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ORIGINAL PAPER

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Decrease of total activity with time at long distances from a nuclear accident or explosion

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Abstract Two data groups were analyzed: (1) the exposure rate in the former Czechoslovakia after the Chernobyl accident in 1986, and (2) the decrease of beta activity of an atmospheric fallout sample taken in Bratislava during 24 h on 30 May 1965. Both quantities decreased with the first power of time. This pattern of decrease is explained by applying the same mathematical formalism as is also used to describe the decrease in postnatal mortality with age. Following this formalism, the decrease of total activity with the first power of time could be seen as a consequence of a log-normal distribution of decay constants in the fallout. This differs slightly from earlier results that show the total activity decreasing with a power of 1.2 immediately after the nuclear explosion.

Introduction

It has experimentally been observed that the decrease in total activity from fission products after a nuclear explosion can be calculated by means of a fairly simple formula:

$$A(t) = A(1)\, t^{-\alpha} \quad \alpha > 0 \text{ and } t > 1\,\mathrm{h} \tag{1}$$

where $A(t)$ is the activity at time t, $A(1)$ is the activity in 1 h, and t is the time after explosion. Immediately after a nuclear explosion, the relationship of the concentration of one nuclide to time depends not only on the respective decay constant, but also on the processes in the other decay chains. The composition of such a mixture of nuclides cannot be described as one decay chain. Way and Wigner [1] have shown that the (theoretical) index α in Eq. 1 has a value of $\alpha = 1.2 = 6/5$. According to this derivation, the value $1 + 1/5$ is a consequence of the proportionality between the decay constant x and the fifth power of disintegration energy E (the dependence follows from the experimental results presented here). The fission products were considered a sort of statistical assembly, calculations were made of the β-disintegrations per second and of the total energy emitted per second at any time after fission. They assumed all energies to be equally frequent ($f(E) \approx$const, where $f(E)$ is a frequency function of energy E). Consequently, they used the following formula:

$$A(t) \approx \int_0^\infty f(E).x(E)\, \mathrm{e}^{[-x(E)t]}\, \mathrm{d}E = \int_0^\infty E^5\, \mathrm{e}^{[-E^5 t]}\, \mathrm{d}E$$
$$= \int_0^\infty y^{1/5}\, \mathrm{e}^{[-yt]}\, \mathrm{d}y \approx t^{-(1+1/5)} \tag{2}$$

where x is a decay constant and t is time (the derivation was simplified here). The external factors were neglected (such as meteorological factors, aerosol size, type of landscape, physico-chemical quality of radionuclides, etc.; for more details see [1]).

The experimental value of the index α has been discussed elsewhere [1–9]. The index α has been assumed to approach a value of 1.2 [3, 5]. Izrael [6] proposed that, if the activity of an instantaneous mixture of fission products is calculated using known fission yields and nuclide decay schemes, then index α also approaches a value of 6/5. On the other hand, the composition of fallout at a long distance from a source had to be affected by many external factors. Two data groups have been analyzed here:

1. Daily measurements of exposure rate at nine locations on the territory of the former Czechoslovakia after the Chernobyl accident in 1986 [10].

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2. The beta activity of an atmospheric fallout sample taken on 30 May 1965 in the region of West Slovakia [11].

At a long distance from the nuclear explosion site (or the nuclear accident site), the total activity can be expressed as the sum of exponential terms, which includes the decay constants of specific nuclides. In this sum, the specific exponential term is multiplied by the ratio that corresponds to the specific nuclide concentration in the mixture. The relative nuclide concentration in the mixture at a long distance from the nuclear explosion site (or the nuclear accident site) can be described by a frequency function $f(x)$ of the decay constant x.

Materials and methods

Exposure rate after the Chernobyl accident

After the Chernobyl accident the exposure rates were measured at nine locations in the former Czechoslovakia (see [10] and additionally http://www.suro.cz/pub/cernobyl/zprava_1987.pdf, pp 10–11). The initial measurements were carried out on 30 April 1986 in all locations with the exception of Ústí nad Labem, where the initial measurement was done on 5 May 1986. Table 1 presents the details, such as name of location, geographic coordinates, and the description of instruments and of measurement sites. The decrease with time was analyzed after the natural background subtraction was estimated. The estimates of the natural background were based on the values measured near the actual site using the same instruments (see Table 1 and [10]). The moment of the Chernobyl accident (26 April 1986, 1:24) was used as the origin of time.

Beta activity of two atmospheric fallout samples taken during the 1960s

The beta activities of two atmospheric fallout samples was measured sequentially with time by Petrasova [11]. The first sample was taken on 12 December 1961. From the beginning of September 1961 to 4 November 1961, a great number of nuclear explosions occurred in the region of Novaya Zemlya (former USSR; for more information, see, for example, http://npc.sarov.ru/english/issues/plutonium.html). For this reason, the date of explosion and the origin could not be identified, which means that the data was not analyzed. Figure 1 shows the decrease of beta activity with time in this sample in with the assumption that the date of the last explosions in the region of the Novaya Zemlya (4 November 1961) was used as origin date.

The second atmospheric sample was taken in Bratislava during 24 h on 30 May 1965. Since in 1965 only one atmospheric nuclear explosion occurred anywhere on earth (on 14 May 1965, in China, see http://cns.miis.edu/research/china/coxrep/testlist.htm), the time origin can reliably be determined for this sample. The beta activity of this sample was measured over the period 31 May 1965 to 7 July 1965 [11].

Results

Exposure rate after the Chernobyl accident

Figures 2, 3, 4, 5, and 6 show the time dependence in Prague, Hradec Králové, Ostrava, Brno, and Bratislava. In all cases, the exposure rates decrease linearly in the log–log scale applied. It was confirmed that the residuals from the linear model were not dependent upon the logarithm of time in all cases (the hypothesis that the deviations of logarithm of measured values from the regression line are not dependent on time was not rejected; 95% confidence level [CL]). The slope $-\alpha$ was calculated using linear regression, and the results for all sites are shown in Table 2.

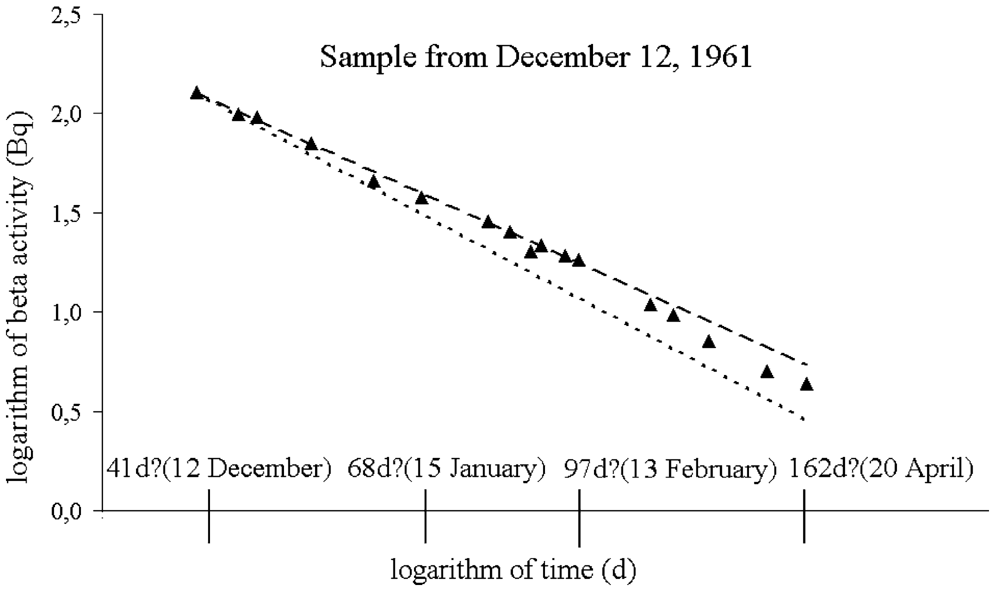
The day when the exposure rate achieved the maximum at a particular site was used as the first point t_1 (most frequently on 1 May 1986; see Table 2). The day when the exposure rate achieved the minimum (for the first time after subtracting the natural background) was used as the last point t_2. It was done separately for every site (all values of exposure rate are also in the website: http://www.suro.cz/pub/cernobyl/zprava_1987.pdf, pp.

Table 1 Locations and measurement methods

Location	Geographic coordinates	Instrument	Description
Prague	50° 04′/14° 26′	RSS-111 (ionization chamber)	1 m above grass
České Budějovice	48° 58′/14° 29′	LB-1310 (proportional counter)	1 m above asphalt
Ústí nad Labem	50° 41′/14° 00′	RUST – 3 (GM tube)	1 m above grass
Hradec Králové	50° 13′/15° 50′	NRG – 302 (plastic scintillator)	1st floor[a]
Ostrava	49° 50′/18° 15′	LB-1310 (proportional counter)	1 m above grass
Brno	49° 11′/16° 39′	RSS-111 (ionization chamber)	1 m above grass
Bratislava	48° 10′/17° 13′	RSS-111 (ionization chamber)	Roof and grass[b]
Banská Bystrica	48° 44′/19° 10′	RUST – 3 (GM tube)	1 m above grass
Košice	48° 44′/21° 15′	RSS-111 (ionization chamber)	1 m above grass

[a]About 3 m above a sidewalk (from the window of the 1st floor)
[b]Roof of annex early, and 1 m above grass later (no more details available)

Fig. 1 Beta activity of atmospheric fallout sample taken on 12 December 1961 versus time in log–log scale. The origin of the time axis could not be specified because a great number of atmospheric nuclear explosions occurred from the beginning of September 1961 to 4 November 1961, and the slope could not be calculated. The date of the last explosions (4 November 1961) was used as the origin date in this projection. The slope of the *dashed straight line* is precisely equal to −1, and the slope of the *dotted straight line* is equal to −1.2. Both these *straight lines* fit the maximum precisely

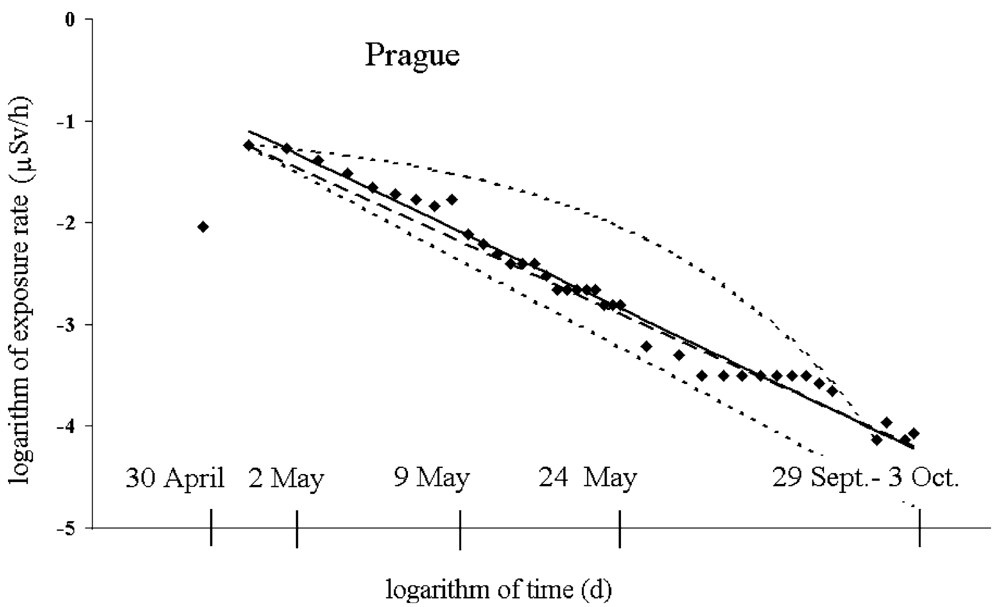


10–11). Both values t_1 and t_2 are shown in Table 2. The hypothesis that slope = −1 was not rejected in all locations with the exception of Bratislava, where the decrease with time was lesser.

The decrease with time can be described for the whole country if one representative value is calculated for all locations of one particular day. Nine values of one particular day can not be used to estimate the type of statistical distribution, and the decision between the arithmetic and geometric mean cannot be realized. Therefore, all exposure rate values were multiplied by the factor t^{α} (see corresponding α values in Table 2), and it was determined that these normalized values for all locations and days together were log-normally distributed ($P < 0.024$). Consequently the geometric means were used and were calculated for all locations together for each day separately. The resulting values have been labeled "Former Czechoslovakia I" (Fig. 7). Since the period was very short and the maximum values small in Usti nad Labem and Kosice (Table 2), a subset without these two locations was created and the geometric means are labeled "Former Czechoslovakia II." The geometric means indicated as Former Czechoslovakia I are shown in Fig. 7. The slope of the set Former Czechoslovakia I is equal to −0.95 (95% CI, −1.03 to −0.87) and it is −1.01 with (95% CI, −1.06 to −0.97), if Usti nad Labem and Kosice (subset Former Czechoslovakia II) are excluded.

The arithmetic mean of the first nine slopes in Table 2 is equal to −1.08 with a standard deviation (SD) of 0.25 for all locations. If the locations Usti nad Labem and

Fig. 2 Decrease of exposure rate along with time (log–log scale) in Prague after the Chernobyl accident. The slope of the *dashed straight line* is exactly equal to −1, and the slope of the *dotted straight line* is equal to −1.2. Both these *straight lines* fit the maximum precisely. The *solid straight line* is the regression line. The *dotted curve* fitting both the maximum and the minimum shows the exponential decrease from the maximum to the minimum



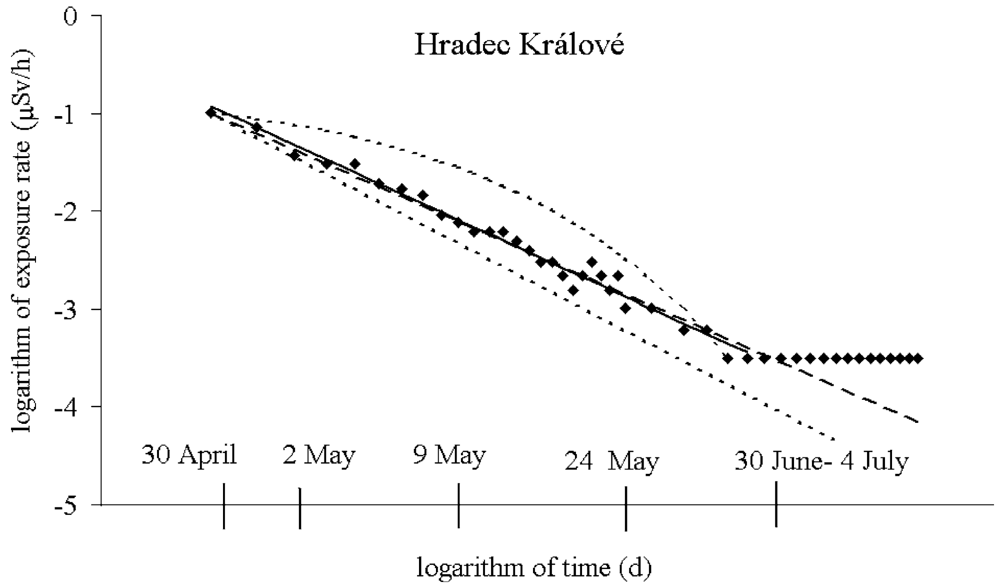


Fig. 3 Decrease of exposure rate along with time (log–log scale) in Hradec Králové after the Chernobyl accident (see description of *lines* in Fig. 2)

Kosice are excluded, then the arithmetic mean is -0.99 with $SD = 0.09$.

The model with the slope -1 ($\alpha = 1$ in Eq. 1) describes the data very well. The straight line, which was drawn with the slope -1 through the maximum (dashed straight line), is practically identical with the regression line (solid straight line) in Hradec Králové with the highest coefficient of determination $r^2 = 0.98$ (r^2 represents the fraction of variability in y that can be explained by the variability in x, it is simply the square of the correlation coefficient; see Table 2 and Fig. 3).

For six of nine locations, the 95% confidence interval does not contain the value -1.2 which is included only only for Ostrava, Ústí nad Labem, and Košice—i.e., the sites with low coefficients of determination and major confidence intervals (Table 2). Consequently, the model with the slope -1.2 was not able to describe the data.

If the fallout contains only radionuclides with one decay constant (in practice one radionuclide) then the activity decreases exponentially with time (corresponding to linear decrease in the semi-logarithmic scale). The heterogeneity of a mixture of decay constants can possibly be demonstrated by the difference between the exponential decrease and the data in Figs. 2–7. The dotted curves in Figs. 2–7 (log–log scale) correspond to the hypothetical exponential decrease. These relationships are linear in the semi-logarithmic scale. Two parameters of this exponential relationship were calculated from the coordinates of the maximum and the minimum value in a particular location (they are un-

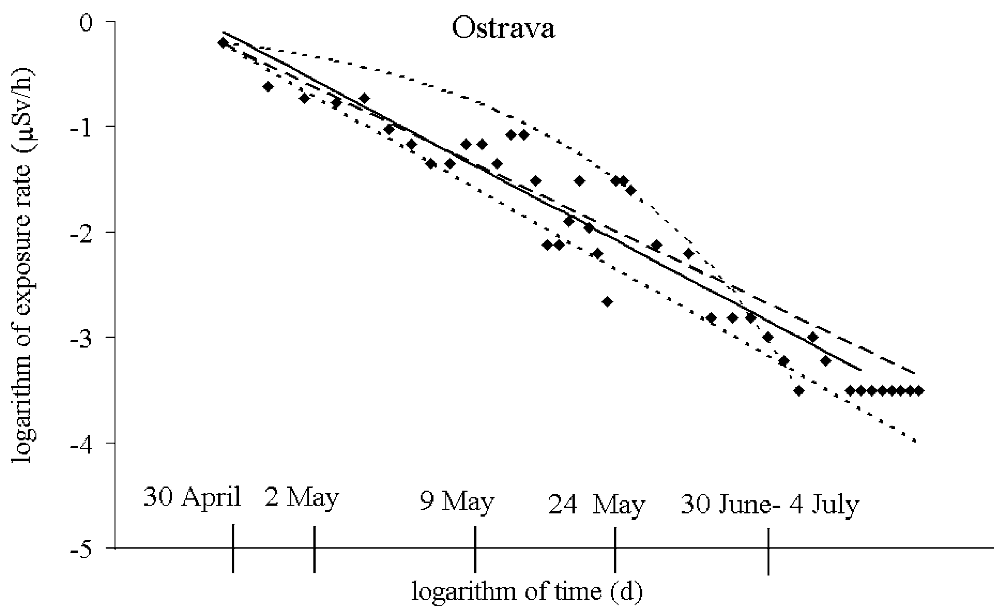


Fig. 4 Decrease of exposure rate along with time (log–log scale) in Ostrava after the Chernobyl accident (see description of *lines* in Fig. 2)

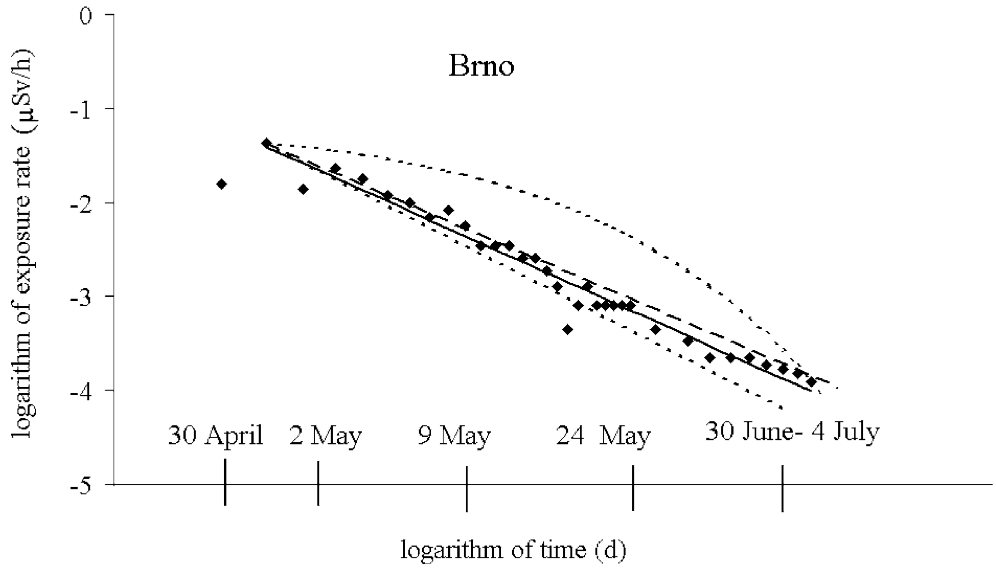


Fig. 5 Decrease of exposure rate along with time (log–log scale) in Brno after the Chernobyl accident (see description of *lines* in Fig. 2)

iquely determined by the coordinates of these two points that are therefore precisely intersected).

Decrease of beta activity in the sample taken on 30 May 1965

Figure 8 (log–log scale) shows the decrease of beta activity with time in the sample taken on 30 May 1965. The slope was calculated using the linear regression in the log–log scale for all values recorded (31 May 1965 to 7 July 1965). It is equal to -1.01 (95% CI, -1.05 to -0.98). Consequently, the hypothesis that $-\alpha = -1.2$ was also rejected. The dashed straight line has a slope equal to -1 and the dotted straight line has a slope equal to -1.2 in Fig. 8. Both these straight lines intersect the first point. The solid straight line which is almost identical with the dashed straight line is the regression line with a coefficient of determination equal to 0.999.

The model with the slope -1 describes the beta activity decrease of this sample also very well. Whereas the model with the slope -1.2 does not.

The activity of the sample taken on 12 December 1961 is also inversely proportional to the first power of time (see Fig. 1) assuming that the date of the last explosions was used as origin date.

Discussion

At a long distance from a nuclear event, the total activity of the mixture of independent non-chain radionuclides is, in general, the sum of exponential terms exp($-\ x_i\ t$)

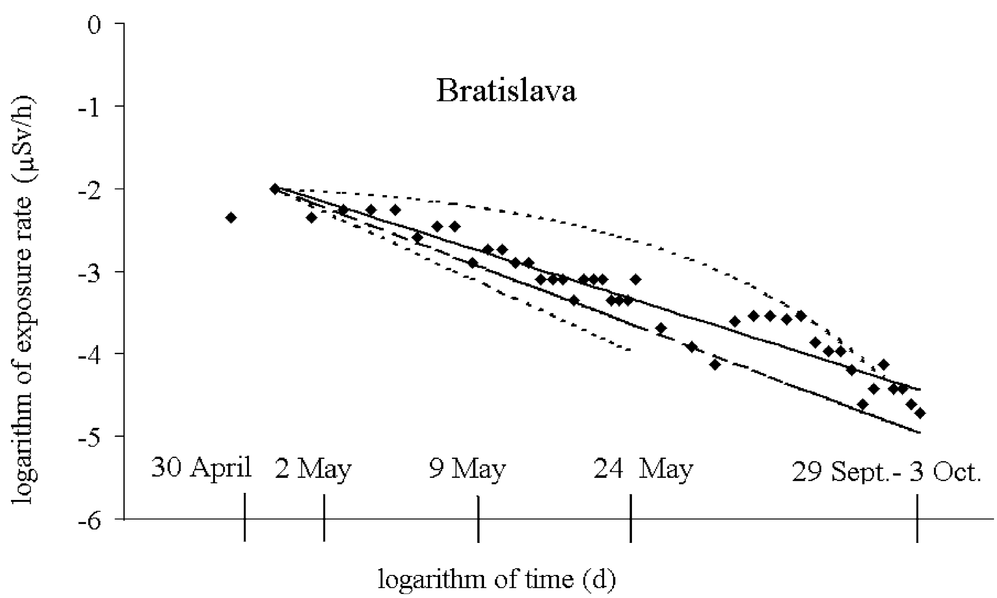


Fig. 6 Decrease of exposure rate along with time (log–log scale) in Bratislava after the Chernobyl accident (see description of *lines* in Fig. 2)

Table 2 Results of the linear regression

Location	Date (t_1)	Date (t_2)	Maximum[a] (μSv/h)	Slope ($-\alpha$)	Lower limit[b]	Upper limit[c]	Natural background (μSv/h)	r^2
Prague	1.5	5.9	0.29	−1.06 (yes)	−1.12	−1.00	0.12	0.97
České Bud.	1.5	1.8	0.51	−0.88 (yes)	−1.03	−0.72	0.1	0.81
Ústí nad L	7.5	17.5	0.11	−1.76 (yes)	−3.78	0.27	0.09	0.30
Hradec Kr.	30.4	20.6	0.37	−1.05 (yes)	−1.11	−0.99	0.11	0.98
Ostrava	30.4	18.7	0.82	−1.11 (yes)	−1.28	−0.94	0.12	0.85
Brno	1.5	25.7	0.26	−1.05 (yes)	−1.13	−0.98	0.105	0.96
Bratislava	1.5	3.10	0.14	−0.83 (no)	−0.91	−0.76	0.095	0.92
Banská B.	1.5	5.9	0.38	−0.98 (yes)	−1.05	−0.91	0.12	0.95
Košice	9.5	24.5	0.04	−1.03 (yes)	−1.28	−0.78	0.1	0.85
Czechosl. I.	1.5	22.8	0.35	−0.96 (yes)	−1.03	−0.87		0.94
Czechosl.II[d].	1.5	22.8	0.32	−1.01 (yes)	−1.06	−0.97		0.98

[a]Maximum calculated after the natural background subtraction
[b]Lower limit of the slope $-\alpha$ (95% confidence level)
[c]Upper limit of the slope $-\alpha$ (95% confidence level)
[d]Set of geometric means calculated only from seven locations (without Usti nad Labem and Kosice). The attribute "yes" means the hypothesis $-\alpha=-1$ was not rejected (see next two columns), the attribute "no" means it was rejected (95% confidence level [CI]). The arithmetic mean calculated from the first nine slopes is equal to −1.08 with SD = −0.25

with different decay constants x_i and with different weights in the entire sum (i.e. with different constants $C_o(x_i)$ independent of the time, where $C_o(x_i)$ is a concentration of specific radionuclides for $t=0$). The relationship of Eq. 3 is valid and the total activity may be a linear—but not a concave—function of time in a log–log scale (see dotted curve in Figs. 2–7).

$$A(t) = \quad \Sigma\, C_o(x_i)\, x_i \exp(-x_i\, t) \rightarrow A(1)\, t^{-\alpha} \tag{3}$$

Equation 3 is not valid in the following two cases:

1. As regards natural radioactivity, the first decay product is in secular equilibrium with the other radioactive isotopes (i.e., x_1 is very small) and the total activity is approximately constant with time.
2. The second exception of the relationship Eq. 3 immediately occurs, when the activity of a partial decay chain depends on the processes in other decay chains (parameters $C_o(x_i)$ are not invariables and $f(x)$ cannot be used). For this case, Way and Wigner [1] assumed the index α in Eq. 1 to be 6/5.

Unlike the situation immediately after a nuclear explosion, the quantity of a certain nuclide at a long distance from the nuclear explosion site is not affected by the decay of other (non-chain) isotopes. In fact, more than one decay product of a particular decay chain in which those products are independent of each other can be present in the mixture, but such a group of nuclides—i.e., primarily a parent–daughter pair—can be

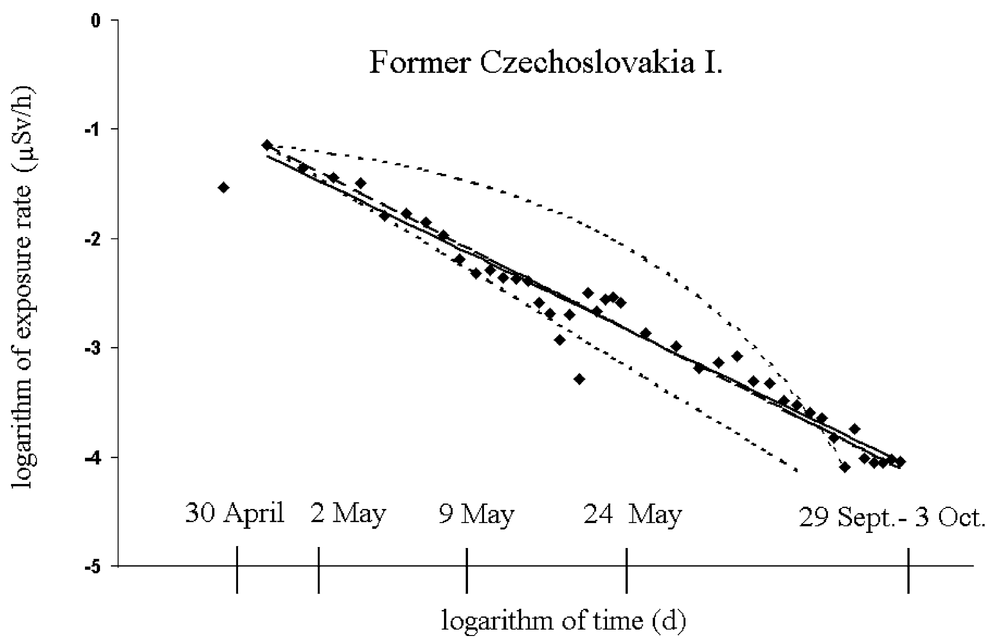
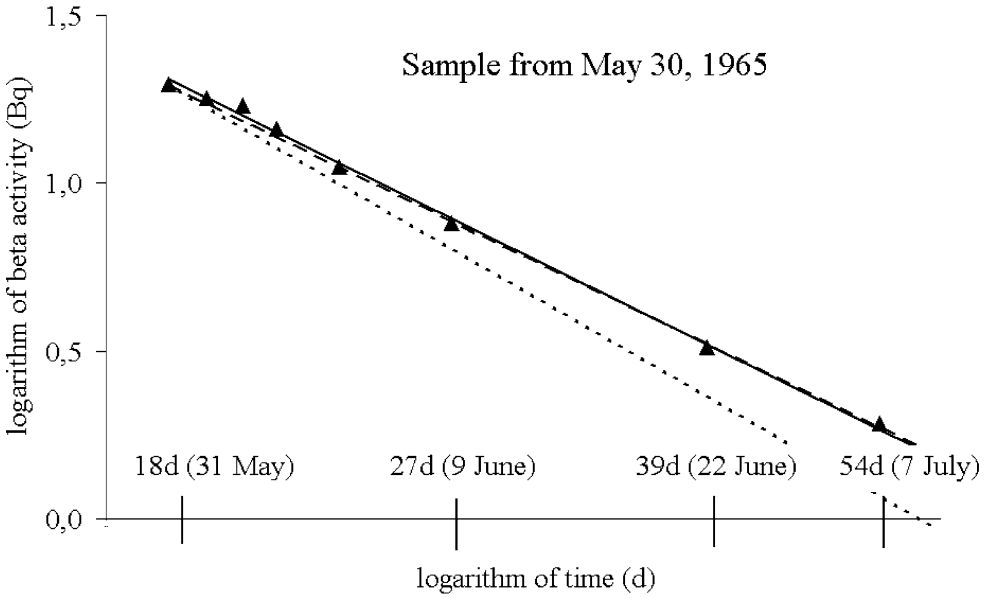


Fig. 7 Decrease of exposure rate along with time (log–log scale) in the former Czechoslovakia after the Chernobyl accident (see description of *lines* in Fig. 2)

Fig. 8 Beta activity of atmospheric fallout sample taken on 30 May 1965 versus time in log–log scale. The origin of time axis is known (14 May 1965). The slope is equal to −1.01 with 95% CI, −1.05 to −0.98; $r^2 = 0.999$. The slope of the *dashed straight line* is precisely equal to −1, and the slope of the *dotted straight line* is equal to −1.2. Both these *straight lines* fit the maximum precisely. The *solid straight line* is the regression line

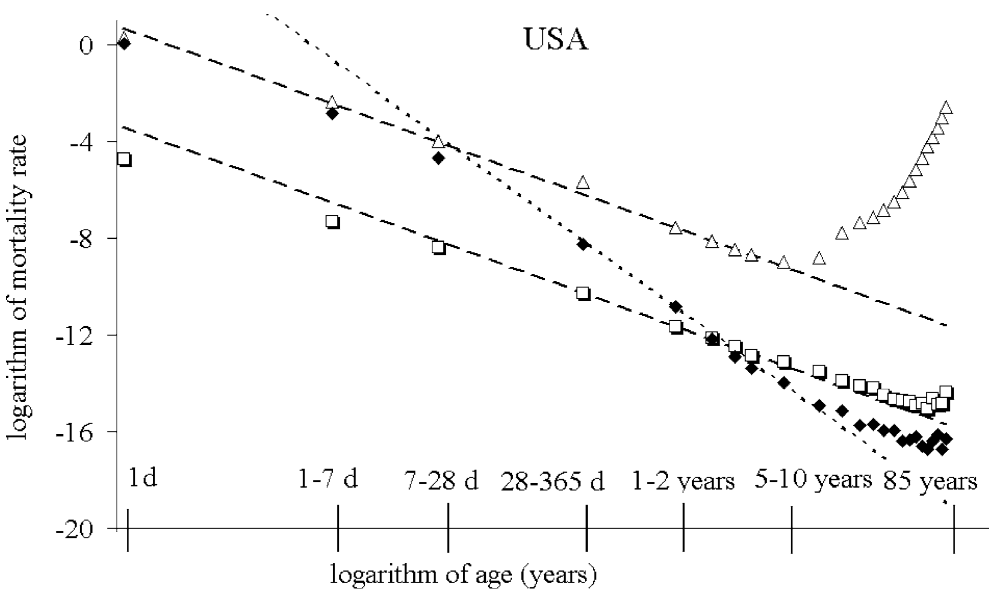


described by one exponential decay term that decreases with time (e.g., ^{140}Ba–^{140}La). The composition of a mixture at long distances from the site is influenced by many external factors, such as meteorological variation, aerosol size, type of landscape, physico-chemical quality of radionuclides. Let f_{actual} $(x;\ t_1)$ be the frequency function of decay constant, x, that describes the existent composition of the mixture at long distances at time t_1 (one portion of the radionuclide with a decay constant equal to x or a quantity $C_o(x_i)$). The parameter t_1 is the period from the moment of explosion/accident to the moment of fallout (e.g., $t_1 = 5.5$ day for 1 May 1986, or $t_1 = 16$ day for 30 May 1965; t_1 is unknown for 2 December 1961). Let f_{actual} $(x;\ t_1)$ be affected by many factors and one of them, radioactive disintegration, be dominating, and separated from the others, if the age of fallout is known. For example, the half-life of ^{137}Cs is about 11,000 days, whereas the half-life of ^{131}I is only 8 days. Hence, the quantity of ^{131}I in the mixture at time t_1 is more affected by radioactive decay. The effect of radioactive decay on the mixture composition within the period $t_1$can be eliminated if the true frequency function f_{actual} $(x;\ t_1)$ is replaced by a virtual frequency function f_{v} (x) at time $t = 0$. The virtual frequency function f_{v} (x) can be defined as:

$$f_{\mathrm{v}}(x) = f_{\mathrm{actual}}(x; t_1)\exp(+t_1 x) \tag{4}$$

The frequency function f_{v} (x) is virtual, because the true composition differs at $t = 0$. The result, f_{v} (x), is of all factors that affect the composition of the mixture during the period t_1 with the exception of the radioactive decay. This virtual mixture composition is due to the

Fig. 9 Three examples of the decrease of mortality rate after birth. The slope of the *dotted straight line* is −2 and the slope of the *dashed lines* is −1 (they precisely fit the point of the age category 1–2 years)



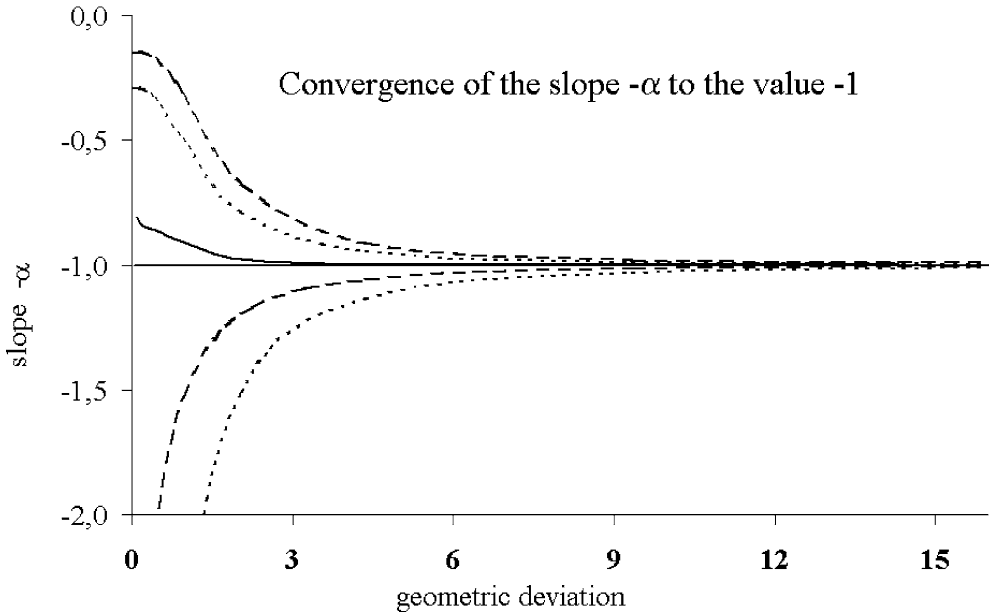


Fig. 10 Convergence of the slope $-\alpha$ to the value of -1 with increasing value of the geometric deviation. The slope $-\alpha$ is equal to -1 if $f(x)$ is the frequency function of log-normal distribution with a wide geometric deviation

fact that different nuclides have different probabilities to achieve a target, and the frequency function $f_v(x)$ describes this aspect (e.g., some inert gases could disappear and their values of $f(x)$ could be zero).

The total activity within the period t (which is counted from the moment of the explosion) is the sum of particular activities $xf_v(x)\exp(-xt)$ of all nuclides, and the following equation can be applied:

$$A(t) \equiv -\frac{\partial N(t)}{\partial t} = \text{const.} \int_{x_{\min}}^{x_{\max}} x\, f_v(x)\ e^{-xt}\, dx \rightarrow \frac{A(1)}{t^{\alpha}} \quad \alpha > 0 \text{ for } t > t_1 \tag{5}$$

where $x_{\min}$ and $x_{\max}$ are the minimum and maximum decay constants in the mixture at the moment of fallout (or at the moment when the measurement of exposure rate has started). Under the assumption of $x_{\min}=0$ and $x_{\max}=\infty$, $A(t)$ is the Laplace transformation of the product $xf_v(x)$. This mathematical formalism is identical with that of the theory of congenital individual risks [12, 13]. The total activity of a mixture of fission products and the mortality rate of a heterogeneous population could be described using the same mathematical formalism (Eq. 5). This theory may explain the linear decrease in the log–log scale and it agrees with the calculation of the slope $-\alpha$ in Eq. 1. Thus, the congenital individual risk in a human population corresponds with the decay constant in the mixture of fission products. Figure 9 presents three examples of postnatal mortality rate decrease with age. Both the dotted straight line with the slope -2 and the dashed lines with the slope -1 are exactly intercepting the point of age category 1–2 years. Both the mortality rate from "All diseases" (all deaths except accidents) and the mortality rate from "Spina bifida and hydrocephalus" decrease with the first power of age. On the other hand, the mortality rate in the category "Certain conditions originating in the perinatal period" decreases with the second power of age ($-\alpha=-2$) [12–14].

In compliance with the theory of congenital individual risks, the slope $-\alpha$ depends on the distribution type of congenital individual risks and on the distribution types of decay constants x, respectively. The theoretical value α does not depend on values of the particular distribution parameters (if x is normally or log-normally distributed in the wide range of these parameters). The slope $-\alpha$ is equal to -2 if the frequency function $f(x)$ is that of normal distribution with some variety or if the frequency function $f(x)$ is invariable (due to uniform distribution [14]). The slope $-\alpha$ is equal to -1, if $f(x)$ is a frequency function of log-normal distribution with notable geometric deviation or if the frequency function can be approximated by $f(x)\approx\text{const}/x$. Figure 10 shows the convergence of the slope $-\alpha$ to the value of -1 with an increasing value of geometric deviation for six values of geometric mean. Similarly, the slope of the integral in Eq. 5 converges to -2, if x is normally distributed [14]).

In general, Eq. 6 is valid:

$$\frac{A}{t^{\alpha}} + \frac{B}{t^{\alpha}} = \frac{A+B}{t^{\alpha}} \tag{6}$$

and, consequently, the relationship of Eq. 1, for one value of the parameter α, is additive. The activity within a particular period, t, is also an additive quantity (activities from two sources can be added), and the convergence condition of the slope $-\alpha$ to -1 could be more generalized. The set of sources of the total activity may be divided into partial subsets. The satisfactory condition is the fact that the decay constants are approximately log-normally distributed with a notable geometric deviation in the partial sources or that the formula $f(x) \approx \text{const}/x$ is valid (however, not necessarily fulfilled for all sources together). The territory after fallout can possibly be divided into more sections. If the decay constants, x, are log-normally distributed with a great deviation in the partial sections, then the slope $-\alpha$ is equal to -1 in all sections. Because Eq. 1 is also additive (for one value of the parameter α), the slope $-\alpha$ on the whole territory could be -1. The activity sources can be divided not only into the territories, but into any quantities with the exception of time. The mixture could be divided into subsets in line with the chemical types of the radionuclides (at the same time, the range of x has to be wide in these subsets).

Conclusions

The results presented here show that the total activity at long distances from a source is inversely proportional to time, if the moment of a nuclear explosion or the moment of a nuclear accident is used as time origin. The model with the power 1.2 is too optimistic and it does not describe the data.

The changes of the amount of a specific nuclide do not depend upon the other nuclides and their decay constants for the fallout at a long distance and, consequently, the function $f(x;t=0)$ could be used. Such a function is independent of the time, and the mixture activity is the sum of exponential terms. It can be assumed that a great number of "external" factors are manifested. If the decay constants are approximately log-normally distributed (its envelop could be approached by the frequency function of the log-normal distribution or by the function $f(x) \approx \text{const}/x$), then the theoretical slope $-\alpha$ is equal to -1.

The estimate of cumulative exposure rate is simple if the relationship between exposure rate and time contains only two parameters (the linear relationship in the log–log scale). If the parameter α in Eq. 1 is known in advance, the cumulative exposure rate can be estimated based on a single measurement. This can be applied if the time of accident or explosion is known and if the composition of fission products mixture depends only on a radioactive decay for $t > t_1$. These conditions were fulfilled for the majority of all measurements in former Czechoslovakia after the Chernobyl accident, and they were automatically fulfilled for the fallout sample taken in Bratislava in 1965. The slope $-\alpha$ is equal to -1 in both cases. This evidence could be explained as a consequence of a log-normal distribution of decay constants in the partial sources or as a consequence of the formula $f(x) \approx \text{const}/x$.

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