MANSOURA JOURNAL OF PHYSICS
Official Journal of Faculty of Science, Mansoura University, Egypt
E-mail: scimag@mans.edu.eg ISSN: 2974-4954

Radiological Impact and Excess Cancer Risk of Terrestrial Radionuclides from Al-Zubair Petroleum Station (Iraq) using NaI(TI) Detector

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Received:17/4/2022
Accepted: 28/5/2022

Abstract: The radioactivity distribution pattern has been estimated for naturally occurring radioactive materials due to exploration and deep drilling to extract crude oil from the Al-Zubair petroleum station from Iraq. The investigated samples were collected from pre-determined locations of Al-Zubair station and their surrounding areas concerning extraction locations of petroleum ores and storage places of TE-NORM residues. The library of radionuclides has been designed to involve the natural uranium and thorium decay series. Based on the measured activity concentrations, the radiation doses were evaluated utilizing the conversion dose coefficients recommended from IAEA, ICRP, and UNSCEAR. For samples from surface to the depth of one kilometer, the resulting values are significantly normal compared to the international and worldwide reference values like the Environmental Protection Agency and IAEA in this regard. The overall mean value of the measured activity concentrations of 226Ra, 232Th, and 40K is (3627.66, 192.92, and 630.89 Bq/kg, respectively). Deeply (> 1 km) where soil coat close to the petroleum layer, which contains an abundance of TE-NORM, the corresponding calculated dose indicated is not permissible level, so strong safety and security precautions must be applied to protect occupational from the radiation hazards. To furnish the calculations data of radiation protection and human safety, integral organ doses and the excess lifetime cancer risk have been calculated to recommend the perfect safety principles to occupational radiation protection programs and perspective on the assessment of environmental TE-NORM risk.

Keywords: Radionuclides; Radiation dose; Radiological impact; Excess Cancer Risk; NaI(TI) Scintillation

1.Introdction

Radioactivity is usually defined as the spontaneous transformation of an unstable atom, often resulting in the emission of radiation, in the form of electromagnetic waves and particles. We inhale radionuclides from the air, soil, water, and food every day since natural radioactivity is common in soil, especially in uranium, mineral mines and petroleum fields. The decay process is accompanied by the emission of nuclear particles or photons carrying the energy in excess. Thus, radioactivity analysis is a complex process aiming to identify and quantify radioactive isotopes since the nuclear radiation may occur in various types, abundances, and energies, characteristic to each radionuclide [1-6]. Technologically Enhanced Naturally Occurring Radioactive Material (TE-NORM) is defined as Naturally occurring radioactive materials that have been concentrated or exposed to the accessible environment as a result of human activities such as manufacturing, mineral extraction, or water processing [7,8].

Al-Zubair Energy Company is situated in southern Iraq, approximately 20 kilometers southwest of Basrah city. The onshore oil field, discovered by Basrah Petroleum Company in 1949, has 4.5 billion proved reserves. A partnership led by Eni (32.81 %), Oxy (23.44 %), KOGAS (18.75 %), and Missan Oil Company (25 %) is redeveloping the field [9]. The field authorization was granted to the consortium in October 2009 for 25 years. The consortium's largest shareholder began managing the oil field in early 2010. Currently, the area produces around 360,000 barrels of oil per day. The rehabilitation project is planned to boost the field's capacity to 850,000 barrels per day in 2018, as indicated in Fig. 1.

The revitalization project includes drilling over 200 wells, installing treatment and storage facilities, and renovating existing infrastructure [9]. Accordingly, the current study aims to investigate and radiological survey of Al-Zubair petroleum station and assess the radiobiological impact.

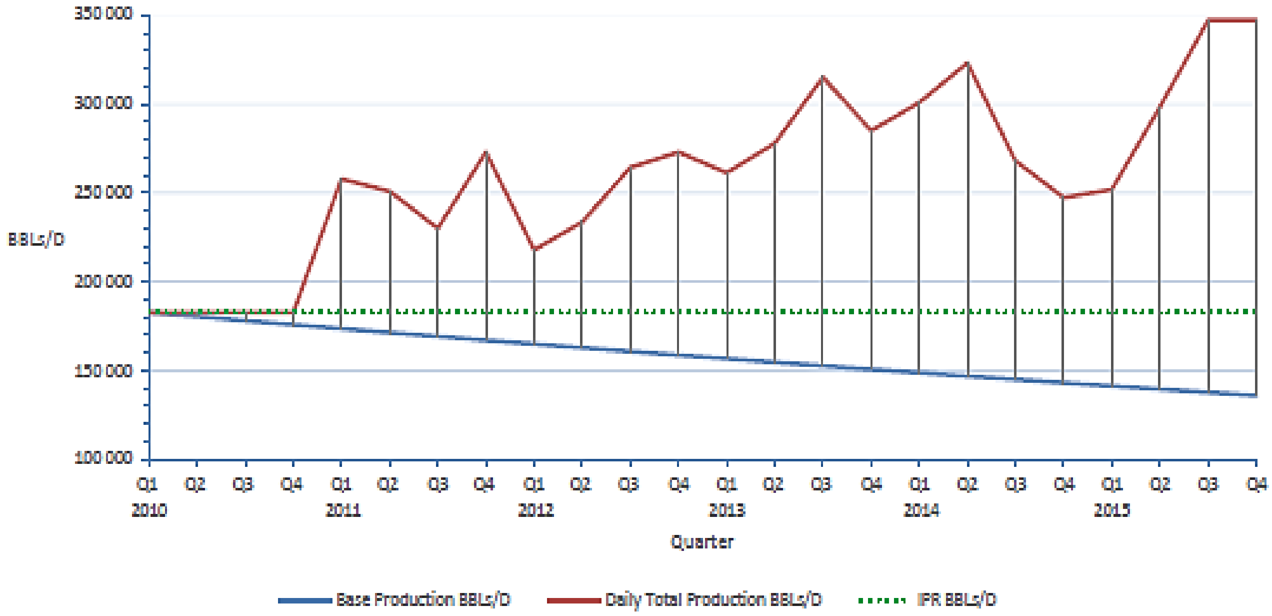

Fig. 1: Oil production of Al-Zubair petroleum field [9]

2. Materials and Analysis Technique

2.1. Sampling strategy

Fulfilling the IAEA protocol, TECDOC-1415, and the Egyptian Geological Survey and Mining Authority recommendations, 31 samples were collected from Al-Zubair station and their surrounding areas [10, 11]. The investigated soil samples were dried in an electric oven at 110°C for 1 hour. All samples were grinded, and ethanol solution was used for cleaning the grinding bowl to avoid contamination from different samples. The grinding process is important to obtain a fine powder recommended for the high quality of efficiency transfer and correction [10, 11]. Sample aliquots were weighted and stored to keep the similar volume of all samples in sealed polyethylene containers, with a diameter of 8 cm and height of 3 cm [1-3, 7, 12].

2.2. Experimental Technique

The samples have been analyzed using a gamma-ray spectrometer of NaI(TI) sodium iodide scintillation detector consisting of a 30% relative efficiency and PCA3-version software developed by the group of nuclear and radiation measurement at Oxford university, including the gamma analysis option. A single crystal of NaI(TI) sodium iodide thallium activated is used as a scintillator. This crystal is a hygroscope; thus, it is sealed into aluminum foil except for one side, which has an optical light-tight contact with a photomultiplier tube. Magnesium oxide coated on the aluminum inside to reflect light. The efficiency calibrations used have been done using the standard source of ^{226}Ra and identical source containers like those used for the samples analyses.

No density corrections have been applied due to the analyzed samples' narrow range of apparent densities, with values close to 1 g/cm^3 [1, 2, 10, 11]. The sample containers used were cylindrical plastic boxes with a diameter of 8 cm and a height of 3 cm, filled with around 170 cm^3 of sample material.

2.2. Evaluation of Radiological Impact

The natural radionuclide concentrations in the soil were used for radium, thorium, and potassium to calculate the radiation hazard indicators for humans resulting from the continuous exposure to the radiation present in the surrounding environment. The samples' activity concentration A_x was calculated by equation (1), where the (1120 KeV) peak of ^{214}Bi was used to determine ^{226}Ra activity, and the (2614 KeV) as a peak of ^{208}Tl was used to determine ^{232}Th activity. The (1460 KeV) energy peak of ^{40}K was used to directly determine the ^{40}K activity concentration [13, 14].

$$A_x = \frac{Net\ Area}{\varepsilon FTM} \quad (1)$$

where ε represents the full energy peak efficiency under the given experimental conditions, *F* is the emission probability of the Gamma-ray produced at the energy peak, *T* corresponds to the measurement time in seconds, and *M* is the sample mass.

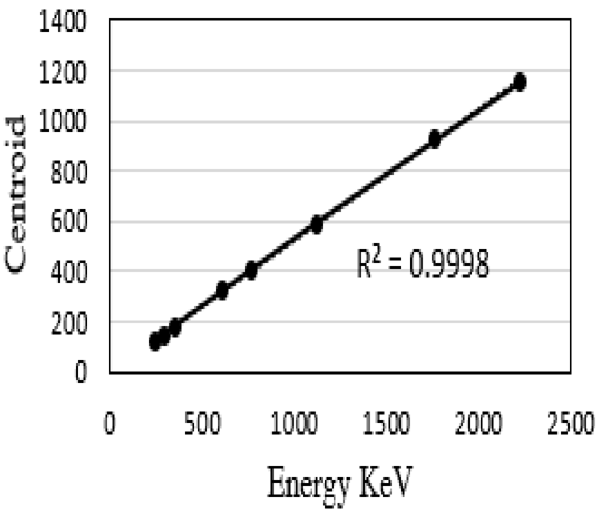

Fig. 2: The curve of energy calibration of ^{226}Ra

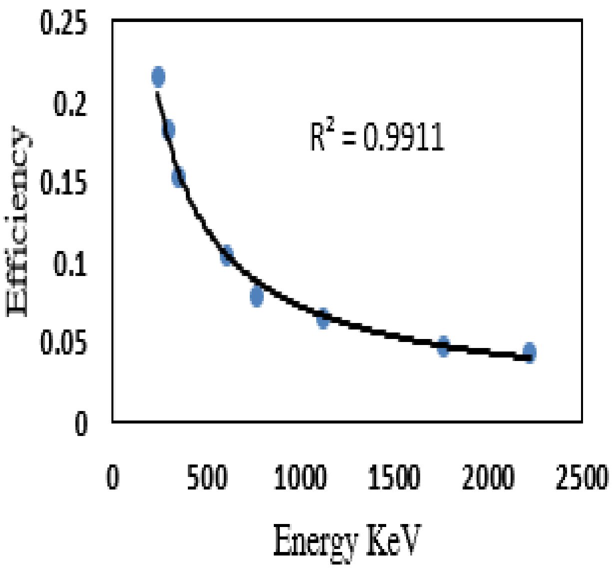

Fig. 3: The curve of efficiency calibration of ^{226}Ra

2.2. Radium Equivalent

The equivalent radium (Ra_{eq}) activity is the radiation factor using the uniform distribution of natural radionuclides represented by radium-226، thorium-232 and potassium-40, and it is measured in units of Bq/kg [13, 14], whereas the equation shall calculate the Radium equivalent has the form [1, 6, 15]

$$Ra_{eq} = C_{Ra} + 1.43 C_{Th} + 0.077 C_K \quad (2)$$

with C_{Ra}, C_{Th}, and C_k are the activities concentration of ^{226}Ra, ^{232}Th and ^{40}K (Bq/kg), respectively.

The external and internal hazard index

The external hazard index due to the emitted gamma ray can be calculated from [1, 6, 15]

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (3)$$

For limiting the annual external gamma-ray dose from materials to 1.5 µGy, the external hazard index $H_{ex} \le 1$. The radioactivity level to which the general population may be exposed is estimated to the limit 0.5 nGy/y [11, 15, 16]. An internal hazard index governs the internal exposure to radon-222 (^{222}Rn) and its daughter products [1, 5, 6], H_{in}, is given by

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (4)$$

and the maximum radium concentration is half the normal, giving acceptable limit $H_{in} \le 1$

Radiation Index

The gamma index I_γ that is used to estimate the level of gamma-ray radiation hazard associated with the natural radionuclides, is defined as

$$I_\gamma = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \le 1 \quad (5)$$

and the reprehensive radiation level index is given by

$$I = \frac{C_{Ra}}{50} F_{Ra} + \frac{C_{Th}}{50} F_{Th} + \frac{C_K}{500} F_K \le 2 \quad (6)$$

where F_{Ra}, F_{Th} and F_K are a fractional percentage to the total rate from ^{226}Ra, ^{232}Th and ^{40}K, respectively, F_{Ra} = 8.09 %, F_{Th} = 47.98 % and F_K = 43.92 % [1, 2, 6].

Exposure Rate E_R and Dose Rate D_R

The radiation exposure rate E_R (μR/hr) is due to the combined radionuclide concentrations were quantified according to

$$E_R = 1.9C_{Ra} + 2.82C_{Th} + 0.179C_K \quad (7)$$

to give the dose rate (mSv/y) as

$$D_R = 0.0833E_R \quad (8)$$

E_R (μR/hr) is equivalent to 2.576×10^{-4} C/kg [6, 7, 17-19].

Annual effective dose equivalent

The annual effective (*AEDE*) in (mSv/y) is calculated using the equation [6,15,20]

$$AEDE = 365.25d \times 24h \times 0.7\times10^{-3} D\,F \quad (9)$$

where D is dose rate in (nGy/h), the facto $365.25d \times 24h$ is the outdoor occupancy time, F denotes occupancy factor as 0.2 for outdoor and 0.8 for indoor, and 0.7×10^{-3} is the conversion coefficient in (mSv/Gy) [14, 20].

Effective Dose Rate D_{organ} toDifferent Body Organ or Tissue

The effective dose D_{organ} (mSv/yr) is considered for the whole body while applying a tissue weighting factor (T_w) of maximum one. For calculating the dose for any specific tissue inside the body, it has to be a fraction of 1, whereas the different tissues are affected differently, and then the tissue weighting factors are not equal and are divided to three categories. Low risk (0.01), medium risk (0.05) and high risk (0.12) [15, 16, 19, 21, 22].

$$D_{organ} = D_{air} T_w \quad (10)$$

where D_{air} is absorbed dose rate (nGy/hr), and T_w is the weighting factor for specific tissue inside the body, as shown in table 1 [21,22].

Table 1: The values of weighting factor utilized to calculate the effective dose

No.	Tissue/organ	T_w
1	Stomach, colon, lung, bone marrow	0.12
2	Gonads	0.20
3	Urinary bladder, esophagus, liver, glands, and breast	0.05
4	Bone surface and skin	0.01
5	Other body organs	0.05

Annual Gonadal Dose Equivalent

The Annually Gonadal Dosage Equivalent (AGDE) measures the genetic relevance of the annual equivalent dose absorbed by the population's reproductive organs. Considering the bone marrow and bone surface cells to be organs of interest in the same context [5]. The concentrations of ^{226}Ra, ^{232}Th and ^{40}K in AGDE (mSv/y) shall be determined using the equation

$$AGDE = \left(3.09C_{Ra} + 4.18\,C_{Th} + 0.314\,C_K\right)\times10^{-3} \quad (11)$$

where C_{Ra}, C_{Th}, and C_k are the activities concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

2.8 .Excess Lifetime Cancer Risk

Excess Lifetime Cancer Risk (*ELCR*) refers to the increased likelihood of acquiring cancer over one's lifetime because of natural gamma radiation exposure to the local population caused by radionuclide concentration in the sample [5, 15, 16, 19] and calculated by the equation

$$ELCR = AEDE \times DL\left(70y\right)\times RF\left(0.05Sv^{-1}\right) \quad .(12)$$

where *AEDE* is the annual effective equivalent dose، *DL* is the life expectancy (70 years), and *RF* is the risk factor for one Sv absorbed dosage, and according to ICRP, this factor is 0.05 Sv^{-1}.

2.8.Content in (PPM) of the ^{238}U, ^{232}Th, ^{40}K

The contents in (ppm) could be calculated from the equation

$$PPM = \frac{C_{Ra}C_{Th}C_K}{WN_C \ln 2} \quad (13)$$

where W is the weight factor of the sample. Therefore, the content in ppm for ^{226}Ra, ^{232}Th, and ^{40}K are estimated, respectively, from the expressions

$$^{226}Ra\left(ppm\right) = 0.0803C_{Ra},$$

$$^{232}Th\left(ppm\right) = 0.247C_{Th},$$

$$^{40}Ra\left(ppm\right) = 3.862\times10^{-3}C_K$$

Table 2: The activity concentration of ^{226}Ra, ^{232}Th, and ^{40}K (Bq/kg) and its equivalent content (ppm)

No.	Ra-226	Th-232	K -40	Ra-226	Th-232	K-40
	Bq/kg			ppm		%
1	257.44	23.03	308.4	23.19	5.67	0.99
2	154.13	29.31	156.69	13.89	7.22	0.5
3	356.42	6.72	299.56	32.11	1.65	0.96
4	269.17	46.91	137.22	24.25	11.55	0.44
5	397.47	9.66	234.67	35.81	2.38	0.75
6	251.95	11.18	136.89	22.7	2.75	0.44
7	674.77	44.17	369.13	60.79	10.88	1.18
8	360.59	62.41	444.83	32.49	15.37	1.42
9	698.7	25.03	516.77	62.95	6.17	1.65
10	258.16	42.64	411.74	23.26	10.5	1.32
11	318.36	28.53	351.98	28.68	7.03	1.12
12	3665.19	298.23	793.26	330.2	73.46	2.53
13	6684.7	301.4	598.33	602.23	74.24	1.91
14	5654.91	137.29	849.84	509.45	33.82	2.72
15	6452.74	314.37	620.15	581.33	77.43	1.98
16	4792.68	262.5	724.79	431.77	64.66	2.32
17	5714.81	277.79	650.29	514.85	68.42	2.08
18	10349.23	504.58	1269.97	932.36	124.28	4.06
19	4678.77	369.2	857.21	421.51	90.94	2.74
20	8219.81	301.55	1157.81	740.52	74.27	3.7
21	4763.89	318.99	972.4	429.18	78.57	3.11
22	3307.95	300.81	867.49	298.01	74.09	2.77
23	5525.14	388.51	926.69	497.76	95.69	2.96
24	2938.78	140.77	575.33	264.76	34.67	1.84
25	3843.69	218.53	583.53	346.28	53.82	1.86
26	3577.01	284.59	881.89	322.25	70.1	2.82
27	4860.04	311.11	823.97	437.84	76.63	2.63
28	3814.9	257.07	810.6	343.68	63.32	2.59
29	6801.84	223.29	718.76	612.78	55	2.3
30	6370.51	164.83	957.18	573.92	40.6	3.06
31	6443.72	275.43	550.19	580.52	67.84	1.76
Ave.	3627.66	192.92	630.89	326.82	47.52	2.02
St. D.	2855.6	140.49	298.11	257.26	34.6	0.95
Max	10349.23	504.58	1269.97	932.36	124.28	4.06
Min	154.13	6.72	136.89	13.89	1.65	0.44

3. Results and Discussion

The natural radionuclides present in sediments such as ^{226}Ra, ^{232}Th and ^{40}K contribute to the amount of radiation effectively. As the most of population is exposed to that radiation in those areas, calculating the value of that radiation is importance for knowing the radiation that humans are exposed to, and then comparing these values with the standard recommended values. The radiometric measurements using NaI(TI) detectors for investigated areas Al-Zubair is shown in table (2) and figure (4). Equation (1) is used to calculate the activity concentration for radium-226, thorium-232 and potassium-40.

It is clear that the ^{226}Ra activity concentration ranged between (154.13 to 10349.23 Bq/kg) with an average of 3627.66 Bq/kg, which is higher than the world standard level of 50 Bq/kg. The thorium-232 activity concentration ranged between (82.8 to 14682.2 Bq/kg) with an average of 2752.87 Bq/kg, which is higher than the world standard level of 40 Bq/kg. Finally, the potassium-40 activity concentration ranged between 19.22 to 18553.3 Bq/kg with an average of 2773.46 Bq/kg, which is higher than the world standard average of 500 Bq/kg [1-3, 5, 12, 24]. Fig. (4) shows a significate correlation between the equivalent dose of ^{226}Ra and the annual gonadal dose due to radionuclides activity concentration. Concerning the hazard index and abovementioned radiobiological impact are calculated by utilizing the conversion factors and presented in table (3).

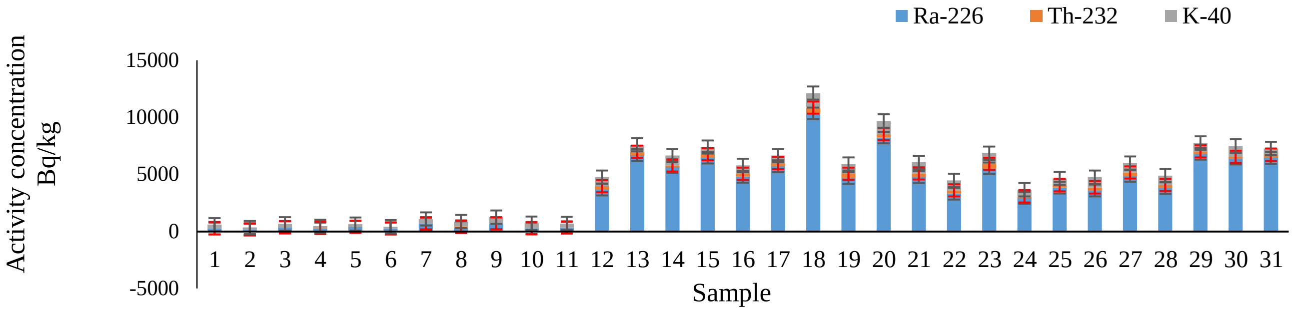

Fig. (4): Comparison between the activity concentration of Ra-226, Th-232, and K-40, respectively.

Conclusions

The pattern of radiation dispersion has been determined for natural radionuclides from Al-Zubair Petroleum station. It has showed an increase in the radiation dispersion because of ongoing deeper drilling and exploratory efforts to obtain crude oil. In pools, waste is collected and dug in preparation for long-term storage. Radiation doses were calculated using conversion dose factors suggested by the (IAEA), (ICRP), and (UNSCEAR). The obtained results for the sample from the surface to a depth of 2 km are considered normal levels compared to international reference values such as EPA and IAEA for settings comparable to the region under study. At depths of more than 1 km, where the soil layer is close to the petroleum layer, which contains an abundance of TE-NORM, the corresponding dose indicates a relatively high level of radioactivity for radium, requiring strict safety and protection precautions to protect occupational and professionals from radiation hazards. The corresponding statistical calculations related to cancer risk in the long term of life are considered to apply an effective and relevant radiation protection procedure to give calculation data related to radiation protection and human safety. Consequently, the authors recommend radiation protection and radiation safety protocols to be undated especially for the areas with deeper drilled soils.

Table 3: The calculated corresponding dose and radiological hazards due to exposure to ^{226}Ra, ^{232}Th, and ^{40}K radionuclides in Al-Zubair Petroleum Station

No.	**Dose Rate**	**Dose indoor**	**Dose outdoor**	**Total**	***AGDE***	**eq. Dose Ra-226**	**Hazard in**	**Hazard ex**	**Gamma index** I_γ	$\times 10^{-3}$ ***ELCR***
	nGy/h	**mSv/y**				**Bq/kg**				
1	14.28	0.02	0.07	0.09	95.69	31.38	0.08	0.15	0.21	0.061294
2	10.92	0.01	0.05	0.07	73.24	24.25	0.07	0.1	0.17	0.046869
3	15.9	0.02	0.08	0.1	106.44	34.55	0.09	0.18	0.23	0.068259
4	18.4	0.02	0.09	0.11	123.36	40.81	0.11	0.18	0.28	0.078965
5	18.05	0.02	0.09	0.11	120.83	39.27	0.11	0.2	0.26	0.077488
6	12.22	0.01	0.06	0.07	81.79	26.67	0.07	0.13	0.18	0.052433
7	34.89	0.04	0.17	0.21	233.69	76.44	0.21	0.37	0.51	0.149762
8	24.61	0.03	0.12	0.15	165.09	54.58	0.15	0.24	0.37	0.105655
9	32.98	0.04	0.16	0.2	220.79	71.89	0.19	0.36	0.48	0.141559
10	17.32	0.02	0.08	0.11	116.18	38.38	0.1	0.17	0.26	0.074352
11	17.66	0.02	0.09	0.11	118.35	38.82	0.1	0.18	0.26	0.075808
12	198.27	0.24	0.97	1.22	1328.15	435.43	1.18	2.07	2.94	0.851065
13	324.41	0.4	1.59	1.99	2171.79	708.53	1.91	3.54	4.76	1.392492
14	256.48	0.31	1.26	1.57	1716.41	558.02	1.51	2.88	3.74	1.100912
15	316.74	0.39	1.55	1.94	2120.58	692.21	1.87	3.44	4.65	1.359575
16	239.73	0.29	1.18	1.47	1605.17	524.41	1.42	2.58	3.53	1.029004
17	280.44	0.34	1.38	1.72	1877.53	612.85	1.66	3.05	4.12	1.203742
18	508.1	0.62	2.49	3.12	3401.77	1110.4	3	3.7	7.46	2.180966

19	251.32	0.31	1.23	1.54	1683.44	551.76	1.49	1.68	3.72	1.078782
20	388.4	0.48	1.91	2.38	2599.84	847.02	2.29	2.94	5.68	1.667166
21	247.2	0.3	1.21	1.52	1655.56	541.77	1.46	1.71	3.65	1.061089
22	183.81	0.23	0.9	1.13	1231.43	404.18	1.09	1.19	2.73	0.788979
23	289.51	0.36	1.42	1.78	1939	634.83	1.72	1.98	4.28	1.242708
24	143.92	0.18	0.71	0.88	963.6	314.48	0.85	1.05	2.11	0.617782
25	193.48	0.24	0.95	1.19	1295.57	423.39	1.14	1.38	2.85	0.830508
26	192.53	0.24	0.94	1.18	1289.65	422.71	1.14	1.28	2.85	0.826409
27	249.98	0.31	1.23	1.53	1674.06	547.62	1.48	1.74	3.69	1.073009
28	198.21	0.24	0.97	1.22	1327.47	434.43	1.17	1.37	2.93	0.850800
29	317.35	0.39	1.56	1.95	2124.09	691.6	1.87	2.43	4.64	1.362202
30	290.49	0.36	1.43	1.78	1944.08	632.21	1.71	2.28	4.23	1.246901
31	310.4	0.38	1.52	1.9	2077.91	677.66	1.83	2.3	4.55	1.332358
Ave	180.58	0.22	0.89	1.11	1209.11	394.92	1.07	1.51	2.66	0.775126
StD	137.86	0.17	0.68	0.85	922.88	301.05	0.81	1.18	2.02	0.591731
Mx	508.1	0.62	2.49	3.12	3401.77	1110.4	3	3.7	7.46	2.180966
Min	10.92	0.01	0.05	0.07	73.24	24.25	0.07	0.1	8.49	2.480156

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