

## CS - 137 AND K - 40 CONCENTRATION IN SOIL AND THEIR TRANSFER TO PLANT

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### Abstract

A total number of 780 samples of soil collected over Greece and measured with gamma-rays spectroscopy, sixteen years after the arrival of the Chernobyl "cloud" and its fallout all over the country. An analysis for the long-lived isotope of  $^{137}\text{Cs}$  and the natural exist  $^{40}\text{K}$  has been performed. It seems that still  $^{137}\text{Cs}$  presents a remarkable geographical variability and an inversely proportionality with the natural  $^{40}\text{K}$  concentration of soil. According to the present data regarding the  $^{137}\text{Cs}$  deposition on Greece, the uneven spotted distribution has been confirmed.

Follow up representative measurements for soil contaminations have indicated minor migration of  $^{137}\text{Cs}$  beyond to 5 cm from the surface layer. The insignificant in depth (beyond 5 cm) radiocesium contamination in Greek soils it was expected knowing the composition of these soils, the majority of which are rich in clay and silt, and contain in excess exchangeable potassium. The radioactivity range varies from 0.4 to 14.4 kBq/m<sup>2</sup>. The transfer of  $^{137}\text{Cs}$  from soil to corn plant was investigated 16 years after a similar work done soon after the Chernobyl accident. Correlations were made between  $^{137}\text{Cs}$  transfer factors and (i)  $^{40}\text{K}$  in plant and soil, (ii) pH of water and KC1. (iii) Exchangeable K, Ca and Mg. The uptake of  $^{137}\text{Cs}$  by corn plant in 2010 seems to be unchanged when compared to the situation in 1988.

**Keywords:** *Radioactivity, soil, plant, transfer factor (TF)*

### Introduction

During Chernobyl accident, radioactive contamination with radionuclides, that reaches Greece, was due to tropospheric fallout (Greek Atomic Energy Commission, 1988). and thus, crops were, sown six months after the accident or later - until today - contaminated with radionuclides, almost exclusively through their root system. The main characteristics, which determine how dangerous are the radionuclides, are their physical and biological half-life and the kind and the energy of their radiation emitted by them. From the long-living radionuclides, cesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) and strontium ( $^{89}\text{Sr}$  and  $^{90}\text{Sr}$ ) isotopes burden the environment for greater time period. The absorbance of radioactive substances by the plants, by the man and animals can be direct and indirect. In the direct absorbance the plant but, also, man and animals are directly incorporating the radioactive substances. This way of incorporation plays an important role in the first days after the accident. The second way of incorporation - the indirect - presupposes the absorbance of radionuclides through the way ground - roots - plant - animal - man. The indirect way of contamination is the most important and influences the food for greater period of time. Soil to plant transfer of various radionuclides is known to be affected by soil properties, plant species and variety, climatic condition and cultural practices. From the soil properties texture, pH, exchangeable potassium and calcium, kind and amount of clays and organic matter content are amongst the most important (Papanikolaou, 1972). Variation of the concentration of radionuclides on the soil surface depends mainly on its

mineralogical composition; its chemical and physical properties, meteorological conditions and the possible transfer of material to deeper soil layers (Missaelidis et al., 1987), (Vosniakos et al. (1998). The possibility of fixation of Cs isotopes by geological material and soil has been the subject of previous studies (Sikalidis et al., 1988). The mechanism of fixation depends strongly on the mineral composition of the soil. For example, soils in Northern Greece have high clay content with vermiculite (mica family) being the predominant mineral, which is a well-known potassium and cesium fixing material (Sikalidis et al., 1988). The existence of  $^{137}\text{Cs}$  in the soil is important because of its possible transfer to the cultivated plants and eventually to animals and humans. It also increases the direct exposure doses received by humans from terrestrial natural radioisotopes by 10% (Kritidis and Kollas, 1992). The majority of N. Greece soils have pH values between 7 and 8, exchangeable potassium. 0.5 meq / 100g (typical value), and are heavy textured because of its high clay (49% typical) and silt (37% typical) contents (Antonopoulos-Domis, 1990). Finally, the transfer of Cs is increased with increasing organic matter content (Berfeijk et al., 1992). Compared to the transfer of Cs in soils with an organic matter content of less than 5%, the effect was approximately 2-5 to 20% increasing up to a factor of 10 on soils with an organic matter content of more than 50%. Organic matter in Greek soils is less than 5% and does not favor the uptake of Cs by plants (Antonopoulos-Domis, 1990). In the pH range of 3.9 - 8.4, transfer of Cs is not affected by soil pH (Berfeijk et al., 1992). Transfer of Cs in clay and loam is lower than in sand, by 5 and 3 times, respectively (Berfeijk et al., 1992).  $^{137}\text{Cs}$  and  $^{40}\text{K}$  have the same oxidation number and almost the same diameter. Absorption of Cs by plant, though, is smaller than the absorption of K probably because of smaller contiguity of Cs with the carriers of the cellular membranes of the plant roots (Mavrogiannopoulos, 1992). Obviously, another reason is that  $^{40}\text{K}$  concentrations are about one order of magnitude greater than  $^{137}\text{Cs}$  in the soil and Cs has to compete with stable K too, which is even in higher (a further 4 orders of magnitude) concentrations.

Firstly the aim of the present work is to identify the level of the existing  $^{137}\text{Cs}$  contamination over Greece sixteen years after the Chernobyl accident and a comparison between the 1986  $^{137}\text{Cs}$  - distribution and the present measured one in more - less the same areas of Greece, has been attempted. The  $^{40}\text{K}$  (0.0118% of natural K) concentration in soils as ratio  $^{137}\text{Cs}/^{40}\text{K}$  has been, examined, even this ratio is not as constant in biological systems as the ratio Sr/Ca (Eisenbud, 1973). Secondly, is to investigate the soil to plant transfer of  $^{137}\text{Cs}$  and  $^{40}\text{K}$ . The areas chosen were Drama (East Macedonia) and Thessaloniki (Central Macedonia). The sites are characterized by different soil contamination levels ( $^{137}\text{Cs}$  contamination increases in the East - Central - West direction). The plant chosen was corn because it constitutes main kind of food for cows in the country.

### Materials and methods

During the period of January 2003 - May 2010, 780 soil samples of surface soil (0-5 cm) were collected over Greece (Fig. 1). It was tried the soil samples to be taken from apparently undisturbed sites in open areas at the ground surface. Deeper soil samples (5-50 cm) were collected as well, but no didactable amount of  $^{137}\text{Cs}$  has been recorded, as it was expected since the mobility for Cs is very low,  $0.2 \text{ y}^{-1}$  (Bonazzola et al., 1993). The sampling of surface soil, of about  $500 \text{ cm}^3$  each, were taken from geographic divisions of Greece with emphasis to those where in 1986 serious depositions of  $^{137}\text{Cs}$  (from  $15 \text{ kBq/m}^2$  and more) were observed (JRC-Rem Project, 1986). The  $^{137}\text{Cs}$  concentration, near the soil surface is strongly time dependent, because of its variable deposition rates over many years and its gradual depletion by decay, erosion and leaching (NCRP Reports, 1988). The uptake of  $^{137}\text{Cs}$  from soil has been show to be inversely proportional to the K content of soils (Eisenbud,

1973) that was also the case in the present work.

The  $^{137}\text{Cs}$  average deposition in Greece was ranged in 1986 between, 0.01 to 14.4  $\text{kBq/m}^2$  (Simopoulos, 1989). Similar measurements of  $^{137}\text{Cs}$  concentration in England for the period of 1990-1991, shows, a range of 0.7-0.8  $\text{kBq/m}^2$  (Aarkrog et al., 1988), in Denmark the total deposition of  $^{137}\text{Cs}$  ranged from 0.66 to 3.6  $\text{kBq/m}^2$  (Aarkrog et al., 1988) while the  $^{137}\text{Cs}$  deposition in Italian soils had a mean value of  $30 \pm 17$   $\text{kBq/m}^2$  immediately after the Chernobyl accident (Kritidis and Kollas, 1992). The present work estimates the  $^{137}\text{Cs}$  accumulation in the Greek soils sixteen years after the Chernobyl accident, to be ranged between 0.4-14.4  $\text{kBq/m}^2$ . The  $^{40}\text{K}$  activity is between 5.1 and 16.5  $\text{kBq/m}^2$ .  $^{40}\text{K}$  and the radionuclides of the U and Th series contribute most of the naturally occurring radioactivity in soils. It is known that  $^{40}\text{K}$  concentration in soils ranges between 0.51-15.54  $\text{kBq/m}^2$  (Eisenbud, 1973). All samples were kept in sealed containers for at least 15 days to allow equilibration between the isotopes  $^{226}\text{Ra}$  and  $^{222}\text{Rn}$ .

*Plant and soil sample collection and preparation.*

At least 15 plants (5 from each site) were collected. Each plant was taken by digging the soils carefully to collect the whole plant (roots, plant and grain) and the soil surrounding the roots. Collection points were at least 40 m apart to cover as much variability of the site as possible. Plants were harvested at maturity from openfields. Corn plants were separated from their roots and grains, thoroughly washed, and cut to small pieces. They were air-dried for two days in the laboratory and then in the oven (Melag) kept at 80 °C. They were weighed (mass of dry plant) and burned at 500 °C in another oven (Nabertherm) for at least 24 hours to get carbon free white ash. This was done because Cs in a certain volume of ash is 8-9 times more than the Cs in the same volume of dry plant. It should be mentioned that Cs is not lost by oven-drying at 500 °C (boiling points of Cs and K, 687°C and >750°C). Plant ash was filled in the standard geometry white cup (cylindrical. 7 cm d. and 2 cm h.), recommended for measurement by the Greek Nuclear Research Centre - Demokritos.

Soil samples were also air-dried, oven-dried (80 °C), pulverized, sifted (2 mm sieve) and weighed (mass dry soil). Then they were put to fill standard cups.

*Measuring device (Gamma ray spectroscopy)*

*Detector:* Hp Ge semiconductor coaxial p-type. CP series Tennelec;

*Energy range:* 40 KeV - 10 MeV;

*Detector temperature:* Liquid nitrogen in vacuum cryostat;

*Sample chamber:* Low background shielding (5 cm lead. 0.5 cm copper);

*High voltage:* 2100 volts DC;

*Amplifier:* Tennelec 242 TC;

*PC-system:* In place of pulse generator and MCA;

*Energy calibration:*  $^{60}\text{Co}$  and  $^{137}\text{Cs}$  standard sources linear CHL - KeV relation ( $R = 0.99999$ );

*Activity calibration:* Standards, dry soils and dry plants ( $^{137}\text{Cs}$ : 1 count/sec corresponds to 90 Bq;  $^{40}\text{K}$ : 1 count/sec corresponds to 276 Bq.);

*Counting times:* 76000 sec;

*Resolution:* FWHM 1.95 KeV at 1332 KeV of  $^{60}\text{Co}$ . *Dead time:* 0-3%.

## Results and discussion

In Fig.2 the  $^{137}\text{Cs}$  concentration versus that of  $^{40}\text{K}$  is plotted. From this figure it seems that  $^{137}\text{Cs}$  is inversely proportional with  $^{40}\text{K}$  in the soil. Besides in Fig. 3 where the logarithm of  $^{137}\text{Cs}$  concentration is plotted versus that of  $^{40}\text{K}$ , it is clear that the experimental data are concentrated along the solid line described by the equation  $y = C [1 - A \exp(Bx)]$  where  $y = \log^{137}\text{Cs}$ ,  $x = \log^{40}\text{K}$  and A, B, C are constant coefficients estimated by the experimental

data, showing that the uptake of  $^{137}\text{Cs}$  from soil is inversely proportional to  $^{40}\text{K}$  content of soil, in which there is a potassium deficiency. It is obvious from Fig. 4 shows that, the ratio of  $^{137}\text{Cs} / ^{40}\text{K}$  is not constant, even cesium is a congener of potassium (Eisenbud, 1973).

Activities of  $^{137}\text{Cs}$  and  $^{40}\text{K}$  in Bq/kg dry wt are given in Table 1. Transfer factors are defined as activity concentration in plant over activity concentration in soil.  $^{137}\text{Cs}$  activities in plant vary between 0.01-2.13 Bq/kg and in soil between 23.1-51.1 Bq/kg. Transfer factors vary between 0.0015-0.0771. Transfer factor values are in agreement with the ones found 16 years ago by another group (Aarkrog et al., 1988) meaning that Cs fixation mechanism in the soil has not really affected the availability of Cs and its uptake by the plants.

$^{137}\text{Cs}$  concentration in the plant decreases as  $^{40}\text{K}$  in the plants increases. The correlation coefficient is -0.66 for a linear fit. Fig. 5 shows a similar correlation for  $^{137}\text{Cs}$  transfer factor versus  $^{40}\text{K}$  in the soil. This finding implies that plants grown in soils poor in K (airport case) are expected to have higher Cs uptake.

Table 1 shows the pH variation of water and KCl ranging between 7.53-8.32 and 7.08-7.56 respectively. Low R values (Fig. 6) show that pH does not affect transfer factors (TF) at least when pH is less than 8.4. This is in agreement with other results (Berfeijk et al., 1992).

Table 1. Activities of Cs and K, transfer factor and soil pH and exchangeable cation status

Site	Activities and transfer factor and soil properties (Ag = Agios, Air = Airport, Dr = Drama; Exch. = exchangeable cations)										
	Activities ( $\text{Bq kg}^{-1}$ )				Transfer		Soil pH		Exch. ( $\text{meq kg}^{-1}$ )		
	Plant		Soil		factor		$\text{pH}_{\text{H}_2\text{O}}$	$\text{pH}_{\text{KCl}}$	K	Ca	Mg
	Cs	K	Cs	K	Cs	K					
Ag1	0.07	428	42.4	549	0.0015	0.780	7.6	7.3	11.0	240	29
Ag2	0.02	391	-	-	-	-	7.5	7.2	10.0	225	39
Ag3	-	-	-	-	-	-	7.8	7.3	9.3	242	33
Ag4	0.00	333	51.1	531	0.0000	0.628	7.6	7.1	14.0	240	32
Ag5	0.01	314	41.5	530	0.0003	0.593	7.8	7.4	11.0	244	27
Air1	1.68	297	38.3	199	0.044	1.488	8.3	7.6	5.4	261	62
Air2	2.10	251	27.8	219	0.075	1.146	8.2	7.5	5.9	267	72
Air3	0.90	337	39.8	211	0.023	1.593	8.2	7.5	6.2	263	64
Air4	2.06	246	34.5	204	0.060	1.203	8.1	7.5	6.1	256	63
Air5	2.13	233	27.6	234	0.077	0.998	8.3	7.5	4.6	255	60
Dr1	1.17	352	23.6	545	0.050	0.646	7.7	7.2	7.8	248	15
Dr2	1.13	321	23.6	464	0.048	0.692	7.6	7.2	7.3	244	16
Dr3	1.16	411	25.9	430	0.045	0.956	7.8	7.2	9.2	260	16
Dr4	0.97	405	26.2	466	0.037	0.868	7.8	7.3	8.5	255	17
Dr5	1.34	423	23.1	472	0.058	0.896	7.6	7.1	9.4	257	16

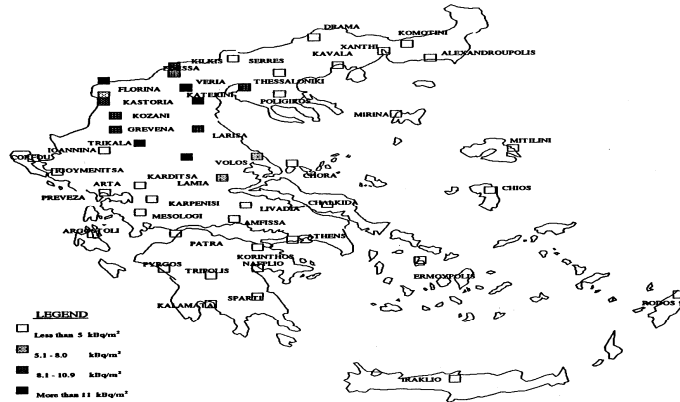


Figure 1. Map of radioactive contamination in Greece

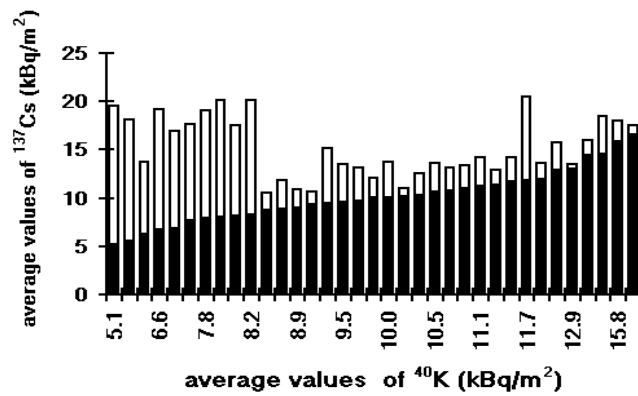


Figure 2. The  $^{40}\text{K}$  average concentrations versus that of  $^{137}\text{Cs}$  (kBq/m<sup>2</sup>)

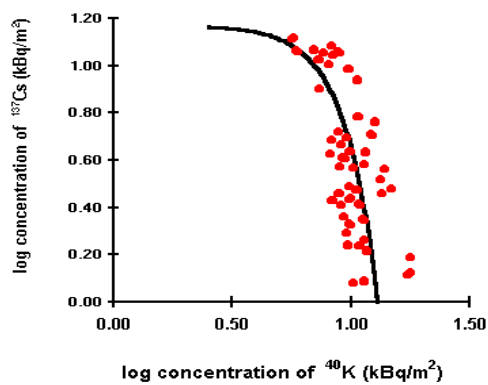


Figure 3. The logarithm of  $^{40}\text{K}$  concentration versus that of  $^{137}\text{Cs}$  (kBq/m<sup>2</sup>)

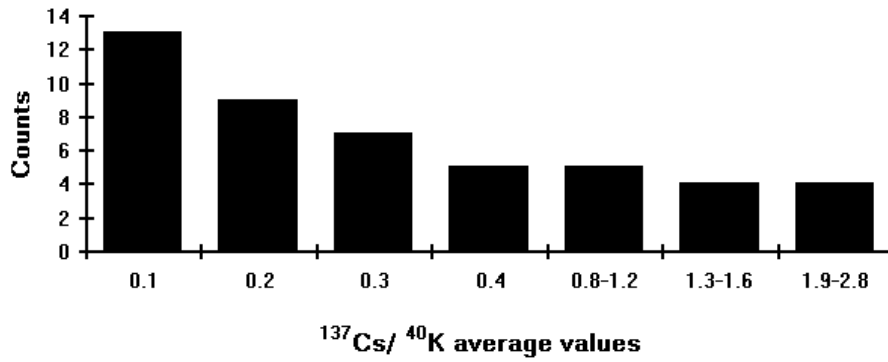


Figure 4. The ratio  $^{137}\text{Cs}/^{40}\text{K}$  from average values

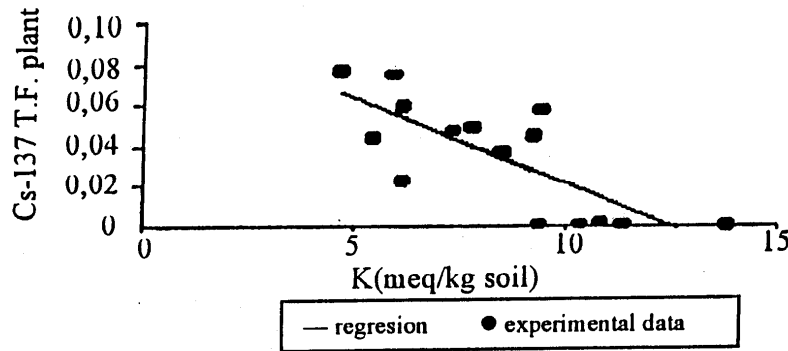


Figure 5.  $^{137}\text{Cs}$  transfer factor versus  $^{40}\text{K}$  in soil

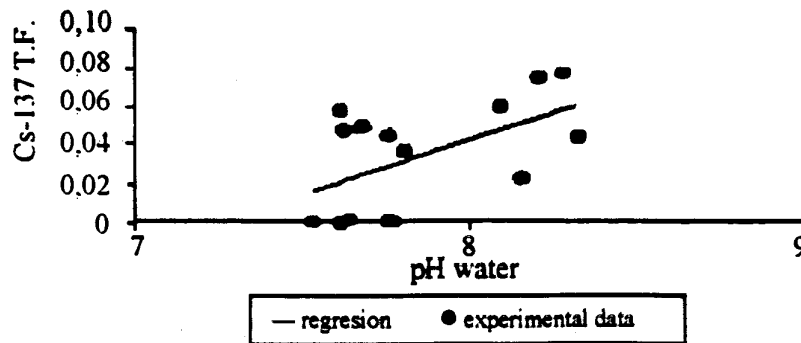


Figure 6. Variation of transfer factor versus pH.

Table 3 shows the content of exchangeable K, Ca and Mg in soil. Fig. 7 shows the correlation between Cs TF and exchangeable K in soil ( $R=-0.78$ ).

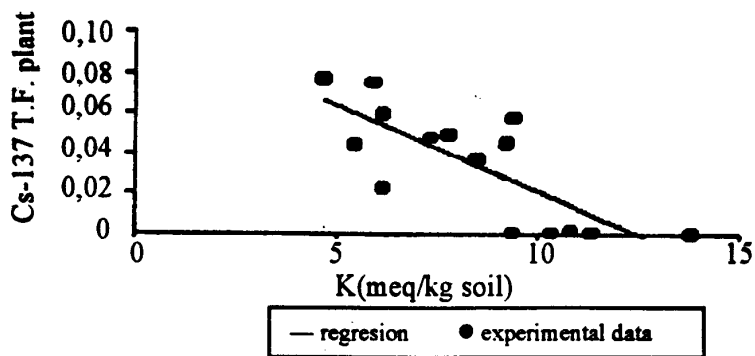


Figure 7. <sup>137</sup>Cs transfer factor versus k<sup>+</sup> in soil

Fig. 8 shows the correlation between Cs TF and exchangeable Ca in soil (R=0.70). Fig. 9 shows the poor fit for Mg (R=0.29). Table 2. shows the mean values of %clay, silt and sand.

Table 2. % clay, silt and sand in soil.

Name	% clay	% silt	% sand
Agiosl	30,1	45,9	24,4
Drama	27,8	24,5	48,6
Airport	34,5	41,8	24,5

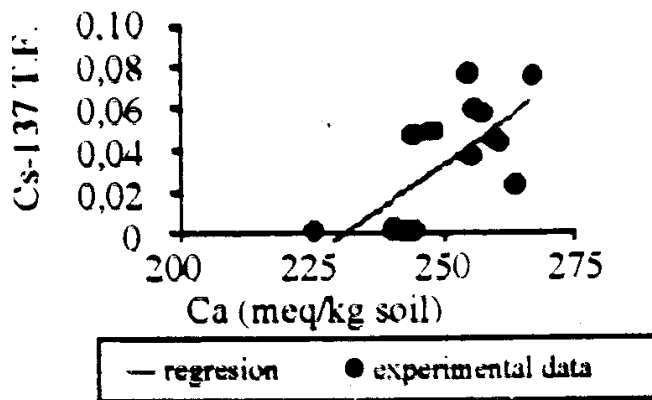


Figure 8. <sup>137</sup>Cs transfer factor versus Ca<sup>++</sup> in soil

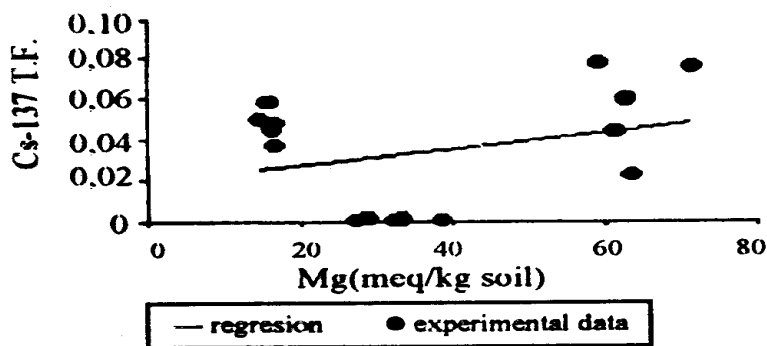


Figure 9. <sup>137</sup>Cs transfer factor versus Mg<sup>+</sup> in soil.

## Conclusions

It may be concluded that  $^{137}\text{Cs}$  uptake by corn in 2010 has not changed compared to the situation in 2003. The Cs fixation in soil does not seem to have affected the uptake as much as initially anticipated.

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