# Ecological Risk Assessment for the Terrestrial Ecosystem under Chronic Radioactive Pollution

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**ABSTRACT:** A methodology of ecological risk assessment for the terrestrial ecosystem under chronic radioactive pollution of a biotope near a regional radioactive waste storage has been developed in terms of the critical environmental loads analyzed. It consists of five stages: determination of effect indicators and assessment of their values; establishment of reference species and indices; assessment and analysis of critical loads by plotting "dose-effect" dependencies; ecological risk assessment from critical loads versus permissible values; plotting of risk functions to calculate the expected adverse alterations in the ecosystem. Based on the results obtained the storage risk for a terrestrial ecosystem is considered to be inadmissible and this implies unstable conditions in the territory in the nearest future. The calculations and mapping have shown that in the territory studied the area with excess critical loads is 48% for CFU and 61% for <sup>90</sup>Sr accumulation coefficient. The analyzed risk functions give evidence of highly probable negative alterations in the tested ecosystem: 85% for CFU and 99% for <sup>90</sup>Sr accumulation coefficient.

Key words: Radioactive waste storage, Sr-90, Ecological risk

# **INTRODUCTION**

Much attention is being given to optimal humanenvironment relationships under the conditions of everincreasing anthropogenic impact on the biosphere. Thus, the developed sanitary-hygienic principles of rationing are based on an anthropocentric approach which considers a human impact of technogenic factors and is aimed at limiting pollutant intake. The anthropocentric principle of rationing was formulated by the International Commission for Radiological Protection (ICRP) in 1970 and confirmed in 1990 (ICRP, 1977; ICRP, 1991). However in 2007 ICRP (ICRP 103, 2007) had stated that in developing the strategy of environmental radiation safety, it is necessary to give the evidence for protecting not only people and but also living organisms; it reflects the ecocentric principle of radiation factor rationing. Therefore many recent papers have developed the ecological approaches to rationing of radiation effects on ecosystems (Udalova et al., 2010; Geras'kin et al., 2009; Spirin et al., 2009; Biya et al., 2003, Ecological Risk Assessment, 2007). At the same time, despite numerous studies a generally accepted concept of environmental load rationing as well as a regulatory and methodical framework are not available yet and even a uniform conceptual apparatus

is not developed for this field of knowledge. It should be noted that there are two approaches to the environmental pollution rationing. On the one hand, it is possible to normalize the pollutant content in environmental objects, and on the other, the degree of environmental transformation as a result of pollution. Numerous studies (Lavrentyeva, 2013; Kolesnikov et al., 2001; Demidova, 2007; Moiseenko, 1999; Ovchinnikova, Vasil'evskaya, 2003) have shown that it is advisable to perform rationing not from pollutant content in the environment but from ecosystem response to this pollution. Such an approach allows one to estimate environmental pollution not from the critical concentration of technogenic pollutants but from variations in the expected recorded ecosystem parameters. Thus, the ecocentric principle of rationing represents a permissible load or rationing of the technological environmental impact within the limits not resulting in ecosystem self-regulation disruption. One of the indices of ecosystem resistance to anthropogenic impacts, including radiation, is the critical load value, i.e. the indicator of ecosystem sensitivity which governs the maximum permissible input calculated for different pollutants (Vorobejchik, 2004; Ovchinnikova, Vasil'evskaya, 2003; Lavrentyeva et al., 2013; Bashkin, 2006; Posch, 2007; Priputina, 2006).

In the light of ecological rationing, the final stage of assessing critical loads for ecosystems is the ecological risk. Now the methods of ecological regulation of a radiation factor from risk assessment are developed poorly. The paper attempts to assess and forecast the terrestrial ecosystem conditions under chronic ionizing radiation by calculating the critical loads.

The paper is aimed at developing a methodology to assess the ecological risk for a terrestrial ecosystem under chronic radioactive pollution in a biotope of a regional radioactive waste storage.

The following problems have been stated:

- to determine experimentally the ecosystem impact indicators and to assess their values;
- to establish experimentally the reference species and indices;
- to analyze the critical loads by plotting "dose-effect" dependencies;
- to assess the ecological risk from critical loads.

## MATERIALS & METHODS

The territory near a regional radioactive waste storage created in 1954 in Obninsk has been chosen as a test object. The required area is 0.54 ha. The area of interest lies within the Smolensk-Moscow landscape province in the central part of the river Protva basin and in the lower part of its terrace slope above the flood-plain. The distance to a river is 1000-1200 m. The storage territory has four trench-type tanks for solid radioactive waste and a reinforced concrete tank for liquid radioactive waste. According to the Radiological Safety and Environment Protection Department (FSUE SRC A.I.Leipunsky Institute of Physics and Power Engineering) the specific activity of 90Sr from 1998 to 1999 had increased from tenths fractions to 109 Bq/l in the wells studied; later this fact was proved by a radionuclide leakage from the storage-tank 4 as a result of unsealing and its flooding by surface and ground water (Vasiljeva, 2007). As a result, the massive radioactive source has formed in the geological environments which can be considered as uncontrolled one; in return, radiation monitoring in the territory of a cross-body ecosystem has been increased. Biotope monitoring of a radioactive waste storage makes clear that the radioecological situation in this territory is stipulated by technogenic 90Sr found in soil, ground water and biota (Kozjmin et al., 2008).

Terrestrial mollusks of a shrubby Snail type (*Bradybaena fruticum*) were chosen as reference species due to their activity to accumulate <sup>90</sup>Sr in shells and the number of colony–forming soil units (CFU) as

reference indices. The number of CFU was determined by inoculation of solid medium; this method is commonly used in soil microbiology. The cells per 1 g air dry soil were calculated from the following formula:

## $CFU = n \times 50000$

where n-number of copies, 50 000 is the coefficient considering a 10 000 dilution and the volume of suspension taken for analysis, i.e. 0.2 ml (Pavlova, 2008; Khaziev, 1990).

Soil and mollusk samples have been collected at most representative sites identified in the previous studies (Fig.1). To assess <sup>90</sup>Sr content in the samples collected, radiochemical separation was used with further radionuclide activity measurements by a "BETA-01C scintillation beta-ray spectrometer according to a standard procedure of <sup>90</sup>Sr content assessment from beta-radiation of its daughter radionuclide <sup>90</sup> $\gamma$  (Kuznetsov *et al.*, 1985).

Ecological risk was calculated from analyzed critical loads using a "dose-effect" dependence. Statistical data processing was realized with Excell 2007 (MicrosoftInc., 2006) and R software programs (RDCT, 2019). The software R was also used for GIS creation.

#### **RESULTS & DISCUSSION**

The method of ecological risk assessment from critical loads on a natural ecosystem exposed to technogenic effects comprises five stages. The first stage consists in determination of effect indicators and assessment of their values. Reference species and indices are identified at the second stage. The third stage involves estimation and analysis of critical loads by plotting "dose-effect" dependencies. Ecological risk assessment from critical loads and its comparison with permissible values are realized at the fourth stage. The fifth stage is aimed at plotting and analyzing risk functions to estimate the expected adverse changes in an ecosystem. Radiation and chemical monitoring of the storage territory has been carried out to determine the degree of biotope pollution. The specific activity of natural (226Ra, 232Th, 40K) and technogenic (90Sr, 137Cs) radionuclides in soil samples collected in the storage territory and cross-border regions has shown that artificial radionuclides make a major contribution to the radioecological situation (Table 1).

The specific activity of technogenic radionuclides exceeds the background values up to 64 times as much as for <sup>137</sup>Cs and 274 times for <sup>90</sup>Sr. Non-uniform <sup>90</sup>Sr radionuclide pollution of soil cover arises from the surface and groundwater runoff directed from sites 1, 1a, 1b to sites 2d, 2c and then to sites 7b, 6, 6a and later to sites 10b, 10d and 10e. Thus, <sup>90</sup>Sr radionuclide activity

Sampling	Radionuclide specific activity, Bq/kg				
site	<sup>226</sup> Ra	<sup>232</sup> Th	<sup>40</sup> K	<sup>137</sup> Cs	<sup>90</sup> Sr
1	$39 \pm 11,7$	$15 \pm 4,5$	$334 \pm 100,2$	28,4±8,5	$299 \pm 89,7$
1a	$70\pm21$	$19 \pm 5,7$	375 ± 112,5	$84,3 \pm 25,3$	$341 \pm 102,3$
1b	52 ± 15,6	$26 \pm 7,8$	$370 \pm 111$	$44 \pm 13,2$	$216,3 \pm 64,9$
2	$60\pm18$	$17,4 \pm 5,2$	$327 \pm 98,1$	$115 \pm 34,5$	$28 \pm 8,4$
2a	$57 \pm 17,1$	$27,4 \pm 8,2$	$350 \pm 105$	$15 \pm 4,5$	$55,7 \pm 16,7$
2b	$79 \pm 23,7$	$45 \pm 13,5$	546 ± 163,8	$2 \pm 0,6$	$170 \pm 51$
2c	$26 \pm 7,8$	$16 \pm 4,8$	$297 \pm 89,1$	$8,2 \pm 2,5$	131,2±39,4
2d	$57 \pm 17,1$	$17 \pm 5,1$	$352 \pm 105,6$	$28 \pm 8,4$	$381,9 \pm 114,6$
3	53 ± 15,9	$20\pm 6$	$334 \pm 100,2$	$5,2 \pm 1,6$	56,4±16,9
4	$33 \pm 9,9$	$13,5 \pm 4,1$	$293 \pm 87,9$	$5,9 \pm 1,8$	$26 \pm 7,8$
6	$37,5 \pm 11$	$32 \pm 9,6$	$407,5 \pm 122,3$	$14,5 \pm 4,4$	$807,3 \pm 242,2$
6a	$68 \pm 20,4$	$27 \pm 8,1$	$245 \pm 73,5$	$38 \pm 11,4$	$5203 \pm 1561$
7b	$44 \pm 13,2$	$22 \pm 6,6$	$364 \pm 109,2$	$16 \pm 4,8$	$62 \pm 18,6$
7в	$95 \pm 28,5$	$14 \pm 4,2$	$316 \pm 94,8$	$33 \pm 9,9$	593±177,9
8	$42 \pm 12,\!6$	$16 \pm 4,8$	$346 \pm 103,8$	$27 \pm 8,1$	$17,6 \pm 5,3$
8a	$42 \pm 12,6$	$21 \pm 6,3$	$367 \pm 110,1$	$25 \pm 7,5$	$21,3 \pm 6,4$
8b	$54 \pm 16,2$	$23 \pm 6,9$	$352 \pm 105,6$	$23 \pm 6,9$	$22 \pm 6,6$
9	51 ± 15,3	$22 \pm 6,6$	$323 \pm 96,9$	$8,5 \pm 2,6$	47±14,1
10	$74 \pm 22,2$	$24 \pm 7,2$	$402 \pm 120,\!6$	$15 \pm 4,5$	497±149,1
10a	$27 \pm 8,1$	$10 \pm 3$	$305 \pm 91,5$	3,6±1,1	$2233 \pm 670$
10b	$55 \pm 16,5$	46± 13,8	$429 \pm 128,7$	$5,0 \pm 1,5$	1108,8 ± 332,6
10c	46 ± 13,8	$19 \pm 5,7$	333±99,9	$2,25 \pm 0,7$	$178,9 \pm 53,7$
10d	$44 \pm 13,2$	$24 \pm 7,2$	$315 \pm 94,5$	$192 \pm 57,6$	$1058,5 \pm 317,6$
10e	44 ± 13,2	44±13,2	$350 \pm 105$	$17,6 \pm 5,3$	$4467 \pm 1340$
control	$37 \pm 11,1$	$25 \pm 7,5$	$325 \pm 97,5$	$3 \pm 0,9$	$19 \pm 5,7$

Table 1. Radionuclide specific activity at sampling sites in soil



Fig. 1. Scheme of sampling sites

is primarily concentrated in the waterlogged low in the storage bottom exteriorly where the main water flow is directed to. The distribution of <sup>137</sup>Cs is of another nature. A slightly enhanced activity of this radionuclide is detected at sites 1a and 1b located near the storage capacity for solid radioactive waste; this storage capacity has no extra barriers preventing <sup>137</sup>Cs leakage outward. However, further radionuclide distribution over the storage territory does not occur, therefore this radionuclide is of no serious hazard to the environment. In addition, the specific activity of natural radionuclides is compared with the background ones, therefore their contribution to radioactive pollution is insignificant.

The concentration of heavy metals Mn, Zn, Ni, Cu, Cr, Co, Cd, Pb, Fe in the samples studied does not exceed the MPC value, therefore chemical pollution is of no concern in terms of priority pollution.

Identification of reference species and indices most properly reflecting the degree and the character of pollution in a studied territory is the next stage to assess a biotope of the radioactive waste storage.

According to the method of reference fauna and flora, proposed in the ICRP Publication 91 "Main assessment principles of ionizing radiation effects on living organisms except human body" (ICRP Publication 91, 2004), the full body of information is necessary on several species typical of the territory studied. It is achieved by collecting the available data as well as by using these species as objects for the subsequent studies to identify and to study their reaction to environmental loads in more detail. To assess the state of a radioactive waste storage under chronic <sup>90</sup> Sr pollution, the following representatives of soil microbiota have been chosen as reference species: the number of colony-forming soil units (CFU), abundance of bacteria Azotobacter as well as catalase, dehydrogenase, urease and invertase soil activity. Such a choice is substantiated by specific features of the low organisms under chronic pollution of territories (Iljin, 2000; Nikitina, 1991; Tsipriyan et al., 1993; Reonnpagel K et al., 1998; Pavlova, 2008). Owing to their small size and relatively large contact surface with the ambient medium, soil microorganisms quickly respond to pollution and this affects all the sides of their vital functions: growth, metabolic and regulatory activity, chemical element accumulation. These organisms are rather sensitive and illustrative because the effect of any forcing can be retraced in tens and even hundreds of microorganism generations due to their high rate of growth and reproduction. The analyzed dependencies of bacteria abundance and fermentative soil activity on the aggregative radioactivity index (Fig.2) have demonstrated that the dimensionless aggregative radioactivity index is the sum of studied-to-reference-soil ratios of radionuclide activity and it allows all the expected radioactive sources to be considered including background radiation from natural radionuclides:

$$I = \sum \frac{A_i}{A_{test}^i}$$

where I is the dimensionless aggregative index, A is the specific activity of the ith radionuclide in soil, Ai<sub>test</sub> is the specific activity of the i<sup>th</sup> radionuclide in soil at the test site. Variations in fermentative activity and the number of bacteria Azotobacter as compared to the aggregative index values are not pronounced, however, an exponential dependence is found for the CFU number. It should be noted that unlike CFU, the values of which differ from each other 1.5 - 48 fold, the fermentative activity and the abundance of bacteria Azotobacter vary only within a particular value. It is likely to be associated with rather high soil pollution that does not make possible the quantitative assessment of pollutant effects on ferment activity and the abundance of bacteria Azotobacter. Therefore the CFU index which adequately responds to forcing even under acute soil pollution should be used as a reference species.

The terrestrial mollusk shrubby Snail (Bradybaena fruticum) has been chosen as a reference species; it turned out to be rather sensitive to radioactive pollution in the territory studied. It should be noted that freshwater and terrestrial mollusks are being profitably employed as testobjects of ecosystem 90 Sr pollution for a long time. It is shown that practically the whole 90Sr content is concentrated in mollusk shells and its activity is governed primarily by the level of territory pollution and does not undergo systematic long-term, seasonal and taxonomical changes. It is also worthy of note that the choice of similar reference species is supported by other researchers (Gudkov et al., 2005; Frantsevich et al., 1995; Vasiljeva, 2007; Gudkov et al., 2009; Polikarpov, Tzytzugina, 1995; Mirzoeva et al., 2009).

Thus, it is assumed that terrestrial mollusks (shrubby Snail) and their response (<sup>90</sup>Sr accumulation lowing in shells) should be considered as reference species and the number of colony-forming units as a reference index for the terrestrial ecosystem polluted by radionuclides. Critical radionuclide loads on the reference species and indices have to be determined for assessing the ecological risk. In this case by the critical load we shall basically mean such radionuclide ingress into the ecosystem which does not cause

irreversible alterations in its biogechemical structure, biodiversity and productivity during a long period. A method has been applied to reveal the critical loads by plotting "dose-effect" dependencies and to analyze them (Vorobejchik, 2004; Reva *et al.*, 2011; Bashkin, 1998). To assess the storage impact on soil biota and terrestrial mollusks from experimental data, the dose dependences with respect to a load gradient have been plotted. In this case the maximum permissible load is the critical point on a dose-effect curve which connects the input loads and the soil biota response. The input loads are considered as radionuclide effects and the response of soil biota and mollusks as a reduction in the number of soil CFU and variations in the coefficient of radioactive strontium accumulation in shells, respectively.

For the reference index (soil CFU number), the following response versus radionuclide specific activity dependence has been plotted:

$$CFU = 2.696 \times I^{-0.657}$$

where *CFU* is the number of colony-forming units, *I* is the aggregation index of radioactivity (Fig. 2A).

The obtained dose curve denotes the dependence between the aggregation index and the CFU number. The critical load values are shown by dashed lines, the CFU values are multiplied by  $10^{-6}$  for convenience.



Fig. 2. Soil microbiota versus radioactivity aggregation index diagrams

The CFU number decreases with increased index values. Thus, the amount of CFU at all points with the increased background radionuclide concentration is by 2.6-61.7 times lower as compared to the reference soil samples. It is a nonlinear dependence and has a distinct power decrease in CFU values with the increased aggregation index.

This power dependence has been analyzed to find the site with such an aggregation index value at which a 10-fold decrease in the CFU number (critical load) takes place; this analysis allowed one to obtain the critical aggregation index value (20) corresponding to the critical CFU value, i.e.  $0.38 \times 10^{-6}$  (in this case, the higher the CFU value, the better is for ecosystem) (Evseeva *et al.*, 2012). Excess critical index values, and hence, excess radiation effects on soil biota cause an irreversible decrease in the colony-forming units of soil. A power dependence of the terrestrial mollusks response to radioactive pollution is plotted as well:

 $AC = 314.627 \times I^{-1.502}$ ,

where AC is the accumulation coefficient, I is the aggregation index of radioactivity (Fig.3).

The dose dependence has been analyzed to find the site with such an aggregation index value at which a 10-fold decrease in the accumulation coefficient of <sup>90</sup>Sr (critical load) takes place; this analysis allowed one to obtain the critical aggregation index value (18.3) corresponding to the critical coefficient value (3.99) of radionuclide accumulation (in this case, the higher the AC values, the better is for ecosystem) (Evseeva *et al.*, 2012). The excess critical value of <sup>90</sup>Sr accumulation coefficient causes alterations in the lifesustaining activity of mollusks, decreased activity of their nutrition and, hence, changes in the need for shell building materials and <sup>90</sup>Sr is proved to be one of them.

To characterize the ecological risk from critical loads, GIS maps for the ecosystem studied have been plotted; these maps are used for determining excess critical loads (Bashkin et al., 1998). A space satellite (Google Company) image with a co-ordinate reference has been used as the GIS basis. All data on the ecosystem load as well the critical load are plotted on the digitized map using R program (R development, 2010). The total territory area according to chosen coordinates is 0.54 ha. A part (%) of the area with excess critical loads is recognized by the software and results are plotted as contour curves and tone drawing (Figs.4 and 5). "Clean" areas (dark) and those characterized by excess critical loads (light) are separated by contour curves and tone drawing. Contour curves specify an aggregation index, the maximum values of which are typical of the areas shown by light.

A 95% ecosystem protection when the area with excess critical loads does not exceed 5% of the total value (0.54 ha) is taken as the permissible adverse storage impact on the environment. The analyzed plotted GIS maps show that in a studied territory the area characterized by excess critical CFU values is 48%, and the critical AC is 61% that exceeds the permissible value (5%). The fact that results on CFU and terrestrial mollusks are close, demonstrates the reliability of our approach to assessing ecological risk from critical loads regardless of a reference index or species position in a food chain.



Fig. 3. Mollusk shells <sup>90</sup>Sr accumulation coefficients versus radioactivity aggregation index

So, the risk of waste storage effects on a surrounding terrestrial ecosystem should be characterized as inadmissible that highlights the unstable state in the territory studied within the next few years. The evaluated values for areas characterized by excess critical loads allow the expected development of adverse changes in the ecological system to be assessed. Radionuclide distribution functions have been plotted from the excess critical load values; these functions represent a difference between the exposure value and the safe exposure level  $(Y_u)$ :



Fig. 4. GIS map for critical loads on soil biota (CFU)





Note: a dotted line - the end of negative changes in an ecosystem

 $N_p = E_{xp} - Y_y,$ 

where  $N_{\rm p}$  is the excess critical load;  $E_{\rm xp}$  is the exposure value;  $Y_{\rm v}$  is the safe exposure level.

The ecosystem effect is evaluated as a ratio of the areas characterized by excess critical loads to the total area under investigation. Thus, a part by area with the excess critical load is determined. The probability of excess critical loads  $P_i(N_p)>0$  is calculated based on  $N_p$  values.



Fig. 5. GIS map for critical loads on terrestrial mollusks





The studied territory areas are calculated by GIS technologies. The ecological risk function  $(R_x)$  is found for each site and represents the distribution  $R_x = F(M) = P(p_x < M)$ , where P is the probability;  $p_x$  is the random variable characterizing the relative territory area with excess critical loads (M  $(N_x) > 0$ ).

Then, the ecological risk is assessed which is a complex index characterizing both the expected development of adverse changes in the ecological system and their amount and nature. The critical load excess will be found within the [M1; M2],  $(P = P_2 - P_1)$  interval.

For a graphical representation of risk functions it is assumed that in case of pollution below the critical load the excess risk is 0 and above this level it is 1. Risk functions are found by recalculating the intermediate values (Figs.6 and 7).

These risk functions have permitted the probability (risk) of excess critical loads or, in other words, the probability of adverse changes to be assessed for the terrestrial ecosystem under studies. Thus, the probability of adverse changes calculated for soil microorganisms is 85% and for terrestrial mollusks it is 99%. A similar value (probability) of adverse changes confirms the impermissibility of risk at a tested territory and highlights the expected loss of this territory as a typical terrestrial biotic community in the short run.

When assessing the ecological risk, some uncertainties have to be considered in such investigations. First, interspecies extrapolation, i.e. data for radionuclide effect on individual reference species have been extrapolated to the whole ecosystem. Second, in the critical load assessment the form of radionuclides found in soil has not been taken into account at the stage of data collection. Third, the process of radionuclide return into soil as a result of plant die-away and mollusk loss has not been considered. Fourth, the unified dimensionless aggregation indices may be not exactly correct for data averaging.

# CONCLUSION

Environmental load rationing is the key problem for ecological safety support. In the light of environmental load rationing the ecological risk is an instrument for assessing the limiting loads on the environment. Now methods for quantitative risk assessment in the territories under chronic ionizing radiation are not developed yet. This paper has studied the set of regulation principles for such ecosystems with a biotope of a radioactive waste storage as an example and the approach to ecological risk assessment from analyzed dose dependencies. The analysis of critical pollutant loads is chosen as a promising approach. The concept of critical loads is based on the assumption of a certain value, i.e. pollutant emission threshold, the excess of which results in irreversible alterations in the ecosystem functioning. There are no essential adverse effects below this threshold. Ensured pollutant emission below the limiting standard may stipulate the normal development of natural ecological systems. However, if the pollutant content exceeds the safe exposure level, the expected consequences of this excess must be evaluated to mitigate adverse effects for ecosystem safety.

The suggested technology of ecological risk assessment for a terrestrial ecosystem under chronic radioactive pollution from critical loads involves the following five stages: determination of effect indicators and assessment of their values; establishment of reference species and indices; assessment and analysis of critical loads by plotting "dose-effect" dependencies: ecological risk assessment from critical loads versus admissible values; risk function plotting to assess the probability of adverse radioactive effects on ecological systems.

The following reference species have been educed from experimental results for the tested territory: the terrestrial mollusk shrubby Snail (*Bradybaena fruticum*) of fauna representatives and the number of colony-forming units (CFU) of soil microbiota representatives.

The calculations and mapping have shown that in the territory studied the area with excess critical loads is 48% for CFU and 61% for <sup>90</sup>Sr accumulation coefficient. The analyzed risk functions give evidence of highly probable negative alterations in the tested ecosystem: 85% for CFU and 99% for <sup>90</sup>Sr accumulation coefficient.

Hence, the environmental impact of this storage should be characterized as inadmissible and implies unstable conditions and the expected loss of current biocenosis in the studied territory in the near future.

It should be emphasized that a concept of critical loads is being extensively used in the world practice as essential measures for efficient monitoring of anthropogenic effects (Moiseenko, 1999).

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