Radiological and Chemo-Toxicological Risk Assessment of Naturally Occurred Uranium in Ground Water from Aurangabad District of Maharashtra

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Abstract: In the present study, 40 water samples from different locations in Aurangabad districts of Maharashtra state were collected to analyze for Uranium and associated water quality parameters. Uranium was analyzed with the help of LED Flourimeter (Quantalas India Pvt. Ltd) Model no. LF-2a. The uranium concentration in collected water is varying in the range of 0.012 ppb to 16.673 ppb with an average value 2.75ppb and Median is 3.77 ppb. The Average concentration of uranium was calculated with the range 0.0003 Bq/L to 0.417 Bq/L with an average value 0.068 Bq/L and median is 0.025 Bq/L. The calculated cancer risk of mortality was found in the range of 1.270 X 10⁻⁸ to 1.7465 X 10⁻⁵ with an average value 2.8466 X 10⁻⁶, and also the calculated cancer risk of Morbidity was found in the range from 1.9440 X 10⁻⁸ to 1.4580 X 10⁻¹ with the mean value 3.6494 X 10⁻³ respectively. In the calculation of Lifetime Average Daily Dose was found in the range from 0.00034 to 0.47637 µg/kg/Day with an average value 0.07764 µg/kg/Day and median value 0.02834 µg/kg/Day. The calculated Annual Effective Dose due to ingestion of Uranium in drinking water is varying in the range from 0.00680 to 9.44796 µSv/Year with an average value 1.53991µSv/Year and the median value is 0.56207 µSv/ Year

Keywords: Uranium, Ground water, LED Fluorimeter E.C.R, Chemical Toxicity Risk, Radiological Risk, Annual Effective Dose, pH, Total Dissolved Solids (TDS), Cumulative Dose, physicochemical parameters.

1. Introduction

The Uranium is the basic key element in nuclear power production. It is a atomic number 92, silvery-white metal in the actinide series in the periodic table. This naturally occurs in rock, soil & water. It is having three isotopes i.e. U^{238} with half-life 4.5 x 10⁹ years, the Second one is U^{235} with Half-Life- 700 Million years and the third one is U^{234} with half Life-244000 years correspondingly. Uranium is undergone series of decay and the final stable product is Lead (Pb²⁰⁶).

The important objects that affect in an individual body are kidney and lungs [1]. Some of the main ores of uranium are Pitchblende, uraninite, carnotite, autunite, and torbernite. Uranium is mostly found in the oxidation states of +3, +4, +5, & +6 and the most common out oxidation states are tetravalent and the hexavalent. Uranium (+4) is insoluble and can form complexes by different inorganic legends, for example, fluoride, chloride, sulfate, and phosphate. However,

the greater solubility of U (+6) as the uranyl $(UO2^{++})$ compounds, is due to its ability to form stable complexes with various organic and inorganic legends. [2]. Uranium is classified as a carcinogenic element (group A) and recommended by the United States Environmental Protection Agency (USEPA) in 1991 is recommended the absolute nonexistence of uranium in drinking water as the safe limit for carcinogenic risk. However present the United States Environmental Protection Agency (USEPA) and World Health Organisation (WHO) has proposed a realistic guideline level as maximum contaminant Level (MCL) for uranium to be 30 ppb [3], [4]. The chemical toxicity is a major adverse health effect of uranium, rather than radiological hazards [5-6].

The primary source of uranium is ingestion through water and food and then it accumulates preferably in the liver, kidneys and bones [7]. The absorbed uranium, 66% is rapidly eliminated via urine, even as the rest is distributed and deposits in the kidneys (12-25 %), bone (10-15 %) and soft tissue [8]. Even though its toxicity, the uranium is highly toxic and it is not normally measured as an indicator of drinking water quality. Hence, the measurement of concentration of uranium in drinking water is very significant. The main purpose of the present study is to measure the level of uranium in drinking water samples and it's become very important to calculate the radiological as well as chemical risk, due to the ingestion of uranium, from the point of view of health hazard.

1.1 Geology of the Study Area

Aurangabad District is located in the western side of India. The Aurangabad city is the administrative headquarters of Marathwada region in Maharashtra state. The district covers the total area is 10,100 km², out of which 141.1 km² is urban area and 9,958.9 km² is rural. The total population of Aurangabad District is 3,701,282 (from 2011 Census). The district comprises nine tehsils (Taluka's) i.e Aurangabad, Kannad, Soygaon, Sillod, Phulambri, Khultabad, Vaijapur, Gangapur, and Paithan. The district is located between North Longitude (Degree) is 19 and 20 and East Longitude (Degree) s 74 to 76 at an average elevation of 568 m (1,864 ft) from sea level. The area of the district is geologically covered by the Deccan Trap lava flows of upper Cretaceous to lower Eocene age. The main rivers in Aurangabad district are Godavari, Tapi, Purna, Shivna & Kham. The rainy season starts in Aurangabad district from the month of June to September. In October to February is a Winter Season and March to May is Summer Season. The Average rainfall of Aurangabad District is 734 mm and the Minimum Temperature is 5.6° C & Maximum Temperature is 45.9 °C. The soils in general in the district are alkaline in reaction clay loam in texture and are fairly high in the content of calcium carbonate. The water supply of the district is mainly based on ground water through tube wells. The numbers of tube wells (bore well) and dug wells are using to extraction of groundwater for irrigation purposes in the districts.

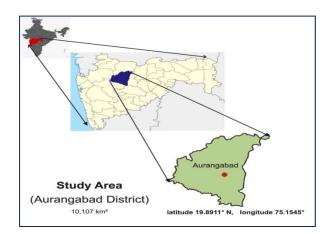


Figure No. 1 Map of the Study Area

2 **Problem Formulation**

2.1 Sample Collections

Before the collection of samples, the air-tight lab grade polypropylene bottles having capacity 1 litre were washed with 10% hydrochloric acid (HCl) to remove contaminants that are absorbed to the inner surface of the bottle. Fresh water samples were taken by running bore well for 2-3 min prior collection to ensure that the fresh water is sampled from the aquifer. The Physical-chemical parameters i.e. Temperature, pH, EC, TDS, Dissolved Oxygen, ORP, Salinity and fluoride were analyzed with the help of Portable Multiparameter Meter kit (Orion Star A326) on the sampling spot. For the analysis of Uranium, the samples are labelled, denoting the details of time, place and date of sampling. The concentration level of uranium was determined in the departmental laboratory in University.

2.2 Measurement of Uranium

For the estimation of uranium content from the collected water samples, LED fluorimeter (Quantalase LF- 2a) was used (Fig.2)



Figure No.2 LED Fluorimeter (Quantalase LF-2a)

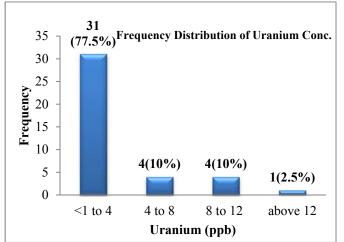
The Fluorescence value varies for different complexes of uranium. Therefore all the complexes were converted into a single form having same fluorescence yield, by addition of (5%) sodium pyrophosphate solution as fluorescence enhancing reagent (Fluren). The concentration of uranium was determined by using LED fluorimeter with the help of equations 1 and 2. The amount of uranium present in the water sample is calculated

Sample counts= (Sample + background) – (background) / (Counts for U Standard) – (background) X 3.....(1)

Conc. Of U in the Sample (ppb)= Sample Counts X 3 / Volume of Sample taken for analysis(2)

2.3 Methodology for Risk Assessment

In the current study, the risk was assessed in two types separately, which is related to the presence of Uranium in drinking water. The first one is a radiological risk which is due to the ionizing radiations emitted by radioactive element uranium and the second one is the chemical risk. The main radiation exposure from uranium occurs when uranium compounds are ingested or inhaled by the individual's body. Uranium is a toxic Heavy metal and it is harmful to human health. The most important chemical effect related to exposure of uranium is kidney toxicity. So it is become very important to calculate the risks associated with uranium.



3 Problem Solution

Uranium concentration of the collected water samples of Aurangabad district and its corresponding risk are tabulated in Table 1. The uranium concentration was varying in the range 0.012 ppb to 16.673 ppb with an average value 2.75ppb and Median is 3.77 ppb. Out of 40 analyzed water samples, 31 (77.5%) were found to be 4 ppb. 4 samples out of 40 (10%) were found to be in 4 to 8 ppb and 4 samples also found to be in 8 to 12 ppb. Only one sample was found above 12 ppb. No sample was found up to 30 ppb, which is the recommended upper permissible limit by USEPA. The Frequency Distribution of Uranium Concentration of samples is shown in the form of a bar diagram in figure 3.

3.1 Assessment of Radiological Risk

Radiological Risk (Excess Cancer Risk) is evaluated using the following equation

- Radiological Risk (Excess Cancer Risk) = U Conc. In Ground Water (Bq/L)* X Risk Factor (Per Bq/L)**......(1)
- *U Conc. (Bq/L) = Measured Value of U (ppb) x Conversion Factor (0.025 Bq/L)
- **Risk Factor = Risk Coefficient (Bq-1) x Water Ingestion Rate (L/Day) x Total Exposure Duration Days..........(2)

The Risk Coefficient in equation (2) for Mortality and Morbidity was taken as 1.19×10^{-9} Bq-1 and 1.84×10^{-9} Bq-1 respectively. The rate for Water ingestion was taken as 1.38 L-Day and total exposure duration was taken 25550 days. Calculated Risk of Mortality and Morbidity is calculated and it is 4.19×10^{-5} and 6.48×10^{-5} respectively. The calculated cancer risk for Mortality and Morbidity was found in the range from 1.25×10^{-8} to 1.75×10^{-5} with the Mean value of 2.85×10^{-6} and in the range from 1.94×10^{-8} to 1.45×10^{-1} with the mean value 3.64×10^{-3} respectively.

3.2 Assessment Chemical Risk

The Chemical Toxicity Risk related to any element is evaluated in terms of Lifetime Average Daily Dose (LADD) and this can be estimated by using the following equation.

Figure No. 3 Bar graph of Frequency Distribution of Uranium Conc in Water Samples.

Lifetime Average Daily Dose [(LADD), $(\mu g/kg/Day)$] = [Cd x IR x EF x LE]/ [BW x AT](3)

HQ = (LADD)/Rfd.....(4)

Where in equation (3), Cd is denoted the uranium concentration in groundwater in (μ g/L), Ingestion Rate (IR) in (L-Day) is which is taken to be 1.38 L/Day. The EF is the Exposure Frequency (Days-Year) which is taken 365 days per year. LE is the life expectancy was taken 70 years. BW is the Body Weight (kg), which is taken as 70 kg. AT is an average time (Days), which is taken as 25550 days. In Equation (4) HQ is said to be the Hazard Quotient (HQ) and Rfd is said to be the Reference dose (μ g/kg/Day), which is taken as 0.857 μ g/kg/Day. [9]

The calculated chemical toxicity Risk i.e Lifetime Average Daily Dose was found in varying in the range from 0.00034 to 0.48 (μ g/Kg/Day) with an Average Value of 0.078 (μ g/Kg/Day). The calculated Hazard Quotient (HQ) was found in between 0.00040 to 0.556, with an average value 0.0905.

3.4 Assessment of Annual Effective Dose

The "Annual Effective Dose" is a biological dose; it determines how unsafe an individual's exposure to radiations. The unit of 'effective dose' is the Sievert. It was estimated using the conversion factors given by

DE = Ac x F x I (annual)(5)

Where in equation (5) is the annual effective dose $(\mu Sv/Year)$, Ac is the average concentration, F is the effective per unit intake $(\mu Sv/Year/ Bq/L)$, which is taken 4.5 x 10-8 and I annual is the annual ingestion, which was taken 503.7 L (1.38 x 365). The estimated Annual Effective Dose was found in the range from 0.00680-9.448 $\mu Sv/year$ with an Average 1.539 $\mu Sv/year$.

The cumulative Dose was calculated for lifetime, and it is found varying in the range from 0.47599-661.343 μ Sv with the mean value 107.791 μ Sv. The Uranium Concentration and Calculated Radiological & Chemical Risks related With Each Water Samples are shown in table no. 1

Sr.	Location details	U	Average	R	R	ECR	ECR	LADD	HQ	DE	Cumulative
No		(ppb)	conc.	(Mortality)	(Morbidity)	(Mortality)	(Morbidity)	(µg/Kg/		(µSv/Yr)	Dose
			(Bq/L)					Day)			(µSv/Lifeti
				1.104105	6 10110 S	0.475.000	0.1.1.50		0.000		me)
1	UNIVERSITY	0.090	0.0023	4.19*10-5	6.48*10-5	9.4E-08	0.1458	0.00257	0.0030	0.0510	3.570
	GATE	0.7(0	0.0100	4.10#10-5	6.40#10-5	0.015.07	1.00105.07	0.022	0.0252	0.4207	20.146
2	SUDHAKAR	0.760	0.0190	4.19*10 ⁻⁵	6.48*10 ⁻⁵	8.0E-07	1.2312E-06	0.022	0.0253	0.4307	30.146
2	NAGAR	2.550	0.0(20	4 10*10-5	(40*10-5	2.7 E 0/	0.000004121	0.072	0.0050	1 4450	101 147
3	BALAPUR(DEVLA	2.550	0.0638	4.19*10 ⁻⁵	6.48*10 ⁻⁵	2.7E-06	0.000004131	0.073	0.0850	1.4450	101.147
4	I) ADGAON BK	0.550	0.0138	4.19*10 ⁻⁵	6.48*10-5	5.8E-07	0.00000891	0.016	0.0183	0.3117	21.816
4	BHALGAON	9.450	0.0138	4.19*10 ⁻⁵	6.48*10 ⁻⁵	9.9E-06	0.000015309	0.018	0.0185	5.3550	374.839
6	ADUL BK	0.690	0.2303	4.19*10	6.48*10 ⁻⁵	7.2E-07	1.1178E-06	0.020	0.0230	0.3910	27.369
7	THAPTI TANDA	0.890	0.0173	4.19*10 ⁻⁵	6.48*10 ⁻⁵	3.0E-07	4.698E-07	0.020	0.0230	0.3910	11.503
8	ANTARWALI	0.290	0.0005	4.19*10	6.48*10 ⁻⁵	2.1E-08	3.24E-08	0.008	0.0097	0.0113	0.793
0	KHANDI	0.020	0.0005	4.19 10	0.48 10	2.112-08	3.24E-08	0.001	0.0007	0.0115	0.795
9	PADHARPUR	2.400	0.0600	4.19*10-5	6.48*10-5	2.5E-06	0.00003888	0.069	0.0800	1.3600	95.197
10	SHIVRAI LIMBE	0.480	0.0000	4.19*10-5	6.48*10 ⁻⁵	5.0E-07	7.776E-07	0.009	0.0160	0.2720	19.039
10	JALGAON	0.100	0.0120	1.19 10	0.10 10	5.01 07	1.1102 01	0.011	0.0100	0.2720	19.059
11	JIKTHAN	0.390	0.0098	4.19*10-5	6.48*10-5	4.1E-07	6.318E-07	0.011	0.0130	0.2210	15.470
12	KADAM	1.475	0.0369	4.19*10-5	6.48*10 ⁻⁵	1.5E-06	2.38918E-06	0.042	0.0492	0.8357	58.499
	SHAHAPUR										
13	NAVIN KAIGAON	6.970	0.1743	4.19*10-5	6.48*10 ⁻⁵	7.3E-06	1.12914E-05	0.199	0.2324	3.9496	276.469
14	AMBEWADI	4.460	0.1115	4.19*10-5	6.48*10 ⁻⁵	4.7E-06	7.22439E-06	0.127	0.1487	2.5270	176.888
	GANGAPUR										
	URBAN										
15	MANJARI	0.358	0.0090	4.19*10-5	6.48*10-5	3.8E-07	5.7996E-07	0.010	0.0119	0.2029	14.200
	(MANULLAPUR										
16	VARKHED	0.891	0.0223	4.19*10 ⁻⁵	6.48*10 ⁻⁵	9.3E-07	1.44407E-06	0.025	0.0297	0.5051	35.358
17	CHOR	0.012	0.0003	4.19*10 ⁻⁵	6.48*10 ⁻⁵	1.3E-08	1.944E-08	0.000	0.0004	0.0068	0.476
	WAGAHLGAON										
18	JAMBARGAON	9.579	0.2395	4.19*10-5	6.48*10 ⁻⁵	1.0E-05	1.55185E-05	0.274	0.3194	5.4282	379.968
19	VAIJAPUR RURAL	10.436	0.2609	4.19*10-5	6.48*10 ⁻⁵	1.1E-05	1.69057E-05	0.298	0.3479	5.9135	413.933
20	FATIYABAD	3.348	0.0837	4.19*10 ⁻⁵	6.48*10 ⁻⁵	3.5E-06	5.42408E-06	0.096	0.1116	1.8973	132.808
21	DIWASHI	0.138	0.0034	4.19*10-5	6.48*10 ⁻⁵	1.4E-07	2.22912E-07	0.004	0.0046	0.0780	5.458
22	KINNAL	2.272	0.0568	4.19*10 ⁻⁵	6.48*10 ⁻⁵	2.4E-06	3.68032E-06	0.065	0.0757	1.2873	90.112

Table1: Uranium Conc. and Calculated Radiological & Chemical Risks related With Each Water Sample

23	MALIWADA	1.995	0.0499	4.19*10-5	6.48*10-5	2.1E-06	3.23174E-06	0.057	0.0665	1.1304	79.129
24	SAWANGI	0.195	0.0049	4.19*10 ⁻⁵	6.48*10 ⁻⁵	2.0E-07	3.1509E-07	0.006	0.0065	0.1102	7.715
25	DAHEGAON	2.113	0.0528	4.19*10-5	6.48*10-5	2.2E-06	3.42355E-06	0.060	0.0705	1.1975	83.825
26	SHIVARAI	2.937	0.0734	4.19*10 ⁻⁵	6.48*10 ⁻⁵	3.1E-06	4.7581E-06	0.084	0.0979	1.6643	116.502
27	MAKARMATPUR	16.673	0.4168	4.19*10-5	6.48*10-5	1.7E-05	2.70103E-05	0.476	0.5559	9.4480	661.343
	WADI										
28	KHANDALA	0.574	0.0143	4.19*10 ⁻⁵	6.48*10-5	6.0E-07	9.2907E-07	0.016	0.0191	0.3250	22.748
29	HILALPUR	0.275	0.0069	4.19*10-5	6.48*10-5	2.9E-07	4.45014E-07	0.008	0.0092	0.1557	10.896
30	CHIKATGAON	1.092	0.0273	4.19*10-5	6.48*10-5	1.1E-06	1.76969E-06	0.031	0.0364	0.6190	43.331
31	TALWADA	0.107	0.0027	4.19*10-5	6.48*10-5	1.1E-07	1.7334E-07	0.003	0.0036	0.0606	4.244
32	DAULATABAD	4.003	0.1001	4.19*10-5	6.48*10-5	4.2E-06	6.48437E-06	0.114	0.1334	2.2682	158.769
33	NANDRABAD	0.315	0.0079	4.19*10-5	6.48*10-5	3.3E-07	5.10462E-07	0.009	0.0105	0.1786	12.499
34	PALASWADI(SHE	1.494	0.0374	4.19*10-5	6.48*10-5	1.6E-06	2.42044E-06	0.043	0.0498	0.8467	59.264
	KAPUR)										
35	GALLEBORGAON	9.060	0.2265	4.19*10 ⁻⁵	6.48*10-5	9.5E-06	1.46767E-05	0.259	0.3020	5.1338	359.358
36	TAPARGAON	7.559	0.1890	4.19*10 ⁻⁵	6.48*10-5	7.9E-06	1.22457E-05	0.216	0.2520	4.2835	299.835
37	SHIVARAI	1.162	0.0290	4.19*10 ⁻⁵	6.48*10 ⁻⁵	1.2E-06	1.88212E-06	0.033	0.0387	0.6583	46.083
38	MAKARAMPUR	0.879	0.0220	4.19*10-5	6.48*10-5	9.2E-07	1.42333E-06	0.025	0.0293	0.4979	34.850
	(K. RURAL)							1			
39	HIVARKHEDA	0.067	0.0017	4.19*10 ⁻⁵	6.48*10-5	7.0E-08	1.08378E-07	0.002	0.0022	0.0379	2.654
	(GAUTALA)										
40	PURANWADI	0.594	0.0148	4.19*10-5	6.48*10 ⁻⁵	6.2E-07	9.61632E-07	0.017	0.0198	0.3364	23.545

Table no.2. Statistical parameters of the obtained data

Statistical	U (ppb)	Avrage conc.	ECR	ECR	LADD	HQ	DE	Cumulative
parameter		in (Bq/L)	(Mortality)	(Morbidity)	(µg/Kg/Day)		(µSv/Yr)	Dose
S								(µSv/Lifetime)
MIN	0.012000	0.000300	1.2570E-08	1.9440E-08	0.00034	0.0004001	0.00680	0.47598600000
MAX	16.673	0.417	1.7465E-05	1.4580E-01	0.47637	0.5558593	9.44796	661.343
Average	2.718	0.068	2.8466E-06	3.6494E-03	0.07764	0.0905986	1.53991	107.791
Median	0.992	0.025	0.000	0.000	0.02834	0.0330688	0.56207	39.344

Earlier it was also stated that the uranium is a ubiquitous element. A good amount of literature is present for its occurrence in groundwater worldwide. Distribution of Uranium concentration in drinking water samples from different cities of India and from some other countries are tabulated in table 3 and table 4 respectively

Table 3: Concentrations of Uranium in Drinking Water Samples from Different Cities of India

Sr.	Cities	Basic Source	U Conc. (µg/l)	References
No.				
1	Shri Ganganagar (Rajasthan)	Groundwater	2.5 - 171	[11]
2	Churu (Rajasthan)	Groundwater	13 - 95	[11]
3	Sikar (Rajasthan)	Groundwater	3 - 136	[11]
4	Himachal Pradesh	Groundwater	0.56 - 10.11	[12]
5	Khalilabad, Gorakhpur, Maharajganj, Kushinagar	Bore well, River water Tap water, open	0.02 - 64.00	[13]
	(Uttar Pradesh)	well		
6	Fatehabad (Haryana)	Groundwater	0.3 - 48	[14]
7	Western Haryana	Groundwater	6.37 - 43.31	[15]
8	Mansa (Punjab)	Groundwater	5.90 - 645.22	[16]
9	Bathinda (Punjab)	Groundwater	7.0 - 323.94	[16]
10	Amritsar (Punjab)	Groundwater	0.87 - 42.51	[16]
11	Hoshiarpur (Punjab)	Groundwater	0.48 - 25.19	[16]
12	Present Study (Aurangabad District)	Groundwater	0.0120- 16.673	
	(Maharashtra)			

1 4)	Table 4. Concentrations of Granitan in Drinking from Different Countries in water Samples						
Sr.	Country	Basic Source	Conc. Of U (µg/L)	References			
No.							
1	Amazonas (Brazil)	Groundwater	0.01 - 1.36	[17]			
2	South western Sinai (Egypt)	Groundwater	328 - 560	[18]			
3	Northern Greece	Groundwater	0.01 - 10	[19]			
4	Russia	Groundwater	>477	[20]			
5	Ulaanbaatar (Mongolia)	Groundwater	< 0.01 - 57	[21]			
6	Switzerland	Groundwater	0.05 - 92.02	[22]			

Table 4: Concentrations of Uranium in Drinking from Different Countries in Water Samples

Table No. 5 Permissible Limit of Uranium conc. in drinking water of Different Authority

Sr No.	Authority/Agency	Permissible Limit of Uranium conc. in drinking water	References
1	World Health Organization (WHO)	15 (μg /l)	[1] [10]
2	United States Environmental Protection Agency (USEPA)	30 (µg /l)	[1] [10]
3	United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)	9 (µg /l)	[1]
4	International Commission on Radiological Protection (ICRP)	1.9 (μg /l)	[1]
5	Atomic Energy Regulatory Board (AERB) DAE, INDIA	60 (µg /l)	[9] [10]

Conclusions

The Current study that the mean radiological, as well as the chemical toxicity risks in the study area is insignificant and well below the permissible limit. As per World Health Organization (30 ppb) & Atomic Energy Regulatory Board (AERB) DAE, INDIA (60 ppb) the limits for uranium in the samples are well below the recommended level. Hence the water samples are safe for consumption. However ICRP, (1993) recommended the safe limit at 1.9 ppb or less. On this criteria samples Nos. 3,7,9,13,14,18,19,20,22,23,25,26,27,32,35 & 36 Needs to be reconsidering for potable use. If possible water from sample point should be avoided for drinking purpose.

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