Dietary Content and Evaluation of Metals in Four Types of Tea (White, Black, Red and Green) Consumed by the Population of the Canary Islands

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Abstract

Objective: The aim of this study was to analyze the contents of Cd, Co, Cu, Fe, Mg, Ni, Pb, Zn in samples of four types of tea (Camellia sinensis) consumed in the Canary Islands: white, black, red and green tea, in order to determine their intake through consumption.

Methods: A total of 80 samples (20 of each type of tea) were analyzed by ICP-OES.

Results: The highest concentrations of Cd, Co, Mg, Pb and Zn were found in samples of white tea, those of Cr, Fe and Ni in the red tea, and those of Cu in green tea. While the lowest levels of Cd, Co, Fe, Pb and Zn were found in black tea, those of Cr, Mg and Ni in the green tea, and those of Cu in the white tea.

Conclusion: The calculated intakes of each metal (assuming the consumption of a cup of tea 2 g/day) shows that Cr, Cu, Fe, Mg and Zn make a negligible contribution to the RDI, and the same applies to Cd to the PTMI and Pb to the PTWI.

Keywords: Metals; ICP-OES; Dietary assessment; Spanish population; Canary Islands

Introduction

Tea or blends of tea are understood to be the buds, shoots and young leaves of the shrub Camellia sinensis (Theaceae) [1]. This shrub of Asian origin has been cultivated since ancient times [2] and is currently being grown in much of Asia, Africa, Turkey and other countries. The different types of tea are determined by a number of factors such as the time of harvest (depending on leaf development), the processing method, drying, roasting and fermentation processes [3].

The consumption of tea has been estimated to be about 18 to 20 billion cups of tea a day worldwide, meaning that it is the second most consumed beverage after water in the world [4,5].

Note that the four types of tea which are the objects of this study (white, black, red and green) have beneficial properties for human health, thus, black tea has antioxidant properties [6], hepatoprotective [7], antiarteriosclerotic [8], lipolytic, thermogenic [9,10], and anticarcinogenic [11]. Antioxidant properties are attributed to green tea [12-14], antiarteriosclerotic [15], hepatoprotective [16], lipolytic, thermogenic [17,18], anticarcinogenic [19,20] and reducing platelet aggregation [21]. White tea has neuroprotective properties [22,23], anticarcinogenic [24,25], protection from ultraviolet radiation when used topically [26] and inhibition of acetylcholinesterase [27]. Red tea has antioxidant features [28], antiarteriosclerotic [8], lipolytic, thermogenic [29] and anticarcinogenic [30].

Although metals are perhaps the oldest known most toxic agents, interest in them has not declined and knowledge concerning their potential toxic effects and mechanisms of action has increased in recent years [31]. Metals such as iron, copper and zinc are considered essential minerals for humans, although high concentrations could be potentially toxic [32-34]. Heavy metals, like lead and cadmium, are regarded as toxic environmental contaminants in food [35]. Compared with other toxic substances, heavy metals are considered to be most damaging to living systems. Food and water are the primary source of these metals for humans [36,37].
in the biosynthesis of certain proteins like collagen and elastin [42,43].

Magnesium is a crucial element in the generation and use of adenosine triphosphate, and is required for oxidative phosphorylation. It is involved in energy metabolism, glucose utilization, protein synthesis, synthesis and degradation of fatty acids, ATPase functions, hormonal reactions, neuromuscular transmission signalling and cardiovascular health. Magnesium is also associated with the maintenance of cellular ionic balance through its relationships with sodium, potassium and calcium [44-46].

Although nickel is an essential trace element for various animal species, its biochemical role in humans and higher animals has not been demonstrated. However, it may serve as a cofactor or structural component of several metalloenzymes with a variety of functions (hydrolysis, redox reactions) and in gene expression [34,47].

Zinc is involved in biochemical processes, such as cell respiration and the use of oxygen by cells, both DNA and RNA synthesis, the preservation of the cell membrane integrity, and the elimination of free radicals, a process executed through a cascade of enzymatic systems [42,48].

Cadmium and lead are not essential for the human organism [34]. Cadmium has a long residence time in human tissues (10-40 years), especially in the kidneys and liver, where it bio-accumulates [49,50] and its toxic effects are noticeable in various ways. It can interfere with some of the organism’s enzymatic reactions, substituting zinc and other metals, manifesting its action in several pathological processes such as renal dysfunctions, hypertension, arteriosclerosis, inhibition of growth, damage in the nervous system, bone demineralisation and endocrine disruption [37]. The main toxic effect of lead is the dysfunction of the nervous system in the foetus and infants. In adults, lead causes: adverse blood effects, reproductive dysfunctions; damage to the gastrointestinal track; nephropathies; damage to the central as well as the peripheral nervous system and interferences in the enzymatic systems that synthesise the HEME group [37].

Therefore, and assuming that food consumption is the main route of exposure of the general population to metals, that tea is a highly consumed food, that its metallic levels and the intake these are responsible for is unknown in the Canary islands, the main objective of this work has been to determine the concentrations of Cd, Co, Cr, Cu, Fe, Mg, Ni, Pb and Zn in tea (white tea, black, red and green) and to discover the dietary intakes resulting from its consumption.

**Material and Methods**

**Samples**

A total of 80 samples of different brands of tea sold on the island of Tenerife were analyzed: 20 black tea, 20 green tea, 20 red tea and 20 white tea. The samples were purchased in supermarkets, health food shops and pharmacies for four months (between January and April 2014).

Before sample preparation, all laboratory materials were washed with Acationox laboratory cleaning agent (Merck, Darmstadt, Germany) to avoid contamination and eliminate any possible trace with Acationox laboratory cleaning agent (Merck, Darmstadt, Germany) and the temperature was gradually increased (50°C / hour) until it reached 425 ± 15°C, this temperature was maintained for 18-24 hours to destroy the organic matter in the sample. The white ashes obtained by this procedure were dissolved in 1.5% nitric acid to a final volume of 25 ml.

The digested samples were transferred into polyethylene containers and stored for a maximum of one month until analysis. The elements in the samples were determined by ICP-OES using a Thermo Scientific iCAP 6000 series spectrometer (Waltham, MA, USA). This reference technique for metal determination is highly sensitive with excellent data reproducibility [34]. The settings were as follows: approximate RF power, 1.2 kW; gas flow (nebulizer flow; auxiliary flow), 0.5 L/min; pump rate, 50 rpm; stabilization time 0 s. All analyzes were performed in duplicate.

The quality controls used in this work were chosen by the criterion of method accuracy. This was established by the average recovery obtained with reference material measured under reproducible conditions. The reference materials SRM 1515 Apple Leaves and SRM 1573a Tomato Leaves from the National Institute of Standards and Technology (NIST) were used. Recovery rates obtained from the reference materials were over 94.4% (Table 1). Instrumental detection and quantification limits in terms of reproducibility were calculated as three and ten times the standard deviation (SD) resulting from the analysis of 15 targets of acid digest [51] and are shown in table 2.

**Statistical analysis**

The SPSS 22.0 software (IBM, USA) was used for the statistical analysis of the results. The normality of the data was checked using the Kolmogorov-Smirnov and Shapiro-Wilk [52], while the Levene test was used to check homogeneity of variance [53]. Given that the results of these tests showed neither normality nor homogeneity of variances, it was decided to perform a non-parametric statistical test (Kruskal-Wallis) to check whether there were significant differences between the studied samples and the Mann Whitney U test to clarify between which types of tea there were differences. P values<0.05 were considered statistically significant.

**Results and Discussion**

The mean concentrations of the metals which were the object of this study (wet weight) and their corresponding standard deviation are shown in Table 3.

Cadmium concentrations ranged from 38.20 mg/kg in white tea and the levels in black tea were below the detection limit (<2.5 mg/kg). The maximum concentration for cobalt of this element, 0.70 mg/kg, was found in white tea and the minimum, 0.44 mg/kg, was in black tea, although very similar to those in the red and black teas, 0.45 and 0.46 mg/kg, respectively. In the case of chromium, 2.56 mg/kg was the maximum content which was found in the red tea and the minimum was 0.26 mg/kg which was found in the green tea. The highest copper content was found in the green tea (8.40 mg/kg) and the lowest in the white tea (5.46 mg/kg). With respect to iron, the red and black teas had the greatest and least amounts of this element (227.43 and 90.00 mg/kg, respectively). The highest concentration of magnesium was detected in the white and that of nickel in the red tea, (142.33 mg/kg of magnesium
and 3.87 mg/kg of nickel), but the lowest amounts of these metals were found in the green tea (38.13 and 2.31 mg/kg respectively). Finally, as for lead and zinc, the largest quantities of both these elements were observed in the white tea (154.05 mg/kg and 7.35 mg/kg, respectively) and the smallest quantities in the black tea (26.00 mg/kg and 5.04 mg/kg, respectively).

Therefore, cadmium, cobalt, magnesium, lead and zinc were found to be present in the greatest quantities in the samples of white tea, while chromium, iron and nickel had the greatest presence in the red tea, and copper in the green tea. The lowest levels of cadmium, cobalt, iron, lead and zinc were observed in the black tea, chromium, magnesium and nickel levels were lowest in the green tea, and copper in the white tea.

It is noteworthy that, in some of the analyzed teas, the variability of the results is high. However, this variability in biological samples is considered normal, since the metal content in foods, both plants and animals, depends on various factors ranging from environmental conditions to the production and processing methods [54].

Significant differences were observed for the metal concentrations depending on the type of tea. In the case of cadmium, white tea and red tea had significantly greater differences (p<0.05) compared to black tea and green tea. In the case of cobalt, white tea differed significantly (p<0.05) from black, red and green tea. As regards chrome, red tea was different from white tea, and they in turn differed from black tea and green tea (p<0.05). With regard to copper content, this was significantly higher (p<0.05) in red and green tea when compared to black and white tea, and similar to that observed for iron, although in this case, red tea was differentiated from black tea, and both of these in turn from white tea and green tea (p<0.05). As for magnesium concentrations, those in the white tea differed from those in the red tea, and both of them differed from the black and green teas (p<0.05). Red tea had

### Table 1: Quality control of the method.

| Element and wavelength | Detection limit (LOD) (mg/l) | Quantification limit (LOQ) (mg/l) |
|------------------------|-------------------------------|----------------------------------|
| Cd (226.5 nm)          | 0.0003                        | 0.001                            |
| Co (228.6 nm)          | 0.0006                        | 0.002                            |
| Cr (267.7 nm)          | 0.003                         | 0.008                            |
| Cu (327.3 nm)          | 0.004                         | 0.012                            |
| Fe (259.9 nm)          | 0.003                         | 0.009                            |
| Mg (279.1 nm)          | 0.583                         | 1.943                            |
| Ni (231.6 nm)          | 0.0007                        | 0.003                            |
| Pb (220.3 nm)          | 0.0003                        | 0.001                            |
| Zn (206.2 nm)          | 0.002                         | 0.007                            |

LOQ: Quantification limit

### Table 2: Detection and quantification limits.

| Element and wavelength | White (µg/Kg, wet weight) | Black (µg/Kg, wet weight) | Red (µg/Kg, wet weight) | Green (µg/Kg, wet weight) |
|------------------------|---------------------------|---------------------------|-------------------------|---------------------------|
| Cd (µg/Kg, wet weight) | 38.20 ± 50.12             | <LOQ                      | 31.00 ± 27.15           | 7.75 ± 13.88              |
| Co (mg/Kg, wet weight) | 0.70 ± 0.98               | 0.44 ± 0.09               | 0.45 ± 0.16             | 0.48 ± 0.20               |
| Cr (mg/Kg, wet weight) | 1.12 ± 0.38               | 0.38 ± 0.15               | 2.56 ± 0.54             | 0.26 ± 0.08               |
| Cu (mg/Kg, wet weight) | 5.46 ± 1.32               | 5.64 ± 1.19               | 7.76 ± 1.33             | 8.40 ± 4.5                |
| Fe (mg/Kg, wet weight) | 90.00 ± 28.85             | 33.29 ± 13.72             | 227.43±68.39            | 65.66 ± 26.77             |
| Mg (mg/Kg, wet weight) | 142.33±103.22             | 58.03±38.53               | 99.99±41.85             | 38.13±19.79               |
| Ni (mg/Kg, wet weight) | 3.38 ± 0.86               | 2.82 ± 0.53               | 3.87 ± 0.48             | 2.31 ± 0.54               |
| Pb (µg/Kg, wet weight) | 154.05 ± 183.74           | 26.00 ± 25.93             | 102.40±47.51            | 38.90 ± 22.92             |
| Zn (mg/Kg, wet weight) | 7.35 ± 1.60               | 5.04 ± 0.80               | 7.20 ± 1.06             | 5.16 ± 1.11               |

1LOQ: Quantification limit

### Table 3: Cd, Co, Cr, Cu, Fe, Mg, Ni, Pb and Zn contents in the different types of tea: mean ± standard deviation.
significantly higher concentrations of nickel (p<0.05) with respect to the white, green and black teas. In case of lead, red tea and white tea are significantly differentiated by their higher lead contents (p<0.05) with respect to the black and green teas and finally the zinc contents in the white tea and the red tea were significantly differentiated from the black tea and the green tea (p<0.05).

A correlation study was also performed using the Pearson correlation test, yielding the following positive correlations between elements: Cr / Fe, Cr / Ni, Cr / Zn, Cr / Mg, Cr / Cd, Co / Ni, Co / Zn, Co / Mg, Fe / Cr, Fe / Cu / Ni, Fe / Zn, Fe / Cd, Ni / Zn, Ni / Mg, Ni / Cd, Ni / Pb, Zn / Mg, Zn / Cd, Zn / Pb / Mg / Cd, Mg / Pb, Cd / Pb and the only negative correlation was between Co / Cu (Table 4).

The comparison of the concentrations of metals in this study with those of other authors, could only be carried out in the case of black and green tea, because in the cases of white and red tea no references from other studies were found to perform the said comparison. Table 5 shows the comparison of the metal concentrations in black tea obtained in this study with other previous studies of metals in black tea. It is observed that the cadmium levels obtained in the present study (<0.0025 mg/kg), are lower compared to those obtained by the other studies, particularly with regard to teas from Saudi Arabia (1.1 mg/kg) [3]. Cobalt levels (0.44 ± 0.09 mg/kg) are consistent with other studies, except for the case of tea from Saudi Arabia (10.98 mg/kg) [3] and Turkey (14.5 ± 7.1 mg/kg) [56], which are in the order of 25 and 33 times higher, respectively. The values of chromium (0.38 ± 0.15 mg/kg) are much lower than those observed by other studies, and were up to 34 times lower than the tea from Turkey (13.0 ± 1.7 mg/kg) [56]. The concentration of copper in the present study (5.64 ± 1.19 mg/kg) is about 18 times greater than that reported in the study on teas produced in Taiwan (7.92 mg/kg) [57], lower than the other works, and was up to 5 times lower than the value reported by Matsuura et al., in 2001 in Japan (27.7 ± 0.7 mg/kg). The iron levels (33.29 ± 13.72 mg/kg) are only greater than those from Taiwan (0.9 mg/kg), about 37 times higher, but lower than for the other authors. As regards magnesium concentrations, the levels of the present study (58.03 ± 38.53 mg/kg) are lower than those described by Matsuura et al., in 2001, in Japan (2070 ± 30 mg/kg) and Shen and Chen, in 2008, in Taiwan (135.3 mg/kg). In the case of nickel, the levels of the study here (2.82 ± 0.53 mg/kg) are in line with other authors, except for Saudi Arabian tea (16.8 mg/kg) and Turkish tea (23.3 ± 9.6 mg/kg). The lead levels reported in the present work are low (0.026 ± 0.0259 mg/kg), particularly when compared with those of Turkish tea (17.9 ± 7.1 mg/kg) [56] (approximately 688 times higher). The same applies in the case of zinc, the level reported in the teas by Narin et al., in 2004, Turkey (129.0 ± 12.9 mg/kg) are much higher than those found here (5.04 ± 0.81 mg/kg).

The comparisons of the metal concentrations in green tea obtained in this study with those of previous studies are shown in Table 6. The levels of cadmium and cobalt found here are very similar to those of other studies. The chromium concentration in Thai tea (1.476 mg/kg) [32] is 6 times higher than those consumed in the Canary Islands (0.26 ± 0.08 mg/kg). The results for copper are similar, where the copper levels reported by Nookabkaew et al., in 2006, also in Thailand (15.20 mg/kg), are 2 times higher than those consumed in the Canary Islands (8.40 ± 4.5 mg/kg) although Taiwanese tea (0.4 mg/kg) [57] has copper concentrations which are 21 times lower. The iron concentration found in the present study (65.66 ± 26.77 mg/kg) is much higher than that reported by Shen and Chen in 2008 in Taiwan (0.6 mg/kg), but lower than that of other authors. The magnesium levels in the study (38.13 ± 19.79 mg/kg) are much lower than those from Thailand [32] and Japan [59] which were 2017 and 2200 ± 60 mg/kg, respectively. The nickel concentration reported here is similar to that of other authors, with that of lead being lower. Specifically, the lead levels reported in the teas from Thailand [32] are 103 times higher than those found in the present study (3.930 mg/kg versus 0.03890 ± 0.02292). The zinc levels found here (5.16 ± 1.11 mg/kg) are similar to those described by Shen and Chen, 2008, in Taiwan (6.3 mg/kg) but lower than the rest.

In order to calculate the intakes of the metals in this work, the percentage of each metal transferred to the infusion of tea according to the references consulted has been taken into account (Table 7). Since transfer data were not found in the literature consulted here for all of the types of tea studied, a value of 100% was given to the metal transfer in the cases where data was not found. This procedure was performed in such a way, by assuming the highest transfer value, in order to consider the case of maximum exposure to the metals studied for tea consumption. In other words, to consider the cases of maximum intake. The aim of this measure is to provide the maximum protection for the health of consumers. In cases where more than one transfer data for the same type of tea has been found and, also and as in the previous case, the highest consulted transfer value has been chosen to protect the health of consumers.

The total intakes of cadmium, cobalt, chromium, copper, iron, magnesium, nickel, lead and zinc from the consumption of black, green, white and red tea in the Canary Islands are shown in Table 8. The calculation of the intakes was based on a daily consumption of tea prepared with 100 ml of boiling distilled water in which 2 g of tea were brewed for 5-10 minutes. Distilled water was used in the preparation of the different types of tea to prevent any interference from metals that may be present in drinking water.

The white tea provided the greatest intake of cadmium (0.0764 µg/day). The maximum intake of cobalt (1.4 µg/day) came from the white tea and the minimum (0.0226 µg/day) from the black tea. The greatest chromium intake was 5.12 µg/day from the red tea and the lowest was 0.0645 µg/day from the green tea. The red tea provided the largest intake of copper (15.52 µg/day) and the lowest intake of copper came from the black tea (2.4590 µg/day). The red and the green tea supplied the highest and lowest intakes of iron (14.3139 and 454.86 µg/day, respectively). The highest intakes of magnesium and lead came from the white tea (284.66 and 0.3081 µg/day, respectively), and lowest from the green tea (26.380 and 0.0055 µg/day, respectively). Red tea provided the highest intake of nickel (7.74 µg/day) and the lowest nickel intake came from the black tea (2.8476 µg/day). Finally, the highest intake of zinc was from the white tea (14.7 µg/day), and this was similar to the zinc provided by the red tea (14.4 µg/day), while the black tea provided the lowest zinc intake (0.9677 µg/day).

If the average consumption of tea by the Canary population is one cup (2 g) per day, the contribution of these teas to the Dietary Reference Intakes (DRI) of chromium, copper, iron, magnesium and zinc should be taken into account. Although the DRI are dietary recommendations, both Canadian and American, to evaluate the intakes of the metals studied in the food groups of this work, they were selected here because they are the most recent established dietary guidelines.

The DRIs of chromium, copper, iron, magnesium and zinc for adults (men and women) are: Cr 20-35 µg/day, Cu 700-900 µg/day, 8-18 mg (8000-18000 µg) of Fe/day, 240-420 mg (240,000 to 420,000 µg) of Mg/day, 8-11 mg (8000-11000 µg) of Zn/day [60]. Therefore, the contributions to the DRIs (in percentage terms) involving established intakes of chromium, copper, iron, magnesium and zinc from the
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| Metal | Population | Reference | Concentration (mg/kg) |
|-------|------------|-----------|-----------------------|
| Cr    | The Canary Islands | The present study | <LOQ* |
|       | Japan      | [55]      | 0.018 ± 0.0003        |
|       | Turkey     | [56]      | 2.3 ± 0.4             |
|       | Taiwan     | [57]      | 0.07                  |
|       | Saudi Arabia | [3]      | 1.1                   |
|       | India      | [33]      | 0.14                  |
|       | Pakistan   | [58]      | 0.0121                |
| Co    | The Canary Islands | The present study | 0.44 ± 0.09 |
|       | Japan      | [55]      | 0.506 ± 0.042         |
|       | Turkey     | [56]      | 14.5 ± 7.1            |
|       | Taiwan     | [57]      | 0.2                   |
|       | Saudi Arabia | [3]      | 10.98                 |
|       | India      | [33]      | 4.76                  |
|       | Pakistan   | [58]      | 1.14                  |
| Cr    | The Canary Islands | The present study | 0.38 ± 0.15 |
|       | Turkey     | [56]      | 13.0 ± 1.7            |
|       | Taiwan     | [57]      | 7.92                  |
|       | Saudi Arabia | [3]      | 9.8                   |
|       | India      | [33]      | 4.76                  |
|       | Pakistan   | [58]      | 12.63                 |
| Cu    | The Canary Islands | The present study | 5.64 ± 1.19 |
|       | Japan      | [55]      | 27.7 ± 0.7            |
|       | Turkey     | [56]      | 16.5 ± 3.9            |
|       | Taiwan     | [57]      | 0.3                   |
|       | Saudi Arabia | [3]      | 18.1                  |
|       | India      | [33]      | 24.07                 |
|       | Pakistan   | [58]      | 21.39                 |
| Fe    | The Canary Islands | The present study | 33.29 ± 13.72 |
|       | Japan      | [55]      | 134 ± 4.8             |
|       | Taiwan     | [57]      | 0.9                   |
|       | Saudi Arabia | [3]      | 250.46                |
|       | Pakistan   | [58]      | 118.46                |
| Mg    | The Canary Islands | The present study | 58.03 ± 38.53 |
|       | Japan      | [55]      | 2070 ± 30             |
|       | Taiwan     | [57]      | 135.3                 |
| Ni    | The Canary Islands | The present study | 2.82 ± 0.53 |
|       | Japan      | [55]      | 8.06 ± 0.19           |
|       | Turkey     | [56]      | 23.3 ± 9.6            |
|       | Saudi Arabia | [3]      | 16.8                  |
|       | India      | [33]      | 2.53                  |
|       | Pakistan   | [58]      | 6.78                  |
| Pb    | The Canary Islands | The present study | 0.026 ± 0.0259 |
|       | Japan      | [55]      | 0.709 ± 0.020         |
|       | Turkey     | [56]      | 17.9 ± 7.1            |
|       | Taiwan     | [57]      | 2.01                  |
|       | Saudi Arabia | [3]      | 1                     |
|       | India      | [33]      | 0.81                  |
|       | Pakistan   | [58]      | 0.37                  |

*: p<0.05  **: p<0.01

Table 4: Pearson co-relation test.
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| Metal | Population | Reference | Concentration (mg/kg) |
|-------|------------|-----------|----------------------|
| Cd    | The Canary Islands | The present study | 0.00775 ± 0.01388 |
|       | Japan      | [59]      | 0.0405 ± 0.0041      |
|       | Thailand   | [32]      | ND                  |
|       | Taiwan     | [57]      | 0.035               |
| Co    | The Canary Islands | The present study | 0.45 ± 0.20 |
|       | Japan      | [59]      | 0.226 ± 0.004       |
|       | Thailand   | [32]      | 0.294               |
|       | Taiwan     | [57]      | 0.7                 |
| Cr    | The Canary Islands | The present study | 0.26 ± 0.08 |
|       | Thailand   | [32]      | 1.476               |
|       | Taiwan     | [57]      | 0.1                 |
| Cu    | The Canary Islands | The present study | 8.40 ± 4.5 |
|       | Japan      | [59]      | 10.5 ± 0.2          |
|       | Thailand   | [32]      | 15.20               |
|       | Taiwan     | [57]      | 0.4                 |
| Fe    | The Canary Islands | The present study | 65.66 ± 26.77 |
|       | Japan      | [59]      | 112 ± 5             |
|       | Thailand   | [32]      | 167.1               |
|       | Taiwan     | [57]      | 0.6                 |
| Mg    | The Canary Islands | The present study | 38.13 ± 19.79 |
|       | Japan      | [59]      | 2200 ± 60           |
|       | Thailand   | [32]      | 2017                |
|       | Taiwan     | [57]      | 175.9               |
| Ni    | The Canary Islands | The present study | 2.31 ± 0.54 |
|       | Japan      | [59]      | 4.65 ± 0.19         |
|       | Thailand   | [32]      | 5.633               |
| Pb    | The Canary Islands | The present study | 0.03890 ± 0.02292 |
|       | Japan      | [59]      | 0.734 ± 0.021       |
|       | Thailand   | [32]      | 3.930               |
|       | Taiwan     | [57]      | 0.01                |
| Zn    | The Canary Islands | The present study | 5.16 ± 1.11 |
|       | Japan      | [59]      | 28.1 ± 0.8          |
|       | Thailand   | [32]      | 32.17               |
|       | Taiwan     | [57]      | 6.3                 |

1LOQ: Quantification limit

Table 5: Comparison of the Cd, Co, Cr, Cu, Fe, Mg, Ni, Pb and Zn concentrations in black tea: population of the Canary Islands against other populations.

| Metal | Tipo de té | Porcentajes de transferencia |
|-------|------------|-------------------------------|
| Cd    | Black      | 40.3% [57]                    |
|       | Green      | 14.18% [32]                   |
|       | White      | 100%                          |
|       | Red        | 100%                          |
| Co    | Black      | 25.3% [57]                    |
|       | Green      | 49.27% [32]                   |
|       | White      | 59.3% [57]                    |
|       | Red        | 100%                          |

1ND: Not detected

Table 6: Comparison of the Cd, Co, Cr, Cu, Fe, Mg, Ni, Pb and Zn concentrations in green tea: population of the the Canary Islands against other populations.
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| Metal | Type of tea | Intake (µg/day) |
|-------|-------------|-----------------|
|       | Black       | 67.5% [57]      |
|       | Green       | 11.45% [32]     |
|       | White       | 100%            |
|       | Red         | 100%            |
|       | Black       | 21.8% [57]      |
| Cu    | Green       | 12.96% [32]     |
|       | White       | 100%            |
|       | Red         | 100%            |
|       | Black       | 30.9% [57]      |
| Fe    | Green       | 2.39% [32]      |
|       | White       | 100%            |
|       | Red         | 100%            |
| Mg    | Black       | 50.6% [57]      |
|       | Green       | 34.26% [32]     |
|       | White       | 100%            |
|       | Red         | 100%            |
|       | Black       | 50.49% [57]     |
| Ni    | Green       | 67.71% [32]     |
|       | White       | 100%            |
|       | Red         | 100%            |
| Pb    | Black       | 58.6% [57]      |
|       | Green       | 7.11% [32]      |
|       | White       | 100%            |
|       | Red         | 100%            |
|       | Black       | 9.6% [57]       |
| Zn    | Green       | 32.15% [32]     |
|       | White       | 100%            |
|       | Red         | 100%            |

Table 7: Percentages of metals transferred to the brewed tea.
consumption of 2 g of black, green, white and red tea per adult and day are shown in Table 9.

Furthermore, considering the abovementioned average consumption (one cup of tea 2 g/day), the contributions from the daily intake of the types of tea evaluated in this work to the Provisional Tolerable Monthly Intake (PTMI) of cadmium and Provisional Tolerable Weekly Intake (PTWI) of lead have been studied. In the case of cadmium, and because of its long half-life, the daily food intake has a small or even a negligible effect on the overall exposure. Therefore, and in order to assess the risks in the short or long term, the intake should be evaluated over a period of months, specifically, for at least one month. Thus, the Joint Expert Committee on Food Additives of FAO / WHO decided to set the tolerable upper intake level of this metal as a monthly value of 25 micrograms / kg body weight (3.17 mg/day for an adult of 70 kg) [61]. The PTWI value for lead will be 25 mg/kg body weight (250 mg/day for an adult of 70 kg), although since 2011 this value has been outdated and we are waiting for the Committee to set a new PTWI value that is considered protective for people’s health [61]. The contributions to the PTMI and PTWI for cadmium and lead (in percentage terms) are also shown in Table 9.

According to the above, a daily consumption of one cup of tea (2 g) by an adult makes a negligible contribution of the metals studied here, not only to the RDI, but also to the PTMI and PTWI. However, it is noteworthy that although the daily consumption of tea in the Canary population is relatively low, it should be borne in mind that there are other sources that may provide cadmium, chromium, copper, iron, magnesium, lead and zinc.

**Conclusion**

Iron was the most abundant metal in the four types of tea studied in the case of red tea, with a mean value of 227.43 ± 68.39 mg/kg, while the cadmium was the least abundant in black tea, whose value was not quantifiable. The intakes of chromium, copper, iron, magnesium and zinc found from the consumption of 2 g of black tea, green, white and red per adult per day, contribute little to the DRIs, with the maximum contribution being that of the chromium in the case of red tea, assuming 100% metal transfer to the brewed tea. The toxicological analysis revealed that the same consumption of 2 g of black, green, white and red tea per adult per day, contributes between 0069-1.956 % of the PTMI of Cd and between 0002-0123% of the PTWI of Pb to an average person of 70 Kg. One, therefore, cannot suspect that there is a

| Metal | Type of tea | Intake (µg/day) | Contribution to the DRI (%) |
|-------|-------------|-----------------|----------------------------|
| Cr    | Black       | 0.5130          | 1.466-2.565                |
|       | Green       | 0.0645          | 0.184-0.322                |
|       | White       | 2.2400          | 6.400-11.200               |
|       | Red         | 5.1200          | 14.629-25.600              |
| Cu    | Black       | 2.4590          | 0.273-0.351                |
|       | Green       | 3.8472          | 0.427-0.550                |
|       | White       | 10.9200         | 1.213-1.560                |
|       | Red         | 15.5200         | 1.724-2.217                |
| Fe    | Black       | 20.5732         | 0.114-0.257                |
|       | Green       | 14.3139         | 0.080-0.179                |
|       | White       | 180.0000        | 1.000-2.250                |
|       | Red         | 454.8600        | 2.527-5.686                |
| Mg    | Black       | 58.7264         | 0.014-0.024                |
|       | Green       | 26.3860         | 0.006-0.011                |
|       | White       | 284.6600        | 0.068-0.119                |
|       | Red         | 199.9800        | 0.048-0.083                |
| Zn    | Black       | 0.9677          | 0.009-0.012                |
|       | Green       | 6.2642          | 0.057-0.078                |
|       | White       | 14.7000         | 0.134-0.184                |
|       | Red         | 14.4000         | 0.131-0.180                |

| Metal | Type of tea | Intake (µg/day) | Contribution to the PTMI (%) |
|-------|-------------|-----------------|----------------------------|
| Cd    | Black       | -               | -                          |
|       | Green       | 0.0022          | 0.069                      |
|       | White       | 0.0764          | 2.410                      |
|       | Red         | 0.0620          | 1.956                      |
| Pb    | Black       | 0.0305          | 0.012                      |
|       | Green       | 0.0055          | 0.002                      |
|       | White       | 0.3081          | 0.123                      |
|       | Red         | 0.2048          | 0.082                      |
Declarations of Interest

Dr. Dailos González-Weller, corresponding author of the manuscript titled "Dietary content and evaluation of Cd, Co, Cr, Cu, Fe, Mg, Ni, Pb and Zn in four types of tea (white, black, red and green) consumed by the population of the Canary Islands" declares that he has not received any financial, consulting, and personal relationships with other people or organizations, writing assistance, grant support and numbers, and statements of employment that could influence this work.

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