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Natural radiation of chemical fertilisers and radiological impact on agriculture soil

Akbar Abbasi[1,2,3]

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Abstract

To investigate the radiological hazards indices of chemical fertilisers commonly used in agriculture soils of Arasbaran, Iran, the activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in four types of chemical fertilisers were measured by gamma spectrometry system. The concentration range for ^{226}Ra, ^{232}Th, and ^{40}K was obtained from 54 ± 2.2 to 1585 ± 29.4, 6 ± 0.1 to 157 ± 9.1, and 31 ± 1.5 to 1496 ± 32.8 Bq/kg, respectively. The obtained results showed that the fertiliser's average radiation hazard parameters were higher than the reference level. The outcomes give rise to more parameter values over the permissible limit, increasing the likelihood of adverse health consequences.

Keywords Concentration · Radiological hazards · Chemical fertiliser · Dose rate · Reference level

Introduction

Natural radiation sources of terrestrial origin can be found in varying degrees in all media of the environment, including soil [1–3], sediment [4–7], water [8–11], air [12], and even the human body [13]. One of the radioactivity causes in soils other than those of natural origin is the widespread use of phosphate-rich fertilisers for agricultural reasons [14]. There is a direct relationship between the ^{238}U series and fertiliser phosphorus pentoxide (P2O5) content [15].

Phosphate rock is used as a raw material for fertilisers, and it contains radionuclides from the natural ^{238}U (^{226}Ra) and ^{232}Th series and ^{40}K. Chemical fertilisers, mostly compound commercially known as NPK (nitrogen (N), phosphorus (P), and potassium (K)) and NPKs, are currently used to replace nutrients in soils and thus supply substances to achieve high agricultural productivity (sulfate-based fertiliser). Their formulae differ significantly, and the concentrations are selected based on each soil and cultivation [16]. Excessive fertiliser usage may lead to increased radionuclides in soil and groundwater, which are exposure sources for food crops and drinking water. Gamma radiation from natural radioactivity accounts for 85% of the total global annual average ionising radiation [17]. Higher exposure to such radionuclides is hazardous to human health. It may result in various health issues, including lung, pancreatic, liver, hepatic, bone, and kidney cancer [4].

In general, fertiliser use has grown globally over the last decade, with demand for all fertilisers rising from 161 million tons in 2008 to more than 200 million tons in 2018, with nitrogen fertilisers being the most widely used [18–20]. As a result, during the last ten years, chemical fertilisers, particularly nitrogen, have grown, which has typically been to enhance crop production, the most significant of which was wheat and corn [21]. As a result of the increased harvesting due to the use of artificial fertilisers, environmental expenses and soil and ecosystem degradation are incurred.

Recently, research has been carried out and reported on natural radiation and radiological hazards from chemical fertilisers [22–25]. This study's objectives were to evaluate natural radioactivity concentration in chemical fertilisers used in the Ahar area, Iran, and to calculate the radiological risk due to chemical fertilisers used for agriculture.

✉ Akbar Abbasi
akbar.abbasi@kyrenia.edu.tr

[1] Faculty of Engineering, University of Kyrenia, TRNC, via Mersin 10, Kyrenia, Turkey

[2] European Organization for Nuclear Research (CERN), Geneva, Switzerland

[3] Division of Research and Innovation, University of Manitoba, Winnipeg, MB R3T 2N2, Canada

Material and methods

Study area

The area under study covers the fertile regions of district Arasbaran. Arasbaran, located at 38.45138° N, 47.0676° E, is situated on the northern slopes of the Buzghush mountain. The chief river flowing from the west to east of Ahar is Aharchayi. The area is noted for its Moderate mountains. The area's topography involves rugged terrains, ranging from 800 to 1800 m a.s.l. The study area is shown in Fig. 1.

Sample collection and preparation

Four different main types of chemical fertilisers were gathered from farmers and markets. The investigated sample types are ammonium nitrate (AN), ammonium sulfate (AS), ammonium phosphate (AP), and potassium nitrate (PN). For each type of chemical fertiliser, different brands were sampled. Finally, 21 samples were packed in a polyethene container and transferred to the laboratory for measuring. The collected samples were dried in an oven at around 75 °C) for 24 h to eliminate all moisture; the samples were crushed, homogenised, and sieved through a 200 µm sieve. Twenty-one samples were weighed and put in a 0.5 kg Marinelli beaker. The beakers were sealed entirely for four weeks to achieve secular equilibrium between daughter radionuclide ^{222}Rn and parent ^{226}Ra, which occurs when the daughters' decay rate equals that of the parent. This step is required to guarantee that ^{222}Rn gas is restricted inside the volume and that the daughters remain in the sample [26]. Table 1 shows several physical and chemical features of chemical fertilizers.

Table 1 Some physical and chemical characters of chemical fertilizers

Fertilizer type	Chemical formula	Colour	Density (kg/m^3)	Melting point (°C)
AN	NH_4NO_3	White	1725	169.6
AS	$(NH_4)_2SO_4$	Grey	1770	280
AP	$(NH_4)_3PO_4$	Grey	1619	155
PN	KNO_3	white	2109	334

Fig. 1 The geographical location of study area (Arasbaran, East Azerbaijan)

Measurement process

A gamma-ray spectrometer with a high-resolution coaxial p-type vertical HPGe detector was used to measure the radioactivity. The detector has a relative efficiency of 80% when compared to the NaI (Tl) (3 × 3) detector and has a resolution of 1.8 keV at full-width half-maximum FWHM of the 1332.5 keV gamma-ray photopeak from ^{60}Co. The system was coupled to a high count-rate Multi-Task 16 k MCA card. Commercial software Gamma 2000 from Silena-Italy was used for data analysis. The gamma-ray spectrometer's energy calibration was performed with point sources. The International Atomic Energy Authority supplied reference materials RGU-1 (U-ore), RGTh-1 (Th-ore), RGK-1 (K2SO4), and IAEA-375 were used for full energy peak (FEP) efficiency calibration of the gamma-ray spectrometer. The samples and standard materials geometry were the same geometry. Each of these reference materials was counted in the same geometry with samples until a good counting statistic was achieved. The FEP efficiency (ε_γ) for each gamma-ray energy of interest was then computed using the following equation [1]:

$$\varepsilon_\gamma = \frac{Cr}{P_\gamma . A} \tag{1}$$

where Cr is the net count rate of the region of interest's gamma-ray photopeak, P_γ is the probability of the gamma-ray of interest, and A is the radioactivity of the reference material in Bq. Each soil sample was placed on the detector's top and counted for about 80,000 s. To acquire net counts for the sample, background radiations were measured under the same circumstances as sample measurements and removed. The activity concentrations in the ^{226}Ra and ^{232}Th decay series were averaged from gamma-ray photopeaks at various energies, assuming secular equilibrium. The activity concentration of ^{226}Ra was determined using the gamma-ray lines of 351.9 keV from ^{214}Pb and 609.3 keV from ^{214}Bi. The activity concentration of ^{232}Th was calculated using the gamma-ray photopeaks of 911.2 keV from ^{228}Ac and 583.2 keV from ^{208}Tl. The gamma-ray lines at 1460.8 keV were used to quantify the activity concentration of ^{40}K [27].

Uncertainty calculation

Uncertainty assessment was carried out before activity computation, with the activity concentration uncertainty (U_A) computed using the equation [28]:

$$\frac{U_A}{A} = \sqrt{\left(\frac{U_N}{N}\right)^2 + \left(\frac{U_B}{B}\right)^2 + \left(\frac{U_\varepsilon}{\varepsilon}\right)^2 + \left(\frac{U_M}{M}\right)^2 + \left(\frac{U_{P_\gamma}}{P_\gamma}\right)^2} \tag{2}$$

where U_N is the sample counting uncertainty; U_B, background counting uncertainty; U_ε, efficiency uncertainty, U_M, mass measurements uncertainty and U_{P_γ} *G* amma line energy uncertainty [29].

Radiological risk assessment

Some parameters, such as absorbed dose rate, Ambient dose equivalent rate (H*(10)), gamma index, and alpha index, can be used to calculate exposure risk.

Absorbed dose rate

The absorbed dose rate (D, nGy/h) of gamma radiations in one meter above the ground surface by ^{226}Ra, ^{232}Th, and ^{40}K radionuclides is calculated using Eq. (3). The absorbed dosage rate was calculated using conversion factors based on the particular radioactivity levels, A_K, A_{Ra}, and A_{Th} (Bq/kg) of ^{40}K, ^{226}Ra, and ^{232}Th, respectively[30].

$$D(nGy/h) = 0.462A_{Ra} + 0.604A_{Th} + 0.0417A_K \tag{3}$$

Ambient dose equivalent rate (H(10))*

H*(10) is a quantifiable quantity providing an effective dose assessment that measures the radiation exposure risk to human health. The ambient dose equivalent rate of ^{226}Ra, ^{232}Th, and ^{40}K is calculated 1 m above the ground surface. The computation of the ambient dose equivalent rate (H*(10)) is as follows [1, 31]:

$$H^*(10)(nSv/h) = 0.674A_{Ra} + 0.749A_{Th} + 0.0512A_K \tag{4}$$

where A_{Ra}, A_{Th} and A_K (Bq/kg) are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

Gamma index

This factor calculates the degree of γ-radiation risk in connection with phosphate fertilisers. The hazard gamma index (Iγ) in Bq/kg is calculated using the following equation [32]:

$$I_\gamma(Bq/kg) = \frac{A_{Ra}}{150} + \frac{A_{Th}}{100} + \frac{A_K}{1500} \tag{5}$$

To be at the safe level, the value of Iγ must be less than one, which is related to an annual effective dose of ≤ 1.0 mSv/y [33].

Alpha index

Alpha index (Iα) is a significant hazard evaluation aspect used to detect excess alpha radiation from radioactive sources. This parameter is evaluated using the following formula [34]:

$$I_\alpha(Bq/kg) = \frac{A_{Ra}}{200} \quad (6)$$

According to reference [35], the radiative material activity level of ^{226}Ra above 200 Bq/kg is assumed to contribute to ^{222}Rn concentration > 200 Bq/m^3 of ^{222}Rn exhalation, which recommended activity level of ^{226}Ra is 200 Bq/kg, which gives $I\alpha = 1$ [35].

Results and discussions

Radioactivity concentration

The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K of different fertiliser types, together with the measurement uncertainty (1σ) and average, minimum, and maximum values, are presented in Table 2. From the show, results can be seen that the values of activity concentrations in the studied fertilisers varied from 54 ± 2.2 to 1585 ± 29.4, 6 ± 0.1 to 157 ± 9.1, and from 31 ± 1.5 to 1496 ± 32.8 Bq/kg for ^{226}Ra, ^{232}Th and ^{40}K, respectively. The minimum and maximum values of ^{226}Ra were obtained in potassium nitrate (PN) and ammonium phosphate (AP) fertilisers. The ^{232}Th minimum and maximum concentration values were found in samples of potassium nitrate (PN) and ammonium sulfate (AS) fertilisers. While the sample of potassium nitrate fertiliser (PN) sample showed the highest amount, and the ammonium sulfate fertiliser (AS) sample showed the lowest amount of ^{40}K concentration. The various sources of raw material and chemical processing of the raw during fertiliser manufacturing might explain the variance in radionuclide concentrations in the investigated chemical fertilisers. The ^{226}Ra average concentrations in all chemical fertilisers are higher than ^{232}Th concentrations. In comparison, the ^{226}Ra average activity concentrations in all chemical fertilisers except for AS and PN are higher than ^{40}K concentrations. It is because of the high potassium present in the NP samples. Figure 2 compares the average concentration values of ^{226}Ra, ^{232}Th, and ^{40}K in samples of fertilizer as a percentage.

Table 2 The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K in different types of fertilizers

Fertilizer type	Sample code	Activity concentration (Bq kg^{-1})		
		^{226}Ra	^{232}Th	^{40}K
Ammonium Nitrate (AN)	AN-1	218 ± 5.3	108 ± 8.4	132 ± 6.2
	AN-2	184 ± 7.3	22 ± 1.8	85 ± 3.7
	AN-3	72 ± 5.6	46 ± 3.6	117 ± 5.1
	AN-4	397 ± 8.7	97 ± 5.4	209 ± 9.4
	AN-5	152 ± 6.8	39 ± 2.0	57 ± 2.5
Min- Max (Average)		72–397 (204.6)	22–108 (62.4)	57–209 (120.0)
Ammonium Sulfate (AS)	AS-1	118 ± 3.7	58 ± 3.7	126 ± 4.6
	AS-2	187 ± 4.2	80 ± 4.5	84 ± 3.2
	AS-3	93 ± 2.2	157 ± 9.1	501 ± 14.4
	AS-4	219 ± 9.1	95 ± 5.2	217 ± 10.8
	AS-15	198 ± 8.1	24 ± 1.6	31 ± 1.5
	AS-6	212 ± 8.5	91 ± 5.1	162 ± 5.7
Min- Max (Average)		93–219 (171.1)	24–157 (84.1)	31–501 (186.8)
Ammonium Phosphate (AP)	AP-1	1585 ± 29.4	128 ± 6.0	183 ± 9.4
	AP-2	855 ± 12.4	133 ± 6.2	46 ± 2.1
	AP-3	1017 ± 14.7	71 ± 3.7	112 ± 5.2
	AP-4	363 ± 7.1	11 ± 0.3	907 ± 33.8
	AP-5	481 ± 9.6	27 ± 1.8	210 ± 14.6
Min- Max (Average)		363–1585 (860.2)	27–133 (74.0)	46–907 (291.6)
Potassium Nitrate(PN)	PN-1	347 ± 8.5	6 ± 0.1	1496 ± 32.8
	PN-2	286 ± 6.3	108 ± 5.1	1368 ± 25.0
	PN-3	174 ± 4.2	52 ± 3.1	502 ± 14.7
	PN-4	54 ± 2.2	84 ± 3.8	390 ± 27.2
	PN-5	120 ± 3.9	17 ± 0.5	1407 ± 29.6
Min- Max (Average)		54–347 (196.2)	6–108 (53.4)	390–1496 (1032.5)

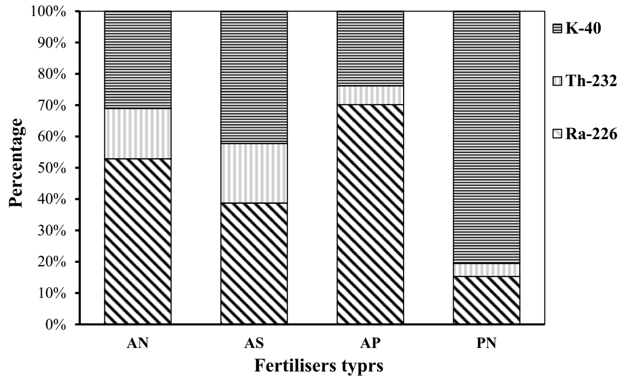


Fig. 2 Percentage comparison between average concentration values of ^{226}Ra, ^{232}Th and ^{40}K in fertilisers samples

Radiation hazard indices

Table 3 shows the gamma index(Iγ), alpha index(Iα), absorbed dose rate (D), and ambient dose equivalent rate (H*(10)) values for all samples under investigation. As shown in Table 3, Ammonium Nitrate (AN), Ammonium Sulfate (AS), Ammonium Phosphate (AP), and Potassium Nitrate (PN) gamma index values ranged from 1.0–8.3 Bq/kg, 1.5–2.6 Bq/kg, 3.1–12 Bq/kg and 1.5–3.9 Bq/kg, respectively. The gamma index (Iγ) average of 2.1 Bq/kg for Ammonium Nitrate (AN) and Ammonium Sulfate (AS) was obtained. Whilst, gamma index (Iγ) average of 6.5 Bq/kg for Ammonium Phosphate (AP) and 2.5 Bq/kg for Potassium Nitrate (PN) were calculated. It is found that Iγ for all fertilisers is less than six except for fertilisers ammonium phosphate type. This value indicates that almost of sample is safe to use following the requirements of the European Commission (EC) [36] ($2 > I\gamma > 6$). The calculated average Iα values for the four group samples under investigation were 0.9 to 4.3 Bq/kg. The recommended limit is more than the unit (< 1) for Iα that is according to 200 Bq kg^{-1} of ^{226}Ra [35]. The alpha index (Iα) in the sample Ammonium Sulfate was less than the given limit. In contrast, ammonium nitrate (AN) and potassium nitrate (PN) were equal to the limit range. Also, the alpha index value in ammonium phosphate (AP) was more than four times. These observed values are

Table 3 The gamma index(Iγ), alpha index(Iα), absorbed dose rate (D), and ambient dose equivalent rate (H*(10)) with maximum-minimum and average values in fertiliser samples

Sample type	Sample code	Hazard indices			
		Iγ (Bq/g)	Iα (Bq/kg)	D (nGy/h)	H*(10) (nSv/h)
Ammonium Nitrate (AN)	AN-1	2.6	1.1	171.5	234.6
	AN-2	1.5	0.9	101.8	144.8
	AN-3	1.0	0.4	65.9	89.0
	AN-4	3.8	2.0	250.7	350.9
	AN-5	1.4	0.8	96.2	134.6
	Max–Min (Avg.)	3.8–1.0 (2.1)	2.0–0.4 (1.0)	250.7–65.9(137.2)	234.6–89.0–(190.8)
Ammonium Sulfate (AS)	AS-1	1.5	0.6	94.8	129.4
	AS-2	2.1	0.9	138.2	190.3
	AS-3	2.5	0.5	158.7	205.9
	AS-4	2.6	1.1	167.6	229.9
	AS-15	1.6	1.0	107.3	153.0
	AS-6	2.4	1.1	159.7	219.3
	Max–Min (Avg.)	2.6–1.5(2.1)	1.1–0.5(0.9)	167.6–94.8(137.7)	229.9–129.4(188.0)
Ammonium Phosphate (AP)	AP-1	12.0	7.9	817.2	1173.5
	AP-2	7.1	4.3	477.3	678.2
	AP-3	7.6	5.1	517.4	744.4
	AP-4	3.1	1.8	212.2	299.3
	AP-5	3.6	2.4	247.3	355.2
	Max–Min (Avg.)	12.0–3.1(6.7)	7.9–1.8(3.4)	817.2–212.2(454.3)	1173.5–299.3(650.1)
Potassium Nitrate (PN)	PN-1	3.4	1.7	226.3	315.0
	PN-2	3.9	1.4	254.4	343.7
	PN-3	2.0	0.9	132.7	181.9
	PN-4	1.5	0.3	91.9	119.3
	PN-5	1.9	0.6	124.4	165.7
	Max–Min (Avg.)	3.9–1.5(2.5)	1.7–0.3(1.0)	254.4–91.9(166.0)	343.7–119.3(225.1)

more than unity and show that the ammonium phosphate fertilisers are not safe from environmental radiation hazards and need more investigation. Figure 3 shows the studied samples' alpha index's maximum, minimum, and average values.

As shown in Table 3, the value of absorbed dose rate (D) in Ammonium Nitrate, Ammonium Sulfate, Ammonium Phosphate and Potassium Nitrate samples ranged from 65.9–250.7 nGy/h, 94.8–167.6 nGy/h, 212.2–817.2 nGy/h and 91.9–254.4 nGy/h, respectively. The maximum and minimum absorbed dose rate (D) was found in Ammonium Phosphate (AP) and Ammonium Nitrate (AN). The average value of absorbed dose rate (D) in all studied fertilisers is higher than the international limit of 59 nGy/h [13].

As illustrated in Table 3, the values of ambient dose equivalent rate (H*(10)) vary between 89.0–234.6 nGy/h, for AN, 129.4- 229.9 nGy/h for AS, 299.3–1173.5 nGy/h for AP, and 343.7–119.3 nGy/h for PN. While the maximum and minimum values of H*(10) were in ammonium phosphate and Ammonium Sulfate fertiliser samples, respectively.

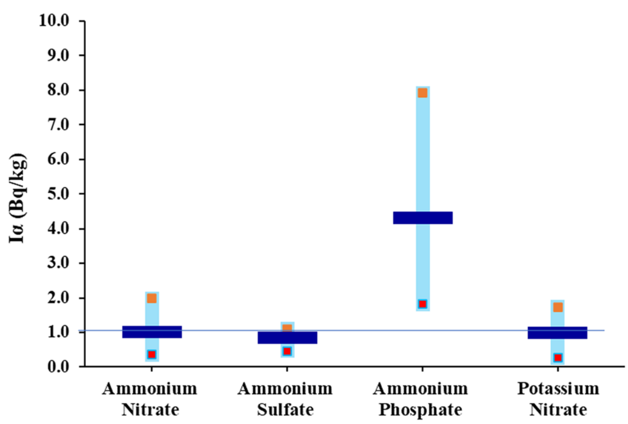


Fig. 3 The maximum and minimum and main values of the alpha index (Iα) in the fertilisers samples

The H*(10) average values in all fertilisers were higher than the average ambient dose equivalent rate value in soil samples(119.16 nGy/h) reported by the author [1]. Figure 4 shows the maximum and minimum with the main ambient dose equivalent rate (H*(10)) in four types of studied fertilisers.

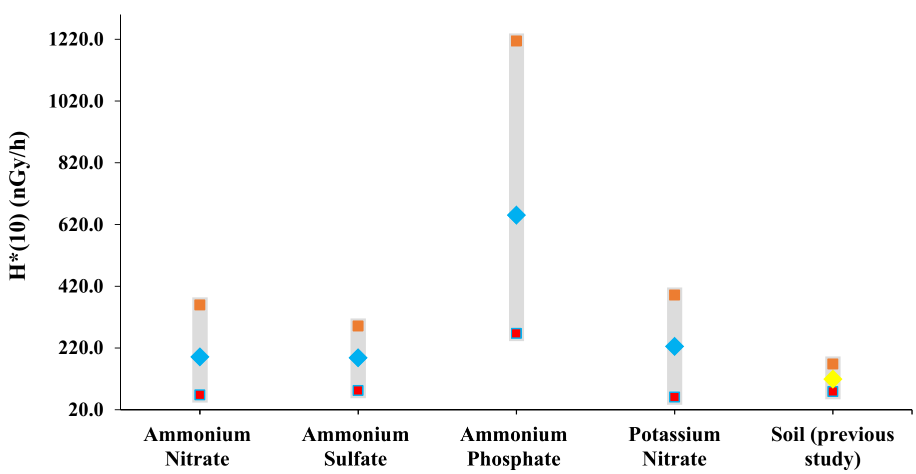
Table 3. The gamma index(Iγ), alpha index(Iα), absorbed dose rate (D), and ambient dose equivalent rate (H*(10)) with maximum-minimum and average values in fertiliser samples.

Conclusion

Low-level gamma spectrometry was used to determine the activity concentrations of natural radionuclides ^{226}Ra, ^{232}Th, and ^{40}K in 21 samples from 4 types of fertiliser in the Arasbaran area, Iran. The obtained results have shown that the radiation hazard parameter Iγ for all fertiliser type samples) is higher than the reference level (> 1), except for the AN-3 sample, which is equal to the limit value. Also, the radiation hazard parameter Iα average value for all fertiliser samples is higher than the reference level > 1, except for the ammonium sulfate (AS) samples. In all investigated samples, the average values of absorbed dose rate (D) due to the fertiliser samples are higher than the reference level of 59 nGy/h [13] and ranged from 137.2 to 454.3 nGy/h.

The computed average ambient dose equivalent rate (H*(10)) values for each group fertiliser were found to be 1.6 to 5.5-fold in typical soil samples (Fig. 4). The use of fertilisers has a significant impact on radionuclide concentrations in agriculture soils, especially phosphorus and potassium-containing fertilisers being one of the causes of the high activity of ^{226}Ra and ^{40}K in the soil. Using these fertilisers causes a buildup of radioactivity in soils, which can be detrimental to the health of farmers, employees, and product consumers.

Fig. 4 The range and main value of ambient dose equivalent rate (H*(10)) in the fertilisers samples and comparison of H*(10) value in previous studied soil samples



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Authors' contributions Abbasi contributed to the design and implementation of the research and the analysis of the results.

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Declarations

Conflict of interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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