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DISTRIBUTION ANALYSIS AND MEAN RESIDENCE TIME OF ^{90}SR IN WET FALLOUT IN ZAGREB

Analiza distribucije i srednje vrijeme boravka ^{90}Sr u mokrom radioaktivnom taloženju u Zagrebu

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Abstract – The results of the analysis of long-term measurements of ^{90}Sr radioactivity in the wet fallout in the Zagreb area are presented. The data are lognormally distributed and the parameters of the lognormal distribution are interpreted. The mean residence time of ^{90}Sr in wet fallout is calculated by means of the obtained distribution parameters.

Key word index: ^{90}Sr, wet fallout, distribution analysis, mean residence time.

Sažetak – Prikazani su rezultati dugogodišnjega mjerenja radioaktivnosti ^{90}Sr u mokrom radioaktivnom taloženju u zagrebačkoj regiji. Pokazano je da su aktivnosti raspodijeljene po logaritamski normalnoj razdiobi. Interpretirano je značenje parametara te razdiobe. Izračunato je srednje vrijeme boravka ^{90}Sr u radioaktivnim oborinama na temelju dobivenih parametara pridijeljene lognormalne razdiobe.

Ključne riječi: ^{90}Sr, mokro radioaktivno taloženje, distribucijska analiza, srednje vrijeme boravka.

1. INTRODUCTION

Radioactive fallout is the material deposited on the Earth's surface, consisting of radioactive particles which have been released into the atmosphere as a result of nuclear explosions and by discharge from nuclear power plants and other nuclear installations. Radioactive fallout is, therefore, produced through nuclear fission and the activation of soil, water and other material close to the site of the detonation. The atmosphere is depleted through *wet fallout* and *dry fallout* (the process of dry deposition on land surfaces or plant cover). Rain-out caused by droplet formation within clouds and wash-out caused by falling raindrops picking up radioactivity are commonly known as wet fallout.

Radioactive fallout, which resulted from the large-scale nuclear weapon tests in the atmosphere conducted during the 1960s, followed by similar, but smaller-scale tests by the Chinese and French in the 1970s and afterwards, was the dominant route for the introduction of fission (i.e. artificial) radionuclides in the atmosphere until the nuclear accident at Chernobyl, former USSR, in 1986. Among these, ^{90}Sr has been regarded as a fission product of great potential hazard to living beings because of the unique combination of its 28-year long half-life, the very energetic beta particle of its ^{90}Y daughter, and its general resemblance to calcium in metabolic processes.

Owing to the increased deposition of radioactive material on the Earth's surface after the most intensive atmospheric nuclear weapon testing in the 1960s, the total ^{90}Sr deposition density (^{90}Sr radioactivity deposited per unit area, i.e. surface deposit) for the Zagreb area for the 1962–1967 period (6 years) was 2896 Bqm^{-2}. In the 1968 to 1993 period (26 years) the total ^{90}Sr surface deposit was 1219 Bqm^{-2}. Although between 1962 and 1967 the precipitation amount in Zagreb, Ksaverska cesta 2, was only 6.1 m, which is 21.2 % of the precipitation for the whole 1962 –1993 period (28.8 m), the surface deposit in that period contributed as much as 70.4 % to the total 1962–1993 surface deposit.

In the explosions and subsequent fire of the graphite moderator in the Chernobyl nuclear power plant approximately 8.0 · 10^{15} Bq (i.e. 4% of the reactor inventory) of ^{90}Sr was released (IAEA, 1986) in three waves (UNSCEAR, 1988). As opposed to more volatile radionuclides (i.e. caesium isotopes), the refractory components of the Chernobyl debris were deposited closer to the accident location. Therefore, due to the refractory nature of strontium, the nuclear accident at Chernobyl has not caused a significant long-term increase (the exception being only May 1986) of ^{90}Sr deposition in the Republic of Croatia, unlike ^{137}Cs (Franić, 1992a). This was especially true of the Adriatic region since, owing to the then prevailing meteorological conditions, air plumes did not pass over the mid-Adriatic (UNEP 1991).

In the Republic of Croatia, increased ^{90}Sr activities persisting for several years were detected only in cistern waters (Franić, 1992b; Franić, 1993). Cisterns are filled with rain water, usually collected from very large surfaces (roofs etc.), which washes out ^{90}Sr (as well as any other) dry fallout, resulting in increased concentrations of fallout radionuclides in cistern waters.

The Department for Radiation Protection of the Institute for Medical Research and Occupational Health in Zagreb has carried out radioactivity measurements in the fallout as part of an extended monitoring programme, since 1962.

This paper summarizes the measurement results and evaluates the data of ^{90}Sr in wet fallout for the period 1962 – 1993.

2. METHODS AND MATERIAL

Wet fallout samples were collected monthly at the location of the Institute, Zagreb, Ksaverska cesta 2. The funnels used for wet fallout collection had a 1 m^2 area. The precipitation amount was measured by means of Hellman pluviometer.

After radiochemical treatment, the radioactivity of ^{90}Sr was determined by beta-counting its decay product, ^{90}Y, in a low-background anti-coincidence, shielded Geiger–Müller counter. The counting time depended on the sample activity, but was never less than 60,000 s.

Efficiency calibration was carried out using sources provided by the International Atomic Energy Agency (IAEA) and the World Health Organization (WHO).

3. RESULTS AND DISCUSSION

Lognormal activity distribution of ^{90}Sr in wet fallout

After the moratorium on atmospheric nuclear weapons testing in the early 1960s, (in 1963) there has been an exponential decrease in ^{90}Sr wet fallout activity. Smaller-scale tests of declining frequency by the Chinese and French in the 1970s and later, as well as a variety of environmental physical factors that naturally fluctuate were the reason for a scanty transient increases in the activity. In order to discern natural or expected variations or differences from unexpected, a distribution analysis of ^{90}Sr wet fallout activities was performed.

There are a number of well-known distributions, the one most commonly used being the normal (Gaussian) distribution. It is applied to natural processes that are influenced by a great number of factors none of which are dominant. But environmental radioactivity data can also be described by other distributions like Weibull (Apt, 1976) or lognormal. The latter is commonly used with data collected over a longer time period (Waite, 1976; Pilar, 1980; UNSCEAR, 1982; Mitič, 1989). Thus, it was expected that the exponential de-

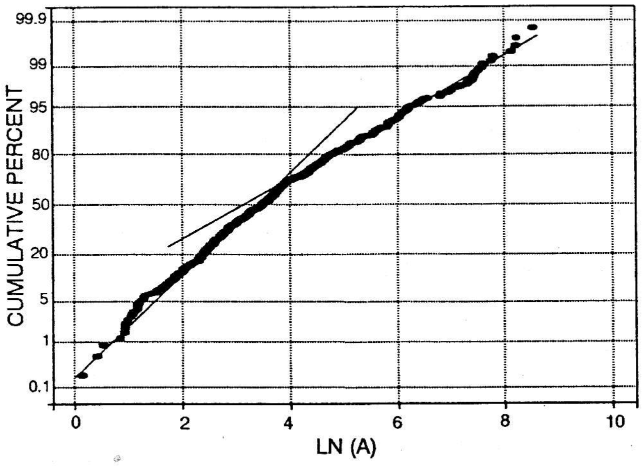
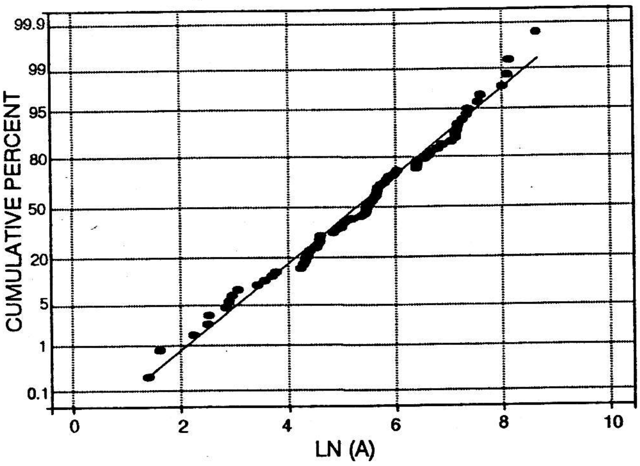


Figure 1. The cumulative distribution curve for logarithms of ^{90}Sr radioactivities (Bqm^{-3}) in wet fallout in Zagreb for the 1962–1993 period.

Slika 2. Krivulja kumulativne razdiobe logaritma radioaktivnosti ^{90}Sr (Bqm^{-3}) u mokrom taloženju u Zagrebu u razdoblju 1962–1993.



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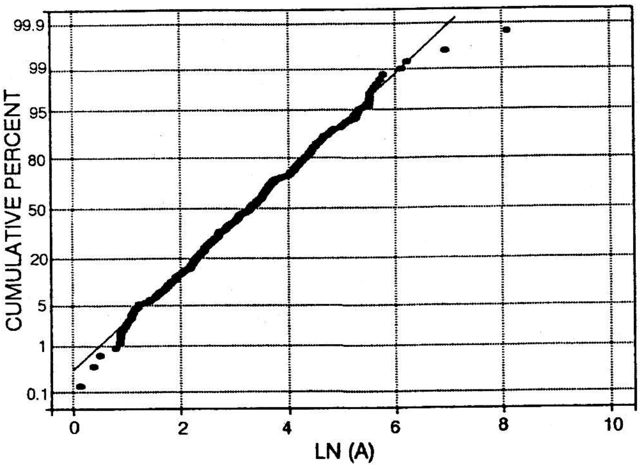

Figure 3. The cumulative distribution curve for logarithms of ^{90}Sr radioactivities (Bqm^{-3}) in wet fallout in Zagreb for the 1968–1993 period.

Slika 3. Krivulja kumulativne razdiobe logaritma radioaktivnosti ^{90}Sr (Bqm^{-3}) u mokrom taloženju u Zagrebu u razdoblju 1968–1993.

crease of ^{90}Sr wet fallout activity in the 1962–1993 period will be reflected in the lognormal distribution.

The distribution of a variable x (in this case activity) is said to be lognormal:

- if the domain of its values is $(0,+\infty)$, and
- if $ln(x)$ is normally distributed.

If $y = \ln(x)$ and the associated normal distribution is $N(\mu,\sigma^2)$, then the probability density function (the familiar Gaussian bell curve) of variable y is given by:

$$f(y) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{(y-\mu)^2}{2\sigma^2}} \qquad (1)$$

where:

μ is the mean value of variable y, and

σ^2 is variance.

The probability density function of variable x is:

$$f(x) = \frac{1}{x\sigma\sqrt{2\pi}}\, e^{-\frac{[\ln(x)-\mu]^2}{2\sigma^2}} \qquad (2)$$

Introducing Z as a function of the random variable x:

$$Z = \frac{ln(x)-\mu}{\sigma} \qquad (3)$$

The distribution function $F(Z)$ of the continuous variable Z is defined as the sum of all probabilities (cumulative probability) of Z:

$$F(Z) = \int_{-\infty}^{Z} f(Z)dZ \qquad (4)$$

Function (4) of the normal distribution, when plotted against a suitable scale, is a straight-line graph (*probability paper*). The mean value μ, corresponds to the 50th percentage of the line, while the slope is given (defined) by the standard deviation σ.

The hypothesis that ^{90}Sr activities, A, in wet fallout samples (collected monthly) are log-normally distributed was tested. Figure 1 shows the cumulative distribution function (4) of activity logarithms for the 1962–1993 period. The *bimodal behaviour* exhibited (data do not fit one straight line) reflects two distinct lognormal distributions. That could also be expected from the data on fallout deposition, since a major fraction of the total surface deposit for the 1962–1993 period was deposited immediately after the most intensive atmospheric nuclear weapon tests had taken place. Therefore, the 1962–1993 period was divided in two parts: the period immediately after the nuclear moratorium in 1963 and the rest.

The best fit of the cumulative distribution functions $F(ln\ A)$ to the straight line were observed for the 1962–1967 (Figure 2) and 1968–1993 periods (Figure 3), exhibiting no bimodal behaviour.

The two data on the Figure 3 which do not fit the straight line reflect the nuclear accident in Chernobyl in April 26, 1986. Fortunately, as stated in the introduction, the Chernobyl nuclear accident did not cause any significant long term increase in ^{90}Sr in the Croatian environment.

Once a distribution type is identified for a set of data, which has been obtained from the distribution of population, it is necessary to deduce the statistical parameters that describe it. The normal distribution of population, in this case activity logarithms ln(A), is characterized by the equation (Pilar, 1980):

$$y = ln(A) = \mu \pm k\sigma \qquad (5)$$

Parameter k stands for the contribution in percentage of the distribution spread which is included. Therefore:

$k = 1$ implies a 68 % confidence level, or that about 2/3 of the distribution is included.

$k = 2$ implies a 95 % confidence level.

In our case the parameters in equation (5) are:

$y = \ln(A)$ logarithms of activity,

μ the arithmetic mean of the activity logarithms and

σ the standard deviation of the activity logarithms.

For the lognormal distribution, the mathematical equation corresponding to (5) is:

$$A = \mu_g g^{\pm k} \qquad (6)$$

where:

$\mu_g = e^{\mu}$ is the geometric mean of wet fallout activity, i.e. the antilogarithm of the arithmetic mean of the wet fallout activity logarithms,

$g = e^{\sigma}$ the geometric standard deviation of the wet fallout activity.

Table 1 shows the parameters of normal distribution of the wet fallout activity logarithms and Table 2 shows the parameters of lognormal distribution of wet fallout activities.

According to Pilar (1980), the parameters in Table 1 may be interpreted by using definition (5) and Table 1, or by applying equation (6) to the data in Table 2.

Therefore, from (5):

1a. In the 1962–1967 period, for 68 % of the time (k=1), ^{90}Sr wet fallout activity was between
$\exp(5.57 - 1.26) = e^{4.31} \approx 74$ Bqm^{-3} and
$\exp(5.57 + 1.26) = e^{6.83} \approx 925$ Bqm^{-3}.
The most probable activity was
$\exp(5.57) = e^{5.57} \approx 262$ Bqm^{-3},
which is significantly different from the arithmetic average for that period (513 Bqm^{-3}).

1b. In the 1962-1967 period, for 95 % of the time (k=2), ^{90}Sr wet fallout activity was between
$\exp(5.57 - 2 \times 1.26) = e^{3.05} \approx 21$ Bqm^{-3} and
$\exp(5.57 + 2 \times 1.26) = e^{8.09} \approx 3262$ Bqm^{-3}.
The maximal measured activity, 3329 Bqm^{-3}, was recorded in April 1963, and the minimal activity, 12 Bqm^{-3}, in November 1967. The arithmetic average overestimates most probable activity in that period by approximately a factor of 2.

Table 1. Parameters of normal distribution

Tablica 1. Parametri normalne razdiobe

Observation period	μ	σ
1962–1967	5.57	1.26
1968–1993	3.22	1.22
1962–1993	3.65	1.53

Table 2. Parameters of lognormal distribution

Tablica 2. Parametri lognormalne razdiobe

Observation period	μ_g	g
1962–1967	262.43	3.52
1968–1993	25.03	3.39
1962–1993	38.47	4.62

average overestimates most probable activity in that period by approximately a factor of 2.

2a. In the 1968–1993 period, for 68 % of the time ($k=1$), ^{90}Sr wet fallout activity was between
$\exp(3.22 - 1.22) = e^{2.00} \approx 7$ Bqm^{-3} and
$\exp(3.22 + 1.22) = e^{4.44} \approx 85$ Bqm^{-3}.
The most probable activity was:
$\exp(3.22) = e^{3.22} \approx 25$ Bqm^{-3}.
The arithmetic average for that period is ≈ 60 Bqm^{-3}.

2b. In the 1968–1993 period, for 95 % of the time ($k=2$), ^{90}Sr wet fallout activity was between
$\exp(3.22 - 2 \times 1.22) = e^{0.78} \approx 2$ Bqm^{-3} and
$\exp(3.22 + 2 \times 1.22) = e^{5.66} \approx 287$ Bqm^{-3}.

The maximal activity, 2947.5 Bqm^{-3}, was recorded in May 1986, immediately after the Chernobyl accident and the minimal activity, 1.3 Bqm^{-3}, was measured in March 1991. The arithmetic average overestimates the most probable activity in this period by more than 2 times.

The mean Residence Time of ^{90}Sr in Wet Fallout in Zagreb

The mean residence time (MRT) gives a measure of how far the released substance can travel before deposition, how much of the substance can the atmosphere load and its flux to the Eearth's surface, being, therefore, an important parameter relevant to models connected with long distance meteorological processes.

The concept of *mean residence time* (sometimes also called *turn-over time*) evolves from the reservoir theory and has been discussed in literature (Rodhe, 1978; Rangarajan, 1992). MRT is defined as the time spent by a particle (or pollutant) in the considered reservoir (compartment) from formation or entry to transformation or removal from the system. When the removal process can be described by first order kinetics, as in radioactive decay, the residence time is the reverse of the removal rate coefficients:

$$MRT = \frac{T_{1/2}}{ln(2)} = \frac{1}{k} \qquad (7)$$

where $T_{1/2}$ is the half-residence time.

The mean residence time of ^{90}Sr in wet fallout was estimated for the periods 1962–1967 and 1968–1993. By function minimization, using the Simplex method (Nelder, 1965), the observed data were fitted to the equation which gives ^{90}Sr wet fallout activity as a function of time:

$$A(t) = A(0)e^{-k_{eff}t} \qquad (8)$$

where:

$A(t)$ is ^{90}Sr wet fallout activity at time t,
$A(0)$ is the initial ^{90}Sr wet fallout activity, i.e. $t=0$,
t is time and
k_{eff} is the effective removal rate of ^{90}Sr wet fallout activity.

For several removal processes:

$$k_{eff} = \sum_{i=1}^{n} k_i \qquad (9)$$

k_i being the effective constant for each removal process respectively.

For two removal processes (radioactive decay and deposition to the Earth's surface) using definition (7) and equation (9) for ^{90}Sr mean residence time in wet fallout is obtained:

$$MRT = \frac{1}{\frac{1}{MRT_{eff}} - \lambda} = \frac{1}{\frac{ln(2)}{T_{eff1/2}} - \lambda} = \frac{1}{k_{eff} - \lambda} \qquad (10)$$

where

MRT_{eff} is the effective mean residence time of ^{90}Sr in wet fallout,
$T_{eff1/2}$ is the effective half residence time of ^{90}Sr in wet fallout and
λ is the decay constant for ^{90}Sr ($7.81 \cdot 10^{-10}$ s^{-1}).

The effective half-residence times and mean residence times were calculated using equation (10) and definition (9), and the values for k_{eff} obtained by fitting (Nelder, 1965) the measured ^{90}Sr wet fallout radioactivity data to equation (8). They are shown in Table 3.

The ^{90}Sr mean residence time in wet fallout for the Zagreb area for the 1962–1967 period is in relatively good agreement with literature data for ^{90}Sr effective mean residence time, MRT_{eff}, of 10 months (0.83 years) for the mid–1960s, after the nuclear moratorium in the year 1963 (UNSCEAR, 1982). If data for the year 1962 for the Zagreb area are excluded (since peak activity is in 1963 after which there were no intensive atmospheric nuclear weapon tests), ^{90}Sr $T_{eff1/2} = 0.91$ years, and $MRT = 1.35$ years, which is in better agreement with the UNSCEAR data.

For comparison, ^{90}Sr effective half-residence time in fallout over the Atlantic ocean (Fare islands) for the period from 1963 to 1981 was estimated to be $T_{eff1/2} = 5.5$ years (Aarkrog, 1984) which corresponds to the mean residence time $MRT = 9.8$ years.

Table 3. Effective half-residence and mean residence times for ^{90}Sr wet fallout activity

Tablica 3. Efektivno poluvrijeme i srednje vrijeme boravka ^{90}Sr u mokrom taloženju

Observation period	T_{eff}	MRT
1962–1967	1.27	1.92
1968–1993	7.94	15.94
1962–1993	5.95	10.80

4. CONCLUSIONS

For the 1962–1993 period, ^{90}Sr wet fallout activities in the Zagreb area were distributed log-normally, but two distinct lognormal distributions were identified: for the intervals between 1962 and 1967 and the one between 1968 and 1993. The most probable ^{90}Sr wet fallout activities for the respective periods were 262 and 25 Bqm^{-3}. Therefore, the arithmetic averages, (513 and 60 Bqm^{-3}) based on the assumption of normal population distribution significantly overestimate the most likely value to be encountered.

The mean residence time for ^{90}Sr in wet fallout for the 1962–1967 period is about eight times smaller compared to that for the 1968–1993 period ($\approx$ 16 years, which equals approximately 1/2 of the ^{90}Sr radioactive half-life). The short *MRT* in the first period can be accounted for by the rapid decrease of fission radionuclides in the atmosphere in the mid–1960s, after the cessation of atmospheric nuclear weapon tests.

The transient increases in wet fallout activity caused by the French and Chinese atmospheric nuclear tests, as well as the peak of 2947.5 Bqm^{-3} in May 1986, as a consequence of the Chernobyl nuclear accident, are the reasons for a relatively long MRT in the second period.

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