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Estimation of Some Rare Earth Elements Levels in Tea in Egypt and Assessment of Associated Health Risks

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Abstract

Heavy metals are widely recognized and extensively studied as food contaminants. However, other potentially toxic elements rare earth elements (REEs), have not received as much attention. REEs present in the environment can accumulate in the human body via the food chain, posing potential health risk. Among the various sources of REEs exposure, tea consumption is notable. Despite its global popularity, there is limited knowledge regarding the REEs content across different types of tea, particular in Egypt. This study was done for revealing the levels of REEs contents in some tea products in Egypt. Cerium (Ce) was the most abundant elements in both green and red tea samples followed by Lanthanum (La). The average concentrations of REEs in red tea were higher than those for green tea. The concentration of total REEs reached up to 1695.7µg/Kg in tea samples. While, the leaching ratios were found to be ranged from 16.2% to 47.6% for soaked preparation and ranged from 28% to 51.3% for boiled preparation. Although, the high concentration of REEs in tea samples, the risk assessment analysis indicated that the estimated daily intake of REEs through tea consumption was considerably below the threshold of acceptable daily intake**.** The study advocates for the evaluation and risk assessment of a broader array of potentially toxic elements that have not yet garnered sufficient attention. Emphasizing the importance of identifies and prioritizing these elements in future research on dietary contaminants.

Keywords: REEs, Toxicity, Tea, Heath risk.

1. Introduction

Over the past few decades, there has been a significant acceleration in technological advancement. That may involve the use of many chemicals including manufacturing; this has changed the natural occurrence of many elements in the environment and the increasing accumulation of pollutants in the soil. This has resulted in the accumulation of various potentially toxic elements in the food chain, which could pose risks to both human and animal health.

Heavy elements such as arsenic, cadmium and lead are widely known and have been studied as food pollutants. Although there are a number of elements that can be toxic but have not received the same degree of attention and this can be attributed to their low toxic activity, namely the REEs **[1]**. Some research indicates that excessive exposure to rare earth oxides (REOs) can be detrimental to human health, with potential links to conditions such as leukemia **[2]**. For individuals not frequently exposed to REE-contaminated environments, dietary intake remains the primary route of exposure to REOs **[3]**. REEs are extensively utilized across various sectors such as industry, agriculture, medicine, and machinery technology. Concurrently, increasing evidence suggests that REEs present in the environment can accumulate in the human body through the food chain **[4]**. These elements tend to accumulate in bones, blood and brain tissue, where they are challenging to eliminate and may pose risks to human health **[5]**.

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Additionally animal studies have indicated that REEs could potentially cause neurotoxicity, pulmonary toxicity, nephrotoxicity and other adverse effect. High doses of REEs can lead to harmful effects, including disrupting the balance of oxidative stress and causing damage to fibrotic tissue **[6]**. A significant toxicological trait of REEs is their tendency to displace calcium from its binding sites in biological systems, which can lead to enzyme inhibition and various biochemical disturbances **[7]**. Additional potential human health effects of REEs include neurological disorders, such as diminished intelligence quotient (IQ) in children, particularly associated with Lanthanum (La), as well as alterations in bone structure **[8,9,10,11]**. As interest in REEs has only recently grown, there is currently insufficient toxicological data available for all individual REEs. Consequently, no health-based guidance values have been established.

In the results of the study **[12]** demonstrated that REEs can enter the human body through various pathways, are linked to multiple diseases, and can induce the production of reactive oxygen species (ROS), leading to DNA damage and cell death. Since 1990, rare-earth fertilizers have been extensively employed to enhance tea production and quality **[13]**.

However, when fertilizers are applied using the spraying methods, REEs tend to accumulate more significantly in tea leaves **[14]**. However, the levels of REEs in herbal teas have received comparatively less attention. It is also important to note that tea leaves exhibit elevated concentrations of REEs due to their extended growth periods and advanced maturity **[15, 16]**. Given that tea has a higher capacity for accumulating REEs compared to other major crops, and considering the use of REE-containing fertilizers in tea cultivation. It is anticipated that tea may exhibit elevated REE concentrations relative to other plants **[17]**. Tea consumption represents one pathway for dietary intake of REEs. However, the potential health risks associated with REE exposure from tea consumption are not yet fully understood.

We cannot overlook the fact that tea is one of the most popular drinks in the world and its consumption is constantly increasing, especially in Egypt. Egyptians consume about 273 billion liters of tea according to statistics from the International Tea Committee. Considering that Egypt depends entirely on importing all its tea needs from abroad and from various sources, it is necessary to evaluate the safety of the product. However, the health risks associated with rare earth element (REE) exposure among tea drinkers in Egypt remain poorly understood. Therefore, the aim of this research was to evaluate REEs contamination levels in different types of tea products in the Egyptian markets and to assess their potential dietary risks.

2. Experimental

2.1. Chemicals and reagents

- High purity deionized water DI (18 MΩ .cm) from Milli-Q system (Millipore, Bedford, MA, USA).
- Nitric acid (69%) and hydrogen peroxide (30%) were purchased from Merck (Germany)
- 13 multi-element REE standard storage solution which are: Cerium (Ce), Lanthanum (La), Neodymium (Nd), Yttrium (Y), Lutetium (Lu), Scandium (Sc), Terbium (Tb), Dysprosium (Dy), Holmium (Ho), Erbium (Er), Samarium (Sm), Europium (Eu) and Gadolinium (Gd); in addition to internal standard storage solutions and tuning solution were purchased from Agilent Technologies Inc (USA) which diluted to the concentration needed.

2.2. Sample preparation and instrumental analysis

Red tea is the most popular drink followed by green in Egypt; they were chosen to conduct this study. 30 Samples of red and green tea were collected from several markets, autonomous regions and municipalities in Egypt, which were imported from different countries of origin (China, Kenya, England and Ceylon). Three sample were chosen from each product, 5 different kind of green tea and 5 of red tea. All these samples were prepared using three different methods as follows:

First preparation method (soaked method): 2 g of each sample was soaked in 50 ml of boiling DI water for 15 minutes then filtered.

Second method (boiling method): 2 g of sample was boiled with DI water for about 5 minutes then filtered and complete to volume 50 ml.

Third method (digestion method): 2 g of sample was digested by 3 mL of HNO_3 and 1 mL H_2O_2 with the use of microwave digestion technic by analytik jena instrument **,** then volume was completed to 50 ml of DI water **[18,19]**. Blank control was made in each preparation method.

Afterwards, samples were analysed for thirteen elements of REEs by inductively coupled plasma- mass spectrometry (ICP-MS/MS Agilent 8800) according to the **[20]** method and to the instrumental manual**.** The ICP-MS-MS helium mode was used to provide lower detection limits and higher sensitivities for elements **[21]**. The 3 samples of each product were analysed as replicate and the results were expressed in mean value and standard deviation SD (n=3) **[22]**.

3. Results and discussion

As shown in Table (1) the concentrations of REEs in tea samples, which have been prepared by soaked method, varied from 0.24 to 126.6µg/kg for the tea samples. Where, Table (2) shows the results for the boiling method, where the concentrations varied from 2.59 to 189.7 µg/kg. Table (3) displays the results for the digestion method, the concentrations of the samples varied from 3.53 to 597.9µg/kg.

From the above results, it is cleared that the concentrations of samples by boiling method was higher than those by soaked method. Also, it is cleared that the samples concentrations by digestion method were higher than both soaked and boiling methods. Cerium element was the most abundant elements in both green and red tea samples followed by Lanthanum element. The study observed that red tea exhibited higher average concentrations of REEs compared to green tea. This finding aligns with previous research on REE concentrations in Oolong, black, and green teas in China. Specifically, Oolong and black teas showed the highest levels of REEs, while green and flower teas had the lowest. Among the REEs, cerium (Ce) was found to be the most abundant, with a concentration of 431.5 µg/kg, followed by lanthanum (La), yttriumm (Y), and neodymium (Nd). **[23]**. It is important to note that research indicates tea plants exhibit a distinct selectivity in their absorption of REEs, with cerium (Ce) being particularly noteworthy **[17]**.

The variation in REEs concentrations across different teas can be attributed to several factors, including the tea plant's growth cycle, the use of rare earth fertilizers, and the plant's specific absorption characteristics. **[24]**. Cerium (Ce) had the highest average content among the REEs, followed by lanthanum (La), scandium (Sc), neodymium (Nd), and yttrium (Y). Together, these elements constituted 83.16% to 86.15% of the total REEs, with lutetium (Lu) being the least abundant. Variations in the content of each element are influenced by factors such as soil composition, the type of tea, and fertilizer application. Among the sixteen elements measured, Ce was the most prevalent in flower herb tea samples, ranging from 27.3 to 3016 µg/kg, followed by La (11.2-1260 µg/kg), Nd (10.2-1100 µg/kg), Y (12.6-699 µg/kg), and Sc (15.3-305 µg/kg). Ce, La, Sc, Y, and Nd collectively represented 84.5% to 89.7% of the REEs in these samples, while Lu had the lowest concentration. **[25]**.

There is limited information on the toxicity of many REEs, and some may exhibit higher toxicity than lanthanum, potentially leading to an underestimation of risk. Nevertheless, since lanthanum, cerium, and yttrium account for 60% of the total REE contamination, the influence of other elements is relatively minor, and risk assessments based on this data are likely to be reliable. REEs in tea enter the human body through the food chain, where trace amounts may have beneficial effects on health. However, excessive intake can lead to accumulation in various tissues, potentially causing health issues such as acute myocardial infarction or leukemia **[26, 27]**. Consequently, there is growing concern regarding the health risks associated with heavy metals and REEs.

In comparison to heavy metals, the levels of REEs in herbal tea have received relatively little attention. Recent concerns have been raised about the high concentrations of REEs in tea and its infusions **[28, 29]**. The total contents of REEs and the leached ratios of each sample with respect to the total content were calculated and shown in Table (4). The leaching ratios were found to be ranged from 16.17 % to 47.56 % for soaked preparation and ranged from 28 % to51.3 % for boiled preparation. In contrast to other studies on flower herb teas, where the leaching ratios ranged from 7.3% for gobe amaranth to 29.8% for carnation [30].

The leached ratios of REEs in flowers were lower compared to most tea leaves (*Camellia sinensis L.*), which exhibited leaching rates of 25.8% to 51.0%. The leached ratios of REEs varied significantly among different flower types. In the feeding study on rats for 90 days **[31, 32]**, in cooperation with The China Scientific Committee on Food Safety established temporary acceptable daily intake (tADI) levels for three primary REEs lanthanum (51.5 µg/kg body weight), cerium (645.0 µg/kg body weight), and yttrium (145.5 µg/kg body weight) based on the no observed adverse effect level (NOAEL) related to decreased body weight gain. Additionally, adverse effects in animals included induced weight loss, reduced erythrocytes, albumin, total bilirubin, and phosphocreatine kinase, along with increased leukocytes **[33]**.

The China Scientific Committee established the lowest temporary acceptable daily intake (tADI) for lanthanum as the health-based guidance value for total REEs, following a precautionary principle. A group tADI of 51.5 µg/kg body weight was derived, with the lowest tolerable daily intake (TDI) for lanthanum at 51.3 µg/kg body weight per day serving as the reference value for all REEs. Lanthanum, being one of the most prevalent elements in analyzed teas alongside cerium and yttrium, significantly contributes to overall contamination. Consequently, an acceptable daily intake (ADI) of 51.5 µg/kg body weight per day was used for guidance in this study.

Table (1): The mean concentrations of REEs (ppb) in different tea samples prepared by soaked method with its standard deviation

Type	samples	Ce	La	Nd	Y	Lu	Sc	Tb	Dy	Ho	Er	Sm	Eu	Gd
Green tea	1	37.43±2.2	49.78±2.7	39.07±2.9	27.6±2.2	11.91±1	6.09±0.6	3.62±0.4	3.59±0.3	0.84±0.08	2.73±0.22	2.59±0.24	1.5±0.11	2.82±0.24
	2	44.84±3.5	45.11±3.1	40±3.1	28.1±1.8	8.1±0.9	3.84±0.3	1.98±0.2	1.6±0.1	0.57±0.06	1.57±0.14	1.12±0.1	0.76±0.06	1.78±0.15
	3	62.37±5.1	47.64±4.7	44.1±3.2	18.9±1.7	5.06±0.5	2.94±0.3	0.97±0.1	0.93±0.1	0.24±0.02	0.86±0.07	0.74±0.66	0.5±0.04	1.16±0.1
	4	68.22±5.2	60.98±5	48.8±2.7	26.5±1.6	5.5±0.5	3.46±0.3	3.92±0.4	2.22±0.2	0.73±0.07	2.86±0.22	1.18±0.1	0.83±0.07	2.03±0.2
	5	66.5±6	58.9±4.8	51.6±5	28.4±1.6	7.84±0.7	5.7±0.5	2.28±0.2	3.92±0.4	0.8±0.6	2.94±0.28	2.45±0.21	0.92±0.08	2.75±0.22
Red tea	6	87.27±6.3	54.55±4.2	66.90±5.4	34.5±2	3.93±0.4	2.19±0.2	1.1±0.1	0.77±0.08	0.25±0.02	0.73±0.65	0.81±0.07	0.45±0.04	0.87±0.07
	7	99.44±7.3	101.6±8.2	44.0±2.9	28.7±1.5	4.27±0.4	4.23±0.4	1.05±0.1	1.03±0.1	0.29±0.02	1.02±0.1	0.88±0.08	0.60±0.05	1.27±0.1
	8	89.9±6.1	86.9±5.8	54.6±3.2	37.5±1.9	5.83±0.5	8.91±0.9	6.64±0.5	4.77±0.5	1.54±0.1	4.97±0.4	3.73±0.33	1.23±0.1	5.32±0.48
	9	96.8±6.6	90.9±7.1	50.8±3	40.5±2.6	7.1±0.7	7.96±0.8	3.62±0.3	3.95±0.4	1.05±0.1	4.86±0.4	2.09±0.2	1.09±0.09	2.64±0.23
	10	126.63±8.4	108.4±8	56.5±3.7	47.8±2.8	13.8±1.2	9.34±0.9	4.17±0.4	3.87±0.4	2.39±0.2	5.04±0.48	3.8±0.32	2.35±0.2	4.83±0.44

Table (2): The mean concentrations of REEs (ppb) in different tea samples prepared by boiling method with its standard deviation

Type	samples	Ce	La	Nd	Y	Lu	Sc	Tb	Dy	Ho	Er	Sm	Eu	Gd
Green tea	1	111.4±9.4	60.34±5.6	58.34±5.5	23.4±2.4	17.6±1.2	16.33±2	7.38±0.7	7.45±0.8	2.91±0.4	5.59±0.6	6.6±0.7	4.12±0.5	6.85±0.7
	2	108.37±9.1	58.60±5.5	53.08±5.3	21.2±2.3	15.3±1.2	15.1±1.8	6.43±0.6	5.93±0.6	3.28±0.5	3.26±0.3	3.78±0.4	2.62±0.3	4.7±0.5
	3	109±9.1	105.73±8.7	49.9±5	18.9±2	10.3±1.1	12.4±1.3	6.83±0.7	5.74±0.6	2.59±0.4	3.89±0.4	3.95±0.5	3.73±0.5	4.97±0.51
	4	154.71±12.5	126.93±8.9	44.1±4.8	19.7±2	17.14±1.4	14.8±1.1	9.96±1	10.07±0.9	3.43±0.5	8.24±0.9	7.1±0.8	4.48±0.6	9.4±1
	5	137.8±11.3	112.7±8.2	48.6±4.7	20.8±2	16.1±1.4	14.6±1.1	8.9±0.9	8.9±0.9	3.9±0.5	9.4±1	7.9±0.83	5.8±0.7	9.98±1.2
Red tea	6	113.81±10	146.34±9.4	55.2±4.8	33.6±2.8	29.3±1.8	19.8±1.7	20±1.8	20±1.9	12.67±0.9	23.6±2	28.6±2.4	29.26±3	31.7±3.3
	7	116.47±10	132±8.3	49.04±4.7	31±2.8	22.6±1.8	18.8±1.7	17.73±1.6	14.16±1.1	13.8±1.5	16.7±1.5	17.4±1.8	14.79±1.3	18.1±2
	8	142.67±12.1	114.2±8.2	58.46±4.8	36.1±3	24.5±1.8	18.9±1.8	18.6±1.6	16.7±1.3	15.7±1.6	18.9±1.9	19.8±2	16.6±1.5	20.6±2.2
	9	189.7±17.6	128.6±8.3	60.5±5.6	38.9±3.3	34.4±2	21.2±1.8	17.8±1.6	32.68±2.9	16.8±1.8	22.5±2.7	23±2.4	21.6±2	16.8±1.8
	10	181.97±17	141.2±8.6	58.21±5.4	53.44±4.9	38.24±2.2	22.5±2	18.8±1.9	32.04±3	18.35±2	25.85±3	26.78±3	24.5±2.5	29.4±3

Table (3): The mean concentrations of REEs (ppb) in different tea samples prepared by digested method with its standard deviation

Type	samples	Ce	La	Nd	Y	Lu	Sc	Tb	Dy	Ho	Er	Sm	Eu	Gd
Green tea	1	388.6±34	216.9±19	155.2±13	113.43±10	62.04±5.3	29.75±2.7	19.12±1.8	44.45±4	9.68±0.87	31.11±3	49.75±4.2	23.89±2.1	28.6±2.6
	2	313.1±27	228±19	145.9±12	95.53±8.6	40.08±3.6	15.62±1.4	22.05±2	24.1±2	5.08±0.46	14.7±1.3	27.16±2.4	13.35±1.2	36.88±3
	3	258.4±22	189.7±16	152.5±13	105.77±9.2	67.16±5.4	11.32±1	18.9±1.7	17.55±1.5	3.53±0.3	11.5±1	21.94±2	11.5±1	30.54±2.8
	4	473.7±32	310±24	220.1±17	100.5±9	48.9±4.1	39.38±3.3	14.8±1.3	53.09±4.8	11.38±1	34.57±3	50.62±4.9	24.49±2.1	66.05±6.2
	5	455.7±31	296.7±23	285±19	87.01±7.8	55.85±4.8	28.6±2.3	17.9±1.6	44.8±4.2	10.23±1	38.1±3.3	42.1±3.9	17.3±1.6	48.4±4.3
Red tea	6	356±28	196.8±17	147.57±12	128.78±10.5	82.99±8	22.13±1.9	24.1±2.1	21.83±2	4.9±0.4	16.29±1.4	21.61±1.9	8.74±0.8	67.41±6
	7	386.2±30	218.6±19	165.76±14	113.94±9.8	73.53±6.7	15.67±1.4	22.5±2	17.97±1.6	4.02±0.36	23.93±2.1	19.5±1.8	8.29±0.78	26.55±2.3
	8	597.9±40	280.3±21	240.53±21	118.36±9.8	65.9±6	43.62±3.9	20.6±1.8	65.44±5.8	19.57±2	35.66±3.1	79.58±7.7	16.83±1.5	72.27±7
	9	497.8±37	364.7±30	288.6±22	142.11±12	88.2±8	38.34±3.5	24.9±2.1	50.78±5	14±1.1	40.2±3.8	66.4±6.1	19.88±1.7	59.8±5.6
	10	521.79±39	358.4±29	332.37±31	138.4±11.4	77.3±6.8	50.75±4.8	28.9±2.5	56.42±5.1	12.6±1	41.56±3.9	57.03±5.1	15.83±1.5	77.35±7.1

Table (4): The total contents of REEs for each prepared method and the leached ratios of the samples

		Total of REEs (ppb)			Leaching (%)	
Type	samples	soaked	boiled	Digested	soaked	boiled
Green tea	1	189.57	328.31	1172.52	16.17	28.00
	2	179.37	301.65	981.55	18.27	30.73
	3	186.41	337.93	900.31	20.71	37.53
	4	227.23	430.06	1298.18	17.50	33.13
	5	235	405.38	1427.69	47.56	28.39
Red tea	6	254.32	563.88	1099.15	23.14	51.30
	7	288.38	482.59	1096.46	26.30	44.01
	8	311.84	521.73	1656.56	18.82	31.49
	9	313.36	624.48	1695.71	43.53	36.83
	10	388.92	671.28	1553	25.04	43.22

The exposure assessment integrated data on REEs **[18]** with consumption data from a survey of Danish consumers, defined as individuals who drink at least one cup of tea per week. The average daily consumption was estimated at 350 mL, with high consumption reaching 1,084 mL per day.

For the risk assessment, an average body weight of 70 kg was used. Additionally, brand loyalty in tea consumption could result in prolonged exposure to high levels of contaminants if the product is significantly contaminated**.**

The hazard quotient (HQ) was calculated to assess whether health risks are acceptable. An HQ value less than one indicates no significant health risk, while an HQ value greater than one suggests potential health risks, with the probability of adverse effects increasing as HQ values rise **[34]**.

The hazard equation (HQ) was estimated using the formula: HQ=ADD/ADI

Where, ADI represents the acceptable daily intake (µg/kg body weight /day),

ADD The average daily dose (µg/kg body weight/day) was calculated using: ADD =Σ ([REEs]*IR*LR) / BW/1000

In this equation, [REEs] is the mean concentration of total REEs (µg/kg) in the tea category, IR is the intake rate of the tea (g/day), LR is the leaching rate of the tea, BW is the individual's body weight (kg).

The hazard quotient (HQ) results, presented in Table (5), indicated that the estimated daily intake of REEs from drinking tea was significantly below the acceptable daily intake for both types of tea. This finding aligns with results that reported, the risk of REE exposure from drinking green tea, even with high REE content, was negligible.

Table (5): The hazard quotient (HQ) estimated daily intake of REEs

hazard quotient (HQ)			
Type	**samples**	**soaked**	**boiled**
Green tea	1	0.009	0.026
	2	0.009	0.026
	3	0.011	0.035
	4	0.011	0.040
	5	0.031	0.092
Red tea	6	0.016	0.080
	7	0.021	0.059
	8	0.016	0.046
	9	0.038	0.150
	10	0.027	0.080

4. Conclusions:

Despite the elevated concentration of REEs in tea samples, the health risk associated with REEs exposure from tea consumption are considered acceptable. Although the above, it is important to avoid excessive tea consumption. Where, consumption of large quantities of tea over long term may lead to the accumulating of these elements within body, potentially leading to serious significant health risk. The study supports priorities future work concerning the surveying and risk assessment of a broad range of elements which may have toxic effect on food.

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