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Evaluation of Ingestion doses and Hazard Quotients due to Intake of Uranium in tap Drinking Water from Aden Governorate, Yemen

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Abstract: In the current study, Uranium concentrations were measured in tap water samples collected from selected regions in Aden governorate using high purity germanium detector (HPGe). The results of this experimental investigation showed that the Uranium concentration in tap water samples were found to range from $(8.58\pm0.964 \text{ to } 124.12\pm13.94)\mu g/L$ with mean value of $(64.48\pm6.96)\mu g/L$. We found that Uranium concentrations in 8 out of 10 (80%) samples exceeded the WHO provisional guideline value of $30\mu g L^{-1}$. The annual effective doses were estimated for different life stage groups. The highest dose was calculated for teenagers (14-18y) male. From radiological perspective, the mean cancer mortality risk and morbidity risk were found to be 0.89×10^{-4} and 3.8×10^{-4} respectively, which are lower than permissible standard which is 1×10^{-3} . The lifetime average daily dose (LADD) was found to range from $(0.632 \text{ to } 9.14)\mu g/kg/day$ with mean value of $4.56\mu g/kg/day$. The mean value of $1.2\mu g/kg/day$ reported by World Health Organization. The Hazard quotient (HQ) was greater than unity, implying significant potential risk of uranium in tap drinking water due to chemical toxicity. All the recorded values of Uranium are compared with the safe limits recommended for tap drinking water by various health and environmental protection agencies.

Keywords: Uranium Concentrations; Annual Effective Dose; Hazard quotient.

1 Introduction

Uranium is a naturally occurring radioactive element that is commonly present in water. Studies show that the contribution of ingested uranium through food products accounts for 15%, whereas drinking water contributes to 85% of the ingested uranium. Hence, the health risk due to consumption of uranium-containing tap water poses a greater risk compared to other causes [1, 2].

Uranium is one of the most serious contamination concerns because of its radioactivity and heavy-metal toxicity. Uranium and its compounds are highly toxic, which is a threat to human health and ecological balance [3]. Uranium is considered a dangerous mineral due to its ability to affect a living cell, which may lead to mutation or cancer [4].

The solubility of uranium varies according to particular substances, and this solubility determines how easily and effectively the body absorbs it through the lungs and the intestines. At the end of the blood flow, uranium accumulated back into the bones of different organs again, causing many health complications, ranging from most forms of cancer, such as kidney failure, skin disorders, respiratory and other diseases, unknown[5]. There are different possible ways by which uranium can reach the human body either in a direct way by inhaling uraniumbearing dust particles or by drinking water which is polluted by uranium, or in an indirect way from the fertile soil layer via the food chain [6].

To reduce the risks of harmful radiation to workers and members of the public through radiation practices, the dose limit must be adhered to, which is; the upper permissible limit for the radiation dose. Dose level the amount of the radiation dose limit may not be increased [4, 7]. Uranium gets into drinking water when groundwater dissolves minerals that contain uranium. The amount of uranium in well water will vary depending upon its concentration in bedrock. However, even within areas that have bedrock types containing uranium, there is a large degree of variation within relatively small areas. Levels of naturally occurring radiation in water are not likely to be high in shallow wells. High levels of uranium indicate the potential for radon and radium also to be present [8, 9]. The uranium intake from water is equal to the total from other dietary components. As a result, these radionuclides may enter the food chain through irrigation waters and the water supply

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through groundwater wells and surface water streams and rivers [10]. The aims of the present investigations is to determine the concentrations of uranium in tap drinking water and compare the observed concentrations with drinking water quality guidelines/standards, to compute age-dependent annual effective doses

(AEDs) and to determine radiological and chemical toxicity risks to humans due to ingestion of uranium in tap drinking water used by people who live in Aden governorate, Yemen.

2 MATERIALS AND METHODS

2.1 Description of Study Area:

In Aden governorate, the household water is supplied from two sources; one from Beer Nasser region in Lahj governorate and other from Beer Ahmed region in the north side of the Aden governorate. In fact, the study area is located inside Aden Governorate which is located in South of Yemen on the Gulf of den. The location of Aden Governorate has been determined using the Global Positioning System (GPS): Latitude: 12°49'.468"N., Longitude: 44°51'.708"E. The map of studied area is shown in Figure (1). Table (1) shows symbol and location name for the different studied regions (sites) in Aden governorate for tap water samples.



Fig. 1. Illustrates the areas under study.

2.2 Collection and Preparation of the samples:

Ten tap water samples were collected from the water networks in dwellings from different locations in Aden Governorate, Yemen, (1 liter) Standard Polyethylene Marinelli beakers (GA-MA & Associates Inc, USA), were used as a sampling and measuring container. Before use,

© 2024 NSP Natural Sciences Publishing Cor. the containers were washed with dilute hydrochloric acid and rinsed with distilled water. Each beaker was filled up to brim and a tight cap was pressed on so that the air was completely removed from it. The collected water samples were left for an overnight period in polyethylene containers to allow setting of any suspended solid materials and for each samples a clear supernatant was separated decantation. The clear solution was acidified by adding 0.5ml of conc. HNO₃ per liter, to prevent any loss of radium isotopes around the container walls, and to avoid growth of microorganisms [11].

The water samples were then homogenized well by shaking. The final acidity of water samples reaches pH=2. The samples were stored for over 30days to reach secular equilibrium between radium isotopes and their respective daughters before radiometric analysis.

2.3 Methods

2.3.1. Radioactivity analysis and Measurements of Activity Concentration of Uranium

All the samples were measured at the nuclear physics laboratory in atomic energy Authority laboratory, Sana'a, Yemen, using a gamma ray spectrometer. The applied low level background gamma ray spectrometer consists basically of an HPGe-detector the detector was coaxial in shape having relative efficiency 30% with respect to NaI (TI) detector and active volume of 180cm³ fitted with beryllium-end window. The detector had closed-end coaxial Gamma-ray detectors (p-type) made up of high purity germanium (HPGe) in a vertical configuration cooled by liquid nitrogen with the following specifications: resolution (FWHM) ≤ 2.000 keV and ≤ 0.925 keV at 1.33MeV and 122keV, respectively, with a relative efficiency of 35%. The germanium crystal was located within a lead shield for the reduction of the environmental background. The detector is connected to preamplifier, main amplifier, analogue to digital converted (ADC) and multichannel analyzer (8192 Chanel). The system was calibrated for energy using standard point sources (⁶⁰Co, ¹³⁷Cs), and calibrated for efficiency using standard QCYB41 [12, 13].

Every sample was placed in face to face geometry the detector for 10 to 24hour for (²³⁸U) Uranium concentrations measurements. Prior to sampling counting, background were taken normally every week under the same condition of sample measurement. The spectra were analyzed by the computer software program Canberra's Genie2000 Canberra Industries, Inc, USA) for the calculation of natural radioactivity.

The radioactivity concentration of 238 U was determined from the photo peaks of 234 Th

(63.29keV) (which was verified by 235 U measurement using the 163keV line). [12, 13]. The activity concentration of Uranium has been calculated by using the following equation (1) [14]:

$$A = \frac{N(E_{\gamma})}{t_c J_{\gamma}(E_{\gamma}) \cdot \varepsilon(E_{\gamma}) \cdot V}$$
(1)

Where N (E_{γ}) is the number of count in a given peak area corrected for background peaks of a peak at energy E_{γ} , $\epsilon(E_{\gamma})$ the detection efficiency at energy E_{γ} , t_c is the counting life time, I_{γ} (E_{γ}) the number of gammas per disintegration of this nuclide for a transition at energy E_{γ} , and V is the volume of the water measured sample.

2.3.2. Health risk assessment:

• Age-dependent dose assessment:

The annual effective dose for different age groups from ingestion of uranium in water was determined as, [15]:

Ingestion dose
$$(Svy^{-1}) = U_A \times DWI (Ly^{-1}) \times DCF (SvBq^{-1})$$

(2)

Where:

 U_A is the Uranium activity in water (Bq/L), DWI is the agedependent daily water intake (Ly⁻¹) and DCF is the dose conversion factor for specific age groups (SvBq⁻¹) [15]. The water intake rates (Lday⁻¹) taken for infants of (0-6) month and (7-12) month old are 0.7Lday⁻¹ and 0.8Lday⁻¹, respectively. For the children of age group (1-3) year and (4-8) year is 1.3Lday⁻¹ and 1.7Lday⁻¹, respectively. For children (9-13) years, 2.4Lday⁻¹ for male and 2.1Lday⁻¹ for female. For teenagers age group (14-18) year, 3.3Lday⁻¹for male and 2.3Lday⁻¹ for female and above 18year (adults), is taken as 3.7Lday⁻¹ (for male) and 2.7Lday⁻¹ (for female) [16]. During pregnancy and lactation, the water intake rate assumed was 3.0Lday⁻¹ and 3.8Lday⁻¹. The dose conservation factor taken for infants, 1year, 5years, 10 years, 15 years and adults are 1.4×10^{-8} , 1.2×10^{-8} , 8×10^{-8} , 6.8×10^{-8} , 6.7×10^{-8} , and 4.5×10^{-8} SvBq⁻¹[15]. The mass to activity conversion factor used for natural uranium was 0.0248Bqµg⁻¹ [17), [18].

• Radiological Toxicity Risk Assessment:

Radiological toxicity risk is expressed in terms of excess cancer risk (ECR), which was evaluated by multiplying the uranium activity concentration (U_A) (BqL⁻¹) and risk factor (RF) (LBq⁻¹) [19].

$$ECR = U_A \times RF \tag{3}$$

The risk factor (RF) was determined as follows:

$$\mathbf{RF} (\mathbf{LBq}^{-1}) = \mathbf{RC} \times \mathbf{IRW} \times \mathbf{ED}$$

Where:

RC is the uranium risk coefficient (Bq⁻¹), IRW is the water ingestion rate (2Lday⁻¹) [20] and ED is the exposure duration (70years), i.e. $70 \times 365 = 25,550$ days. According to the [1], the mortality and morbidity cancer risk coefficients of 1.13×10^{-9} Bq⁻¹ and 1.73×10^{-9} Bq⁻¹, respectively, have been used for the estimation of cancer mortality risk and cancer morbidity risk of uranium over lifetime consumption of tap water[20; 21].

• Chemical Toxicity Risk Assessment:

The chemical toxicity risk from exposure to uranium is quantified in terms of the lifetime average daily dose

(LADD) and hazard quotient (HQ). LADD is defined as the quantity of uranium ingested per kilogram of body weight per day and was evaluated using the following equation [22; 23]:

$$LAAD = \frac{EPCxIRWxEFxED}{ATxBW}$$
(5)

Where:

EPC is the exposure point concentration (μ g/L), IRW is the water ingestion rate taken as 4.05 liters per day, EF is the exposure frequency taken as 365 days per year, [20, 21], ED is the total exposure duration which is taken as 70 years, AT is the average time (25,550days), BW is the weight for a standard man taken as 55kg [24; 25; 26].

HQ is defined as the ratio of the chronic daily uranium intake to its reference dose (RfD). The HQ was calculated using the following equation:

$$HQ = \frac{LAAD}{RfD} \tag{6}$$

Where:

(4)

RfD is the reference dose. Its value is 1.2µgkg⁻¹day⁻¹ [20]. If HQ is found to be less than unity, then no adverse health effects are expected due to the exposure of uranium.

3 Results and Discussion

3.1. Activity Concentrations of the radionuclides:

Table (1) shows the Uranium concentrations in tap drinking water samples collected from different regions of Aden Governorate, Yemen. From this table, the highest value of Uranium concentration is equal to $(124.12\pm13.94)\mu g/L$ was found in tap drinking water sample of (W10) in Beer Nasser region, while the lowest value of uranium concentration is equal to $(8.583\pm0.964)\mu g/L$ was found in tap drinking water sample (W02) Al-Ma"allah region. The mean value of Uranium concentration in tap drinking water samples is equals to $(64.48\pm6.961)\mu g/L$.

 Table 1: Uranium Concentration and daily uranium intake in tap Drinking Water.

		Coord	linates	Uranium	Uranium	
Sam ple code	Name of the Site	Latitu de (N)	Longit ude (E)	Concentra tion (Bq/L)	Concentra tion (µg/L)	
W1	At-Tawahi	12°46'	44°59'1	2.62±1.70	105.582	
W2	Al- Ma''allah	12°47'	45°00'1	0.213±0.13	8.583	
W3	Crutter	12°46'	45°02'0	2.30±1.49	92.686	
W4	Crutter	12°46'	45°02'4	0.446±0.28	17.973	
W5	Khor- Makaan	12°46'	44°59'1	1.54±0.999	62.059	
W6	Ash Shovish''Uth	12°47'	45°00'1	0.777±0.50	31.312	
W7	Dar-Saad	12°46'	45°02'0	2.32±1.51	93.492	
W8	Al- Mangauna	12°48'	45°02'1	1.01±0.656	40.701	
W9	Beer Ahmed	12°58'	44°69'4	1.07±0.695	43.119	
W10	Beer Nasser	12°52'	44°54'0	3.08±2.07	124.119	

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	Maximum	3.08±2.07	124.119±1
	Minimum	0.213±0.13	8.583±0.96
	Mean	1.60±1.04	64.48±6.96

The WHO reference level for Uranium concentration is equal to 30µgL⁻¹ [20], the UNSCEAR safe limit for Uranium concentration is equal to 9µgL⁻¹ [27], the ICRP safe limit is equal to $1.9\mu gL^{-1}$ [28] and the USEPA safe limit for Uranium concentration is $30\mu gL^{-1}$ [29]. On comparing the Uranium values in tap drinking water with the recommended value of ICRP and UNSCEAR, 90% of the samples seem to have Uranium values higher than the recommended level and 10% of the samples seem to have lower than ICRP and UNSCEAR reference level for Uranium. However, when compared the uranium values in tap drinking water with WHO and USEPA safe limit 80% of the samples seem to have uranium values higher than the recommended level and 20% of the samples seem to have Uranium values lower than the safe limits of WHO and USEPA. In the present investigation, the mean value of Uranium concentration in tap drinking water samples were 34times higher than the recommended value of ICRP (1.9µg/L), and 7times higher than the recommended safe limits by UNSCEAR (9µg/L), and 2times higher than the reference level for Uranium (30µg/L) of WHO and USEPA. It can be seen that the Uranium concentration in the tap drinking water samples varied from location to location. The variation of Uranium concentration in study area is due to presence of underlying rocks, sandstones, siltstones, shale, and limestone in the tap drinking water sources [16]. Uranium concentration in all locations in Aden Governorate is shown in Figure (2).



Fig.2: The Uranium concentration $(\mu g/L)$ in tap drinking. water in Aden Governorate

3.2. Age-dependent annual effective dose (AED)

From Uranium concentration, the corresponding annual effective dose for different age groups has been calculated in Table (2). The value of annual effective dose for different age groups ranged from $0.751 \mu S v y^{-1}$ to $278.09 \mu S v y^{-1}$ with

Subaihi and A. O. Bazohair : Evaluation of Ingestion doses and ... an average value of 73.18±7.28µSvy⁻¹. The large variations in the annual effective dose are due to the wide range of uranium concentrations in the investigated tap drinking water. The WHO guidelines (fourth edition) and the European Union Council Directive [30] prescribed the measurement of reference dose level (RDL) of the AED received from drinking water ingestion at 100µSvy⁻¹. This RDL is 4.2% of the average AED of 2.4mSvy⁻¹ from natural background radiation [26; 20]. The Annual effective dose received due to the ingestion of Uranium in tap drinking water in age group of children and Adults for samples W1 Al-Ma"allah region, W3 Crutter region, W7 Daar-Saad region and W10 Beer Nasser region were higher than the recommended limit suggested by WHO guidelines (fourth edition) and the European Union Council Directive [26; 20]. It is noted that, the doses received due to the ingestion of Uranium in tap drinking water by teenagers (male) are higher than that received by infants and adults compared to other age groups. Thus the age group at risk is teenagers (male) because of their intensive bone growth and action should be taken to restrict their intake [22].

Table 2: Age-dependent annual effective doses (μSvy^{-1}) due to daily consumption of Uranium through tap drinking water in Aden Governorate.

Anzaid effective date (givy*)												
Sample Code	(1-145)	(7-13a)	990	(449)	0-05		0410		(Abera 10)		Prepiercy	Jactation 1
					Mab	female	Mab	Tenals	Male	Inab		
π1	9.35	14.56	1471	135.37	153.93	131149	296.87	366.87	15.14	114.69	127,85	19114
111	1.75	1.189	1.39	16.0	12.51	1635	18,22	13.40	12.78	8.52	10.38	15.11
#3	8.31	612	12.92	112.01	718.13	118.34	207.66	1112.0	337.94	100.00	111,51	141.9
114	1.91	1.89	1.99	21.84	26.25	22.85	41.27	29.66	38.79	19.91	21.87	17.48
113	6.45	633	8.65	- 9.6	91.0	9.11	29.04	90.01	92.18	67.36	3434	54.88
Ť6	3.74	310	4.36	19,84	45.65	38.84	78,15	.8.99	46.65	31.68	37.76	47.89
TT!	8.38	1.55	1500	113.99	196.98	119.27	308.4T	145.99	199.14	101.48	112.75	142.42
773	3.56	407	56	8.8	19.34	51.82	91.19	63.55	0.55	11.10	41.00	62.17
TΓP	3.77	4.51	6.81	\$1.39	62.88	89.81	98.65	4.77	64.18	46.80	52.80	65.87
W19	16.87	11.41	17.9	190.80	338,94	15834	1.816	183.82	381.75	104.72	149.69	188,00
Min.	16.87	11.41	37.39	150.80	-188.94	158.14	278.08	185.82	384.79	134.12	149.69	388,06
Mint.	6.751	1.307	1.10	16.45	12.92	16.85	18.19	13.40	12.78	+.31	10.39	10.11
Man	54.1	6.25	8.43	75.35	19,34	79.64	.138.83	M.%	92.22	67.25	74.73	94.67

3.3 Radiological toxicity risk:

The cancer mortality and morbidity risks as evaluated for the people who consume this water for drinking purposes are presented in Table 3. Mortality indicates the incidence of fatal cancers and morbidity indicates the incidence of total cancers (fatal and non-fatal). The lifetime cancer mortality risk due to intake of uranium from the tap water samples ranged from 0.123×10^{-4} to 1.78×10^{-4} with an average value of 0.889×10^{-4} . The lifetime cancer morbidity risk due to intake of uranium from the tap drinking water samples ranged from 0.188×10^{-4} to 2.72×10^{-4} with an average value of 1.36×10^{-4} . It can be seen that the lifetime cancer mortality risk and lifetime cancer morbidity due to intake of uranium from the tap water samples are lower than the international acceptable limit of 1.0×10^{-3} for radiological risk according to [31]. Therefore, there was no radiological health risk that would lead to having lifetime cancer mortality and lifetime cancer morbidity by

consuming uranium from the tap water in the study area. The average value of the lifetime cancer morbidity risks and lifetime cancer morbidity is lower than the international acceptable limit of 1.0×10^{-3} for radiological risk according to [31]. This confirms that the tap water Aden Governorate is radiologically safe for uranium consumption.

3.4. Chemical toxicity risk:

The chemical toxicity health risks associated with the consumption of uranium in tap drinking water in the study area was done by assessing uranium as a heavy metal, and then compares the uranium mass concentration (μ g/L), LADD and HQ with the international acceptable limit of uranium in drinking water [32]. The mean value of the mass concentration of uranium (ug/L) for the tap water samples 3times higher than the permissible limit of uranium in drinking water which is equal to 20µg/L [20; 29; 33; 34]. The values of LADD from tap water samples varied from (0.632 to 9.14)µg/kg/day with an average value of 4.56µg/kg/day, considering the body weight as 55kg of an adult Yemeni reference man. The average value of LADD for the tap water samples is approximately 4times higher than the accepted international threshold daily intake value of $1.2\mu g/kg/day$ [20]. (See figure 4).

This variation in LADD is due to uneven distribution of Uranium in tap drinking water. The mean value of hazard quotient (HQ) was also found to be 3.8 with range 0.527-7.62 as given in Table (3). RfD recommended by WHO 1.2μ g/kg/day. HQ for 90% samples was >1.0, indicating

risk due to chemical toxicity due to the drinking of tap drinking water according to WHO standards, as shown in Table (3). The variation of Uranium concentration in tap water samples has been reported in Table (1). It has been found that the Uranium concentration is higher in At-Tawahi, Crutter, Khor-Makser, Daar-Saad and Beer Nasser regions as compare to other regions in Aden Governorate.

The migration or mixing of chemicals in tap drinking water may be due certain reasons, These reasons can be anthropogenic factors, such as drainage, irrigation, groundwater pumping, waste or wastewater disposal from industry and other is man-made activities such as depleted uranium from the wars. A comparison of the lifetime average daily dose (LADD) obtained in this study with the reference dose (RfD) of 1.2µg/kg/day as the acceptable level, shows that the chemical toxicity risks due to uranium in water samples were all higher than the reference dose. This therefore confirms that the health risks associated with intake of uranium by the inhabitants in the study area through drinking water from the tap drinking water are mainly due to the chemical toxicity risk. The calculated hazard quotient (HQ) using reference dose (RfD) value of 1.2µg/kg/day showed that HQ values are greater than unity (i.e HQ>1.0) as in figure (6), indicating significant potential risk due to chemical toxicity of uranium in water [35]. The study therefore confirm that human risk due to uranium content in water supplies that will result from ingestion may be attributed to chemical toxicity of uranium as heavy metal rather than radiological risk.

		Chemica	l Toxicity Risk	Radiological Risk		
Sample code	Name of the Site	LADD (µg/kg/day)	HQ Hazard Quotient	cancer Mortality Risk×10 ⁻⁴	cancer Morbidity Risk×10 ⁻⁴	
W1	At-Tawahi	7.77	6.48	1.51	2.32	
W2	Al-Ma''allah	0.632	0.527	0.123	0.188	
W3	Crutter	6. 82	5.69	1.33	2.03	
W4	Crutter	1.32	1.10	0.257	0.394	
W5	Khor-Makser	4.57	3.81	0.889	1.36	
W6	Ash Shaykh''Uthman	2.30	1.92	0.449	0.687	
W7	Dar-Saad	6.88	5.74	1.34	2.05	
W8	Al-Mansoura	2.99	2.49	0.583	0.893	
W9	Beer Ahmed	3.17	2.64	0.618	0.946	
W10	Beer Nasser	9.14	7.62	1.78	2.72	
Maximum		9.14	7.62	1.78	2.72	
Minimum		0.632	0.527	0.123	0.188	
Mean		4.56	3.80	0.889	1.36	

Table 3:Radiological and Chemical Toxicity Risk of Uranium in the Tap Drinking Water Samples.









Fig. 5: Comparison of Lifetime average daily dose due to intake of uranium from the tap water samples with WHO Permissible Standards.



Fig.6: Comparison of Hazard Quotient due to intake of uranium from the tap water samples with Standards.

A total of 10 Tap water samples were collected from the water networks in dwellings from different locations in Aden Governorate, Yemen. The results have shown that, the highest value of Uranium concentration in tap water sample is equal to $(124.12\pm13.94)\mu g/L$ is found in (W10) in Beer Nasser region, while the lowest value of uranium concentration is equal to $(8.583\pm0.964)\mu g/L$ is found in (W02) Al-Ma"allah region with mean value of $(64.48\pm6.96)\mu g/L$.

- The mean value of uranium concentration $(\mu g/L)$ for all water samples was higher than the recommended value of ICRP (1.9µg/L), and higher than the recommended safe limits by UNSCEAR (9µg/L), WHO (2011) provisional guideline level (PGV) of $30\mu gL^{-1}$ and United States EPA ($30\mu g/L$). So there is health effect on the habitants of the study area.
- 40% of the samples have annual effective dose due to intake of uranium through tap drinking water in all age groups higher than the recommended limit except infants and children (1-3y) and 60% of the samples have annual effective dose due to intake of uranium lower than the recommended limit suggested by WHO guidelines (fourth edition) and the European Union Council Directive of 100µSvy⁻¹.The teenagers (male) have received relatively high mean AEDs compared to other age groups in all samples. In general 40% of the samples have AEDs exceed the recommended limit.
- The results show that cancer mortality risk and cancer morbidity risk lower than the permissible risk limits of 1.0×10^{-3} .
- Approximately 90% of the samples showed HQ >1, indicating chemical toxicity due to the presence of Uranium in tap drinking water, therefore, unsuitable for drinking.

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