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Quality Evaluation of Several Brands of Bottled Mineral Water from Egypt and Saudi Arabia

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Twenty four brands of uncarbonated ground bottled water in Egypt and Saudi Arabia markets were analyzed using different techniques for evaluating their quality. To identify and classify the major and minor elements concentrations of the studied water samples, inductively coupled plasma optical emission spectroscopy (ICP-OES) technique was used. The water quality is also judged by the percent of total dissolved salts, conductivity and pH. The natural radioactivity was considered as well. The quality of water brand was also determined by analyzing the used plastic bottle, Therefore in the present work, the chemistry of the water bottles was investigated using FTIR technique. The obtained results were compared to several international standards; International Bottled Water Association (IBWA), United States Food and Drug Administration (FDA) and United States Environmental Protection Agency (EPA). All elements were less than the limits of the three international standards, except arsenic in K4, K9 and E4 samples, lead appeared only in K1 and K8, selenium in K1 was more than the limit in International Bottled Water Association but less under United States Food and Drug Administration and United States Environmental Protection Agency.

Keywords: Bottled water, Trace elements, HPGe spectroscopy.

INTRODUCTION

Bottled mineral water gains importance due to its rapid growth in the world markets and its expanding among all human ages in daily use. The spread of bottled mineral water in the entire world due to its price, availability, testiness and fewer impurities[1] leads to customers' trust. Many studies were carried out to identify and predicate its full analysis[2]. There exist over 5000 brands of bottled water which are controlled and tied by many of international standards[3], developed by international organizations to set the secure limits for physical, chemical and microorganisms parameters[4]. Water and its contents of minerals are very important for the human life[5], some of them are very important but others are not or harmful/toxic[6], therefore the elemental composition of bottled water get in focus in the last few years around the world[7].

Water from drilled wells and from free-flowing are the sources utilized in the bottled water industry. The bottled water industry in Egypt depends only on drilled wells, but in Saudi Arabia there are many sources (labeled on all brands). The sales of Egyptian bottled water growth are over 370.5 million liter[8]. A lot of humors spread out about the Egyptian bottled water contamination[9] or containing toxins and polluted because the depth of wells are less than 200 meters[10] and in Saudi Arabia[11,12]. In 2013, Egypt's health ministry listed only 18 brands which are currently licensed bottled water in Egypt[13]. Twenty four brands of bottled mineral water of different drilled wells in different regions of Egypt and Saudi Arabia, collected from supermarkets and all of them in plastic containers.The concentration of trace and ultra-trace elements, pH, conductivity and other different analysis are carried out in the present work. Each water sample was analyzed for 34 chemical elements (As, Ba, Be, Bi, Cd,Cs, Co, Cu, Ga, Fe, Pb, Li, Mg, Mn, Ni, Se, Ag, Na, Sr, Zn, Cr, K, Al, V, Ca, B, Ge, Mo, Nb, P, Re, Si, Ta, Ti). Current results were compared to several international standards; International Bottled Water Association (IBWA), United States Food and Drug Administration (FDA) and United States Environmental Protection Agency (EPA).

EXPERIMENTAL

Twenty-four brands of ground bottled water in Egypt and Saudi Arabia markets were collected randomly from supermarkets and conserved under 10-15 °C. Twelve

Egyptian brands named from E1 to E12 and the Saudi Arabia twelve brands named from K1 to K12. The bottled water samples capacity between 0.5 L and 1.5 L. All analysis was carried out within 1-5 h after samples opening, except in the case of γ-rays measurement the samples were detected overnight.

The analyses were carried out at Egyptian Atomic Energy Authority and KACST, Atomic Energy Research Institute. The elements (As, Ba, Be, Bi, Cd, Cs, Co, Cu, Ga, Fe, Pb, Li, Mg, Mn, Ni, Se, Ag, Na, Sr, Zn, Cr, K, Al, V, Ca, B, Ge, Mo, Nb, P, Re, Si, Ta, Ti) in the bottled water samples were measured using prodigy high dispersion inductively coupled plasma optical emission spectrometer (ICP-OES) (Teledyne Leeman ICP-OES USA). The operational conditions were shown in Table-1. pH was measured by METTLER TOLEDO MP 230 pH Meter. Electrical conductivity (EC) was measured by Profi lab LF597S Conductivity measuring unit. Measurement of γ-rays was carried out by using the HPGe spectroscopy system. The instrumentation used to measure γ-rays from radioactive samples (NORM or man-made) generally consists of a semi-conductor detector, associated electronics and a computer-based, multi-channel analyzer (MCA/computer). The PET container was characterized using Fourier transform infrared FTIR spectroscopy of THERMO model 100.

TABLE-1
OPERATING CONDITIONS OF ICP-OES

ICP spectrometer	Leeman Prodigy Prism ICP-OES
RF Power	1.2 KW
Coolant gas flow	20 L/min
Auxiliary gas flow	0.3 L/min
Nebulizer gas flow	36 psi
Solution Uptake Rate	1 mL/min
Mg II/Mg I Ratio (Robustness)	6
Replicates	3
Integration time	10 sec

RESULTS AND DISCUSSION

The elements Ca, K, Mg and Na are the major elements, but the others are of low concentrations in bottled water[2]. All the elements were obtained by the inductively coupled plasma-optical emission spectrometer (Tables 2 and 3).

Conductivity and pH: The conductivity range in Egyptian bottled water were from 152 to 380 μS/cm accompany with TDS values 102 and 256, but in Saudi Arabia water ranged from 164.4 to 323 μS/cm accompany with TDS values 110 and 217. Tables 4 and 5 show that the conductivity values are related to the TDS values[7]. The pH range values between (7.32 to 8.27) in Egyptian samples but in Saudi Arabia samples from (7.25 to 7.95).

TABLE-2
ELEMENTAL COMPOSITION OF EGYPTIAN SAMPLES BY ICP-OES

Element	E1	E2	E3	E4	E5	E6	E7	E8	E9	E10	E11	E12
As	0.007	< DL	< DL	0.011	< DL	< DL	< DL	< DL	0.009	< DL	< DL	0.006
Ba	0.003	0.004	0.157	0.158	0.199	0.015	0.106	0.015	0.205	0.232	0.04	0.04
Be	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Bi	0.017	0.019	0.026	0.021	0.017	0.018	0.026	0.026	0.025	0.023	0.017	0.010
Cd	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Cs	0.003	< DL	0.001	0.002	< DL	0.002	0.003	0.003	0.004	< DL	0.001	0.001
Co	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Cu	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ga	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Fe	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Pb	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Li	0.015	0.017	0.018	0.030	0.017	0.015	0.030	0.023	0.034	0.019	0.016	0.017
Mg	3.577	7.496	5.657	4.336	5.928	8.414	5.619	6.919	5.070	7.422	4.059	3.649
Mn	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ni	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Se	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ag	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Na	10.220	26.873	21.574	28.206	14.206	12.090	26.096	24.562	24.334	29.569	29.596	33.100
Sr	0.112	0.333	0.396	0.149	0.223	0.054	0.040	0.056	0.051	0.293	0.254	0.202
Zn	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Cr	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
K	1.284	4.847	23.292	17.662	5.143	1.666	14.969	2.92	16.847	6.356	2.236	1.548
Al	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
V	0.001	0.004	0.003	< DL	0.001	< DL	0.001	0.003	0.003	0.005	0.002	0.004
Ca	1.804	2.462	2.098	1.332	2.467	1.552	1.302	2.331	1.184	2.373	1.760	1.723
B	0.142	0.173	0.134	0.151	0.135	0.134	0.158	0.149	0.160	0.171	0.297	0.195
Ge	0.011	0.005	0.006	0.006	0.005	0.005	0.004	0.005	0.006	0.004	0.007	0.002
Mo	0.120	0.044	0.031	0.025	0.016	0.016	0.017	0.018	0.017	0.015	0.015	0.013
Nb	0.069	0.017	0.011	0.021	0.003	0.004	0.016	0.007	0.004	0.003	0.016	0.016
P	0.120	0.039	0.009	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Re	0.001	0.003	0.004	0.005	0.005	0.004	0.004	0.004	0.005	0.005	0.005	0.006
Si	0.610	1.032	0.727	0.695	0.600	0.424	0.634	0.722	0.847	0.543	0.335	0.336
Ta	0.013	0.008	0.010	0.013	0.011	0.012	0.012	0.014	0.014	0.012	0.014	0.008
Ti	0.012	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011	0.011

TABLE-3
ELEMENTAL COMPOSITION OF SAUDI ARABIA SAMPLES BY ICP-OES

Element	K1	K2	K3	K4	K5	K6	K7	K8	K9	K10	K11	K12
As	< DL	< DL	< DL	0.01	< DL	< DL	< DL	< DL	0.01	< DL	< DL	< DL
Ba	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Be	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Bi	0.02	0.04	0.03	0.01	0.05	0.04	0.01	< DL	0.04	< DL	0.01	< DL
Cd	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Cs	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Co	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Cu	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ga	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Fe	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Pb	0.01	< DL	< DL	< DL	< DL	< DL	< DL	0.01	< DL	< DL	< DL	< DL
Li	0.01	0.02	0.02	0.02	0.01	0.02	0.02	0.02	0.03	0.01	0.01	0.02
Mg	9.26	1.47	4.55	4.22	2.83	3.44	3.42	3.93	3.09	7.04	1.19	3.56
Mn	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ni	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Se	0.032	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ag	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Na	18.4	65.08	33.37	27.15	41	22.24	26.74	22.11	22.79	14.16	38.80	40
Sr	< DL	0.07.	0.01	0.02	0.08	0.09	0.16	0.20	0.19	< DL	< DL	0.04
Zn	0.01	< DL	0.01	0.06	0.01	0.02	0.04	0.08	0.01	0.04	0.03	0.02
Cr	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
K	1.47	1.50	0.20	1.63	1.50	2.86	1.51	1.43	1.50	0.20	0.09	3.60
Al	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
V	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ca	1.50	4.83	5.21	3.49	8.47	2.56	3.17	3.47	5.43	5.34	2.41	6.89
B	0.08	0.32	0.14	0.20	0.18	0.08	0.26	0.20	0.26	0.19	0.13	0.34
Ge	< DL	< DL	0.03	< DL	< DL	< DL	0.02	0.02	0.01	0.02	< DL	0.05
Mo	0.01	0.07	0.03	< DL	0.01	0.01	0.01	< DL	0.01	0.01	< DL	0.01
Nb	0.03	0.04	0.02	0.03	0.02	0.02	0.04	0.04	0.02	0.02	0.03	0.02
P	< DL	0.06	< DL	0.01	< DL	0.061	0.11	0.11	0.01	0.08	0.08	0.02
Re	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Si	0.03	0.57	0.03	1.62	1.97	1.71	0.67	1.59	1.23	0.12	0.02	0.22
Ta	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL
Ti	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL	< DL

TABLE-4
pH, CONDUCTIVITY AND TDS FOR EGYPTIAN SAMPLES

	E1	E2	E3	E4	E5	E6	E7	E8	E9	E10	E11	E12
pH	7.61	8.22	7.97	8.05	7.91	7.91	7.93	7.84	7.78	8.27	7.41	7.32
Conductivity	152	343	315	255	243	228	259	288	247	380	270	325
TDS	102	230	216	171	164	149	175	192	166	256	182	221

TABLE-5
pH, CONDUCTIVITY AND TDS FOR SAUDI ARABIA SAMPLES

	K1	K2	K3	K4	K5	K6	K7	K8	K9	K10	K11	K12
pH	7.42	7.36	7.69	7.35	7.49	7.35	7.33	7.25	7.71	7.35	7.95	7.84
Conductivity	185.3	206	304	198.4	199.7	198.3	208	194.7	164.4	221	192.8	323
TDS	124	138	204	133	134	133	140	130	110	148	134	219

Inductively coupled plasma-optical emission spectrometer (ICP-OES): Ca, Mg, Na and K are themajor elements that play important roles in human life[7]. In Egyptian samples, Ca levels were between 1.184 and 2.467 mg/L. Mg levels 3.577 and 8.414 mg/L. The maximum level of Na was 33.1 mg/L and the minimum was 10.22 mg/L. The K levels were between 1.284 and 23.292 mg/L.In Saudi Arabia samples, Ca levels were between 1.5 and 8.47 mg/L. Mg levels were between 1.19 and 9.26 mg/L. Na levels were between 14.16 and 65.08 mg/L. K levels were between 0.09 and 3.6 mg/L. In addition to these elements there were trace elements such as Al, As, Ba, Be, Cd, Cu, Fe, Pb, Mg, Mn, Ni, Se, Ag, Zn and Cr all of them presented in therange ofinternational standards. Some of them are essential for life functions and other are toxic[5]. Arsenic in K4 and K9 was more than the limits in FDA and EPA but in the limits of IBWA but arsenic in the sample E4 is more than the three standards. Lead appeared only in K1 and K8 and was twice the limits in IBWA and FDA but less than the limit in EPA. Selenium in K1 was more than the limit in IBWA but less under FDA and EPA. The elements (Be, Cd, Co, Cu, Ga, Fe, Pb, Mn, Ni, Se, Ag, Zn, Cr and Al) were less than detection limit in all Egyptian samples. The elements (Ba, Be, Cd, Cs, Co, Cu, Ga, Fe, Mn, Ni, Ag, Cr, Al, V, Re, Ta and Ti) were less than detection limit in Saudi Arabia samples.

Measurement of γ-rays: It was carried out by using the HPGe spectroscopy system. Every sample was placed in the HPGe spectroscopy system over night for the full counting.

All results showed that the radio nuclides, which emit γ-rays are less than the detection limits of the HPGe spectroscopy system.

FT-IR spectroscopy of water bottles: FT-IR spectroscopy provides information regarding intermolecular interaction *via* analysis of FT-IR spectra corresponding to stretching or bending vibrations of particular bonds. The position at which these peaks appear depends on the bond type[14]. The monomer of PET polymer consists of the following functional groups; 1) Two ester groups, 2) aromatic ring and 3) one ethyl group.

The FTIR spectra of the 24 water brands used in the present study are presented in Figs. 1 and 2. Two bands exist for C-O stretching bond at 1110 cm^{-1} and 1238 cm^{-1}. The strong band found at 1725 cm^{-1} is due to the stretching vibration of the C=O bond of the ester group. Absorption bands associated with C=O bond stretching are usually strong because a large change in the dipole takes place in that mode. For the ethyl group there is exists two kinds of vibrations for the C-H bond; i) the medium C-H stretching bond which is found at 2970 cm^{-1} and ii) the strong C-H bending bond which is found at 731 cm^{-1}. The C-H stretching band of the aromatic ring exists at 3055 cm^{-1}. Also the band at 1578 cm^{-1} is assigned to the C-H bond stretching vibration of the phenyl ring. The C-C phenyl ring stretching band exists[14,15] at 1408 cm^{-1}. The characteristic band found at 1340 cm^{-1} and 1370 cm^{-1} are clearly observed. These bands corresponding to wagging mode of ethylene units in *trans*-conformation and gauche conformations, respec-

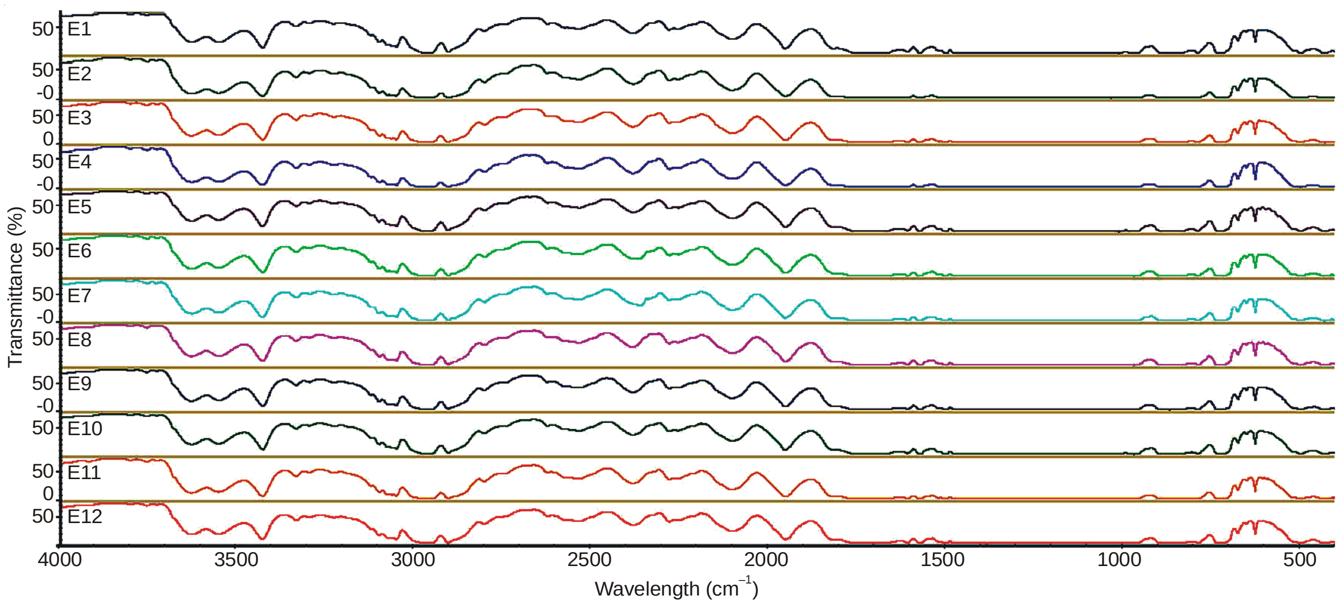

Fig. 1. FTIR spectrum to Egyptian water bottles

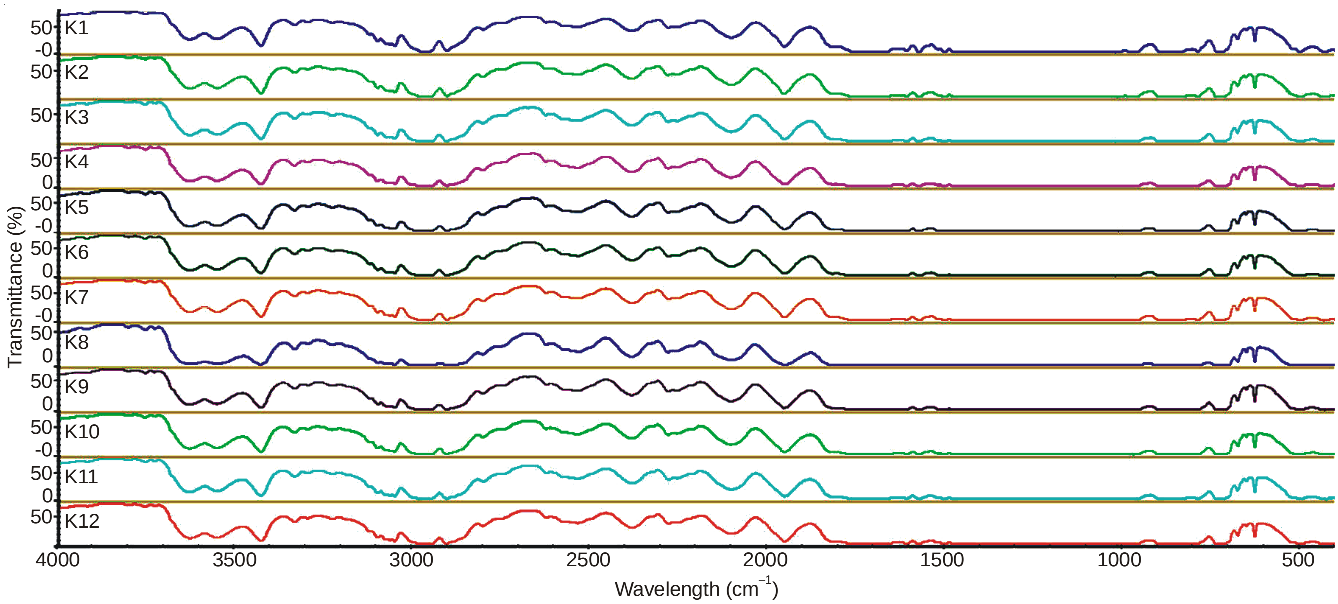

Fig. 2. FTIR spectrum to Saudi Arabia water bottles

tively[16]. The *trans*-form is made favourable by crystallization and stretching, while the gauche conformations are characteristics of the amorphous state[17]. All the bottles have the same chemical structure and nearly the same bonds concentrations as seen from the FTIR spectra which ensure quality of the bottles.

Conclusion

This work provides a survey of the elemental composition and quality of some bottled mineral waters available on Egypt and Saudi Arabia markets. Licensed water brands in Egypt and Saudi Arabia were found to have a good quality and satisfy the quality requirements of several international standards organizations; International Bottled Water Association (IBWA), United States Food and Drug Administration (FDA) and United States Environmental Protection Agency (EPA).

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