, respectively. The cross-correlation coefficients of radionuclides, radiation and soil properties are shown in Table 3. Table 3 shows that natural radionuclides were also relatively well correlated each other, but natural radionuclides were no correlation with artificial radionuclides and soil characteristics. The pH, another representative soil characteristic, had no correlation(r=-0.19) with the concentration of ^{137}Cs in the soil, which is consistent with the results [15]. The correlation (r=0.41) of ^{90}Sr concentration with pH was found a slightly significant, which affected ^{90}Sr mobility in soil. When ^{137}Cs comes into contact with soil, it is strongly absorbed on the surface of organic matter immediately [16]. OMC is major components that make CEC of the soil samples. There was a highly significant correlation between ^{137}Cs concentration and CEC (Fig. 4). The ^{137}Cs concentrations also showed a highly significant correlation with OMC (r=0.73).

CONCLUSIONS

From the measurement of the radionuclides (natural and artificial) concentration and gamma-ray dose rate at 233 locations in Korea, we have calculated radionuclide concentrations and gamma-ray dose rate for each province and the country as a whole.

The mean national concentrations of ^{232}Th, ^{226}Ra, ^{40}K, ^{137}Cs and ^{90}Sr detected in soil samples were 60±31, 33±14, 673±238, 35±9.3 and 5.0±3.4 Bq kg^{-1}, respectively. The mean levels of ^{232}Th, ^{226}Ra and ^{40}K detected were 1.33, 1.06 and 1.65 times higher, respectively than those attributed to the world mean reported by UNSEAR[12] . The mean concentration of ^{90}Sr was 5.0 Bq kg^{-1}, which was found to be 1 to 2 orders of magnitude less than the mean concentrations of natural radionuclides (^{232}Th, ^{226}Ra and ^{40}K) and artificial radionuclides (^{137}Cs). The concentration of ^{137}Cs and ^{90}Sr in soil was presumed to be the results of fallout of radioactivity from the atmospheric tests mainly conducted around the world till 1962. The mean concentrations of the artificial radionuclides (^{137}Cs and ^{90}Sr) detected in Cheju-do were higher than those in the other provinces, while the mean concentrations of the natural radionuclides (^{232}Th, ^{226}Ra and ^{40}K) in Cheju-do were lower than those in the other provinces. Those are due to the presence of volcanic ash over the Cheju-do. The distribution of ^{226}Ra and ^{137}Cs are to be a little skewed to the right, its tails are fatter than those of a normal distribution. The tails of ^{232}Th, ^{40}K, ^{90}Sr and gamma-ray dose rate distribution were thinner than those of a normal distribution. The measured gamma-ray dose rate varied between 23 and 148 nGyh^{-1}, the mean value being 79±18 nGyh^{-1}. Additionally, the relationships between radionuclide and soil properties (pH, OMC, CEC) were studied. The ^{137}Cs had highly significant correlations with OMC and CEC while ^{90}Sr concentration had relatively smaller correlation with pH.

These results will provide a valuable and useful reference for the design and development of studies in specific regions of Korea that show enhanced levels of radiation, natural and artificial radioactivity.

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