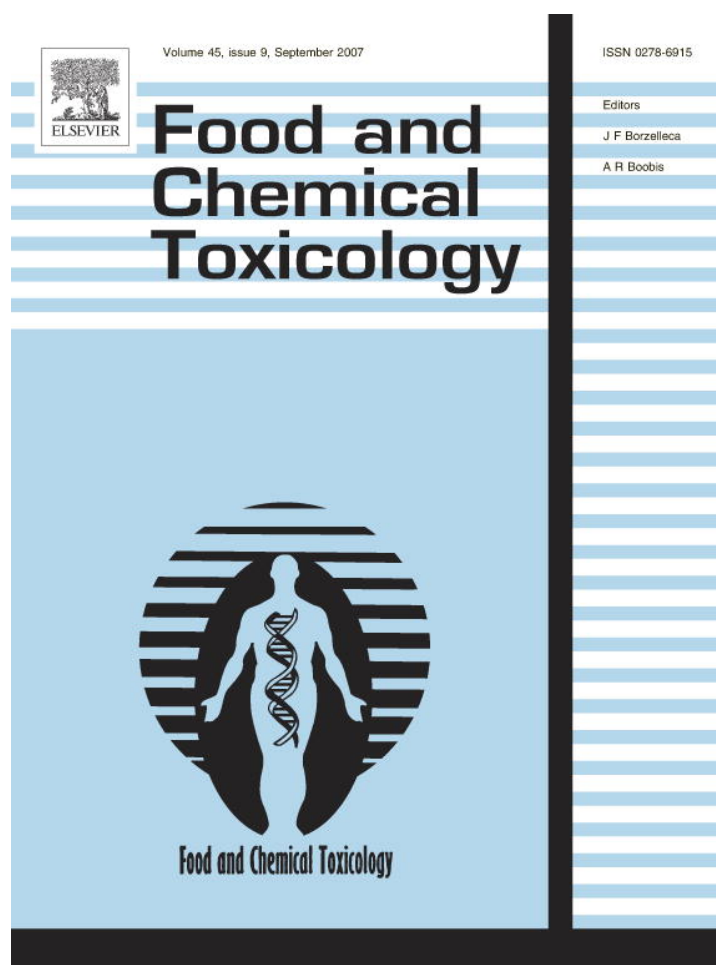


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Content and daily intake of copper, zinc, lead, cadmium, and mercury from dietary supplements in Mexico

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Abstract

This study investigates the presence of Cu, Zn, Cd, Pb, and Hg in 24 dietary supplements purchased in different health stores across the city of Hermosillo, located in the northwest of Mexico. Analysis of metals was done by microwave digestion and atomic absorption spectroscopy. The most abundant elements in dietary supplements were Cu (<0.19–137.85 µg/g) and Zn (<2.83–4785.71 µg/g), followed by Pb (<0.003–66.32 µg/g), Cd (<0.001–2.90 µg/g), and Hg (<0.24–0.85 µg/g). The estimated daily intakes of metals were below those recommended by WHO and the Institute of Medicine, showing that little risk from heavy metals is associated with the consumption of the dietary supplements analyzed. However, some products presented more than 10% of the tolerable daily intake of Pb, indicating that production processes should be improved.

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Keywords: Copper; Zinc; Cadmium; Lead; Mercury; Dietary supplements

1. Introduction

A dietary supplement is defined as a product that intends to supplement the diet, which contains vitamins, minerals, herbs, or other botanicals, amino acids, or any combination of the above ingredients (National Institute of Health, 2003; Dolan et al., 2003; Office of Dietary Supplements, 2005). The human use of these dietary ingredients intends to increase the total daily intake (National Institute of Health, 2003; Dolan et al., 2003), therefore it is essential to ensure the quality of dietary supplements and detect the presence of contaminants.

The industry of dietary supplements has been increased worldwide (National Institute of Health, 2003; Dolan et al., 2003; Office of Dietary Supplements, 2005), because of the growth in their consumption as alternative medi-

cines. This increase is due to consumers' beliefs that these products are natural, safe, and without any adverse effects (Khan et al., 2001; National Institute of Health, 2003; Haider et al., 2004). However, regulatory agencies highlighted the problems with dietary supplements in terms of quality, effectiveness, and safe for human consumption (Sherma, 2003; Haider et al., 2004). According to the Office of Dietary Supplements (2005), in 2004, 55% of United States adults took a dietary supplement and 35% of these users took multivitamins and multiminerals supplements.

Since dietary supplements are regulated as food, and manufactures are not required to register these products before sale (Khan et al., 2001; National Institute of Health, 2003), they must assure that products contain what the label claims, like ingredients in forms that can be absorbed into the body and are free of contaminants (National Institute of Health, 2003).

Metals are widely dispersed in the environment and have a number of applications in the industry (Goyer and Clarkson, 2001). It is now well recognized that some metals like copper (Cu) and zinc (Zn) are essential to human health.

Abbreviations: TW, tablet weight; RP, recommended portion indicate in the label; DSI, dietary supplements intake; TDI, tolerable daily intake.

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The essentiality of Cu and Zn are based on their role as metalloenzymes and as a cofactor of large number of enzymes (FDA, 2001). For these essential metals there is a range of intake over which their supply is adequate to the body (Cu 1.5–3 mg/day, Zn 12–15 mg/day) (Olalla et al., 2004; Singh and Garg, 2006). However, beyond this range, deficiency and toxic effects are observed (FDA, 2001; Olalla et al., 2004; Singh and Garg, 2006). A high supplementation of Cu had been related with liver damage (FDA, 2001). Zn may produce adverse nutrient interactions with Cu. Also, Zn reduces immune function and levels of high density lipoproteins (FDA, 2001). Other metals like lead (Pb), cadmium (Cd), and mercury (Hg) are toxic at lower concentration (Almela et al., 2002; Llobet et al., 2003). Pb is known to induce renal tumors, reduce cognitive development, and increase blood pressure and cardiovascular disease in adults (Goyer and Clarkson, 2001; Haider et al., 2004; Ikem and Egiebor, 2005). Cd may induce kidney dysfunction, osteomalacia and reproductive deficiencies. Hg may cause neurological disorders and has toxic effect on the kidney (Goyer and Clarkson, 2001; Haider et al., 2004; Ikem and Egiebor, 2005).

In Mexico, little information is available about the concentration of metals in dietary supplements. Cueto (1992) indicated high levels of Pb in some remedies used for gastrointestinal infections in Mexicans living in the US and Mexico. These results were very important for the prohibition of Pb-based remedies in Mexico. Because of toxicity of metals, knowledge of intake of metals is essential for establishing regulatory issues. Therefore, it is of great importance for health regulations to determine the status of metal contamination.

In Mexico, as well as the rest of the world, the use of dietary supplements has increased since 1980s. According to the Federal Consumer Agency (Procuraduría Federal del

Consumidor, PROFECO) more than 30% of the population has used supplement. In Mexico, dietary supplements are regulated on its manufacturing process and label information, manufactures also, are requested to ensure the quality of these products (PROFECO, 2005). However, the increase on its commercialization, reports of its inadequate efficacy, and absence of quality programs has determined that health authorities are concern for the national and international distribution of these kinds of products. In addition, Mexican legislation does not regard maximum values of metals in supplements.

Because of the global interest and used of dietary supplements, this study is intended to provide information of intake of metals to those involves in the manufacture, regulatory activities, health authorities, and consumers of these products. The aim of this study was to investigate the presence of metals in dietary supplements and provide information useful to evaluate its possible toxicological risk.

2. Material and methods

2.1. Dietary supplement selection and preparation

Consumers in different health food stores asked questions like *The name of dietary supplement they used and Why?* A list of 100 dietary supplements was obtained, and the first 24 were selected for the study. Products were purchased in 2003 in Hermosillo, Sonora, Mexico. Tables 1a and 1b show the 24 products and their labeled ingredients.

Dietary supplement samples are sold in dried form therefore each product was ground with a mortar and stored at -20°C until analysis.

2.2. Instruments

Digestion of the samples was performed with a microwave system (MARSX, CEM Corp., Matthews, NC) equipped with temperature and pressure control of 200°C , 800 psi and a 1200 W magnetron.

Table 1a
Code and label ingredients of dietary supplements

Code	Principal label ingredients and scientific name ^a
A-SA	Ginkgo biloba, opuntia, nettle, Indian chestnut
B-SA	Ginseng extract (<i>Panax ginseng</i>)
C-SA	Ma Hung root extract (<i>Ephedra sinica</i>), guarana seeds extract (<i>Paullinia cupana</i>), green tea leaf (<i>Camellia sinensis</i>), kola nut (<i>Cola acuminata</i>), hawthorn berries (<i>Crataegus oxyacantha</i>), ginkgo biloba leaf (<i>Ginkgo biloba</i>), beet root (<i>Beta vulgaris</i>), bladderwrack (<i>Fucus vesiculosus</i>), Fo Ti root (<i>Polygonum multilorum</i>), ginger root (<i>Zingiber officinale</i>), royal jelly, saw palmetto berries fruit (<i>Serenoa repens</i>), white willow bark (<i>Salix alba</i>), siberian ginseng root (<i>Eleutherococcus senticosus</i>), yerba mate leaf (<i>Ilex paraguariensis</i>), chromium, vitamin B12
D-SA	Vitamin B1, B3, B6, B12, C, E, potassium citrate, calcium carbonate, calcium pantotenate, PABA, zinc, magnesium citrate, manganese citrate, chromium picolinate, selenium, boric acid, inositol, betaine, pepsin, yeast
E-SA	Ginkgo biloba, nopal
F-SA	Ginkgo biloba extract, calcium carbonate, avicel
G-SA	Ginseng pure powder
H-SA	Guaco stem (<i>Mikania guaco</i>), red vine leaves (<i>Vitis vinifera</i>), horse plant (<i>Equisetum arvense</i>), gorongoro bark (<i>Gorongoro officinalis</i>), gelatin qsp
I-SA	Spirulina algae
J-SA	Beer yeast, glutamic acid, soy protein, ginkgo biloba, vitamin C, dibasic calcium fosfate, royal jelly, soy lectin, toronjil herb, azahar leaf, garlic, zinc
K-SA	Ginseng (<i>Panax ginseng</i>), ginkgo biloba, guarana (<i>Paullinia cupana</i>), glutamic acid
L-SA	Flower pollen

^a Scientific name is indicated only in those products marked in the label.

Table 1b
Code and label ingredients of dietary supplements

Code	Principal label ingredients and scientific name ^a
M-SE	Isolate soy protein, calcium carbonate
N-SE	Maca, pollen, damiana, soy, quinoa, <i>S. palmetto</i> , ginseng, ginkgo biloba, vitamin A, E, C, B1, B6, B12, selenium, iron, sodium, zinc, calcium
O-SE	Vitamin B12, chromium picolinate, kola nut (<i>Cola acuminata</i>), caffeine, guarana seed extract (<i>Paullinia sordilis</i>), green tea leaf extract (<i>Camellia sinensis</i>), garcia cambogia fruit extract, yerba mate (<i>Ilex paraguariensis</i>), roses root extract, corn silk (<i>Stigmata maydis</i>), buchtu leaf (<i>Diosma betulina</i>)
P-SD	Mallow, sen leaves, china cambogia root, lima, tlanchalagua, cascara sagrada, plum extract, seaweed plants, chromium picolinate
Q-SD	Dull brown seaweed plant (<i>Fucus vesiculosus</i>), water plantain leaves (<i>Plantago cordata</i>), china root-root (<i>Smilax china</i>), gelatin qsp
R-SD	Wit grass
S-SD	Apple fruit (<i>Pyrus malus</i>), boldo leaves (<i>Peumus boldus</i>), sen leaves (<i>Cassia acutifolia</i>), uva ursi fruit and leaves (<i>Arctostaphylos pungens</i>), tamarind fruit (<i>Tamarindus indica</i>), isolate soy protein
T-SD	Sen leaves (<i>Cassia angustifolia</i>), toronjil herb (<i>Cedronella Mexicana</i>), boldo (<i>Peumus boldus</i>)
U-SD	L-Carnitine, Chromium picolinate, soy lectin, seaweed plant (<i>Fucus vesiculosus</i>), spirulina algae, fitolaca
V-SA	Garlic (<i>Allium sativum</i>), opuntia (<i>Cactus streptacanta</i>), parsley (<i>Petroselinum sativum</i>), soy lectin, vegetal fiber, lactose, dextrose
W-SE	Chorine complex, inositol, chromium picolinate, cayenne fruit, burdock root, uva ursi leaf, carnitine complex, damiana leaf, betaine, vitamin B6, calcium
X-SA	Calcium proteinate, vitamin B1, B2, B6, B12, nicotinamide, calcium pantothenate

^a Scientific name is indicated only in those products marked in the label.

The quantification of metals was performed with an atomic absorption spectrometer (AAS), Varian model SpectrAA-20 (Varian, Victoria, Australia), equipped with graphite furnace (GTA-96), PSD-56 autosampler, and a hydride vapor generation (VGA-76).

2.3. Reagents

HPLC water was used for the preparation of reagents and standards. All chemicals were trace metal grade. Calibration standard solutions were prepared from commercial standard solutions (High-Purity Standards, Charleston, SC). Concentrate nitric acid (HNO₃) 70% v/v, concentrate hydrochloric acid (HCL) 36% v/v, and concentrate hydrofluoric acid (HF) 48% were obtained from Mallinckrodt (Phillipsburg, NJ, USA). Hydrogen peroxide (H₂O₂) 30% v/v, were obtained from Productos Químicos Monterrey (Mexico). Boric acid (H₃BO₃) 4%, potassium dichromate (K₂Cr₂O₇), and stannous chloride (SnCl₂) were obtained from Sigma (St. Louis, MO, USA).

2.4. Microwave digestion

Homogenized dry samples (0.35 g) were weighed in a microwave digestion lineal vessel and 4 mL of concentrate HNO₃, 4 mL of HF (48% v/v), and 1 mL of concentrate HCl were added. Forty vessels were digested at a time (Table 2). First, they were digested for 15 min at 175 °C and 1 min at 200 °C. Then 12 mL H₃BO₃ (4% v/v) was slowly added to the vessels. The vessels were returned to microwave oven, and a third digestion of 20 min at 165 °C was applied. After the microwave digestion, the samples were cooled, vented, and diluted with HPLC water. This digestion procedure was carried on two times, one for Cu, Zn, Pb, and Cd determination and the other for Hg determination.

Table 2
Microwave digestion program

Parameters	Step		
	I	II	III
Power (%)	100	100	100
Time (min)	15	1	20
Pressure (psi)	200	200	60
Temperature (°C)	175	200	165
Cold (min)	5	5	5

2.5. Atomic absorption spectrometry measurements

For Cu, Zn, Pb, and Cd quantification, the digested samples were diluted at 25 mL. Cu and Zn were determined by atomic absorption spectrometry (Bordajandi et al., 2004) and Cd and Pb by furnace atomic absorption spectrometry (Khan et al., 2001). The oven program employed is described in Table 3. For Hg quantification, 0.5 mL of K₂Cr₂O₇ (1% w/v) and 4 mL of HNO₃ (50% v/v) were added to digested samples and afterwards were diluted at 50 mL with HPLC water. Hg was determined by cold vapor technique after reduction with SnCl₂ (20%, w/v) in HCl (20%, v/v) (Khan et al., 2001).

2.6. Daily intake estimates

The daily intakes of Cu, Zn, Pb, Cd, and Hg from each supplement were calculated by multiplying the daily intake indicated on the product label by the mean unit mass and by the element concentration determined in the study. The metal intakes were compared with the tolerable daily intakes for metals recommended by the WHO (1989) and by the Institute of Medicine (2001).

2.7. Quality control

All glassware was treated with Pierce solution 20% (v/v) (Pierce, Rockford, IL), rinsed with cold tap water followed by 20% (v/v) nitric acid and then rinsed with double-distilled water. For quality control purposes,

Table 3
Instrumental conditions for Pb and Cd determination in dietary supplements by ETAAS

Conditions	Lead	Cadmium
Wavelength (nm)	283.3	228.8
Slit width (nm)	0.5	0.5
Graphite tube	Partition	Partition
Dry temperature (°C)	95	95
Ashing temperature (°C)	400	300
Atomization temperature (°C)	2100	1800
Sample and standard volume (μL)	20	10
Matrix modifier	–	–
Background correction	Deuterium	Deuterium
Signal mode	Peak height	Peak height

blanks and duplicates samples were analyzed during the procedure. Standard Reference Material, SBR-1515, Apple Leaves from the National Institute of Standards and Technology (NIST, Gaithersburg, MD) was analyzed for quality assurance. Percent recovery means were: Cu (103%), Zn (101%), Pb (89%), Cd (105%), Hg (97%). The variation coefficient was below 5%. Detection limits ($\mu\text{g/g}$) were determined by the blanks method: Cu (0.19), Zn (2.83), Pb (0.003), Cd (0.001), Hg (0.24).

3. Results and discussion

Metal concentrations found in dietary supplements are shown in Table 4. Cu, Zn, and Pb showed wide range. This could have been a consequence of the supplements' components that may be the source of metals, the geographical source of the supplement, and the number of ingredients in the supplement. Only in three supplements (H-SA, O-SE, W-SE) were detected all five study metals. These products are recommended for losing fat and increasing energy. For the rest of dietary supplements were detected three to four metals at time.

3.1. Copper and zinc

The most abundant elements in dietary supplements were Cu and Zn. The highest concentration of Cu was detected in D-SA (137.85 $\mu\text{g/g}$) and the highest levels of Zn were found in J-SA (4785.71 $\mu\text{g/g}$) and D-SA (2250 $\mu\text{g/g}$). The J-SA ingredients are ginkgo biloba, garlic, azahar leaves, beer yeast, zinc, and vitamins B6 and B12. The D-SA is made of vitamins C, B, E, and also of minerals like Ca, potassium (K), Cu and Zn. Moreover, Cu and Zn are widely available in food and can come from both plant

and animal sources (Goyer and Clarkson, 2001; FDA, 2001).

Compared to the values described in the literature for Cu and Zn, the levels found in this study were lower than those reported by Hight et al. (1993) in other dietary supplements, but higher than those observed in medicinal plants (Haider et al., 2004). This difference may due by the number and kind of ingredients in dietary supplements. Naithani and Kakkar (2005) reported lower levels of metals in herbal teas with fewer ingredients than in polyherbal teas.

3.2. Lead and cadmium

Of the nonessential metals, Pb showed the highest variation (0.03–66.32 $\mu\text{g/g}$). The highest concentration of Pb was detected in X-SA supplement. This product is composed of calcium proteinate and pantothenate, vitamins B1, B2, B6, B12, nicotinamide, and it is recommended to take 1 tablet three times a day as nutritional supplement. Cd ranged from 0.18 to 2.90 $\mu\text{g/g}$, the highest concentration (2.90 $\mu\text{g/g}$) was found in N-SE product used as energizer. Pb and Cd are considered environment contaminants (Goyer and Clarkson, 2001; Sherma, 2003; Haider et al., 2004). In the present study many dietary supplements are derived from natural products and it is possible that metals detected in these products have a natural origin. In order to reduce health risk it is important that manufactures be concern about this situation as well as the equipments used to dry, ground, and packet dietary supplements.

Table 4
Metal concentration in dietary supplements ($\mu\text{g/g}$, dw)

Code	Origin	Cu	Zn	Pb	Cd	Hg
A-SA	Mexico	5.71	9.28	0.51	0.35	<0.24
B-SA	Korea	8.57	10.00	0.11	0.57	<0.24
C-SA	USA	9.29	13.93	0.31	0.59	<0.24
D-SA	USA	137.85	2250.00	0.59	0.78	<0.24
E-SA	Mexico	3.57	27.85	0.03	0.18	<0.24
F-SA	Mexico	1.79	<2.83	0.21	0.53	<0.24
G-SA	Mexico	6.43	3.57	<0.001	0.37	0.81
H-SA	Mexico	7.15	24.64	1.26	1.53	0.85
I-SA	Mexico	22.14	27.86	1.38	1.63	<0.24
J-SA	Mexico	2.14	4785.71	1.81	2.10	<0.24
K-SA	Mexico	1.43	<2.83	0.10	0.19	<0.24
L-SA	Mexico	6.43	17.86	15.92	1.12	<0.24
M-SE	Mexico	11.43	30.00	0.25	0.62	<0.24
N-SE	Mexico	8.57	31.43	3.04	2.90	<0.24
O-SE	USA	6.43	37.15	0.33	0.61	0.80
P-SD	Mexico	5.00	17.86	0.65	0.54	<0.24
Q-SD	Mexico	4.29	17.14	2.49	2.80	<0.24
R-SD	Mexico	10.00	35.00	1.32	1.07	<0.24
S-SD	Mexico	2.14	10.71	1.99	2.23	<0.24
T-SD	Mexico	6.43	24.64	2.26	2.12	<0.24
U-SD	Mexico	1.07	9.28	6.16	2.20	<0.24
V-SA	Mexico	9.29	26.43	0.33	0.42	<0.24
W-SE	USA	3.57	27.50	8.30	2.03	0.80
X-SA	Mexico	<0.19	15.00	66.32	1.36	0.29
Range		<0.19–137.85	<2.83–4785.71	<0.003–66.32	<0.001–2.90	<0.24–0.85

Compared to the values described in the literature for Pb, the levels found in this study were similar to those reported by Hight et al. (1993), Au et al. (2000) and Dolan et al. (2003) in other dietary supplements, but higher than those observed in ginseng products (Khan et al., 2001) and medicinal plants (Haider et al., 2004). For Cd, the levels found in the present study were similar than those reported in other dietary supplements (Hight et al., 1993) and in medicinal plants (Haider et al., 2004), but higher than those reported by Khan et al. (2001) and Dolan et al. (2003). As indicated previously these differences may due by the kind and number of ingredients presented in the study dietary supplements. According to WHO (1989), the tolerable limit for Pb in medicinal herbs is 10 µg/g and 0.3 µg/g for Cd. In the present study 4 dietary supplements were higher than these Pb limit, but lower than those reported (7.70–94.10%) previously in Pb-based remedies used for gastrointestinal infections in Mexico (Cueto, 1992). For Cd almost all samples were higher than Cd permissible limits.

3.3. Mercury

The Hg concentration ranged from 0.29 to 0.85 µg/g. The highest value was found in H-SA product which is recommended one capsule before meals as nutritional supplement. This product is elaborated with guaco stem (*Mikania guaco*), red vine leaves, horse plant, and gorongoro bark

(*Gorongoro officinalis*). Mercury is a common contaminant that is occasionally included as an ingredient in the form of cinnabar (HgS) in Chinese medicinal products as an active ingredient to treat diseases such epilepsy, sore throat, and asthma (Bin et al., 2001; Hong et al., 2002). In the present study 19 supplements were manufactured in Mexico, four in Unites States, and only one in Korea (in which there was no detectable mercury).

Compared to the values described in the literature for Hg, the levels found in this study were mostly lower or similar than those reported in other dietary supplements (Au et al., 2000; Hong et al., 2002; Dolan et al., 2003), but higher than those values (0.0026–0.010 µg/g) reported by Bin et al. (2001) in Chinese medicinal herbs.

3.4. Metal daily intakes

Daily intakes of metals for each dietary supplement are shown in Table 5. The highest, Cu intakes, corresponded to D-SA (68.93 µg/day) represented the 0.69% of the TDI level of 10000 µg/day established by the Institute of Medicine (for 19–>70 years male and female). There is negligible risk for toxicity. For Zn the intake ranged from 3.21 to 7178.57 µg/day, corresponded the highest intake to J-SA supplement. This intake represented the 17.9% of the TDI of 40000 µg/day recommended by Institute of Medicine (for 19–>70 years male and female). Therefore,

Table 5
Daily intake of metals by dietary supplements (µg/day)

Code	TW (g)	RP (day)	DSI (g/day)	Cu	Zn	Pb	Cd	Hg
A-SA	0.5	6.0	3.0	17.13	27.84	1.53	1.05	<0.24
B-SA	0.3	3.0	0.9	7.71	9.00	0.10	0.51	<0.24
C-SA	0.9	2.0	1.8	16.72	25.07	0.56	1.06	<0.24
D-SA	0.5	1.0	0.5	68.93	1125.00	0.30	0.39	<0.24
E-SA	0.4	6.0	2.4	8.57	66.84	0.07	0.43	<0.24
F-SA	0.6	2.0	1.2	2.15	<2.83	0.25	0.64	<0.24
G-SA	0.5	4.0	2.0	12.86	7.14	<0.001	0.74	1.63
H-SA	0.5	3.0	1.5	10.73	36.96	1.89	2.30	1.15
I-SA	1.0	3.0	3.0	66.42	83.58	4.14	4.89	<0.24
J-SA	0.5	3.0	1.5	3.21	7178.57	2.72	3.15	<0.24
K-SA	0.5	6.0	3.0	4.29	<2.83	0.30	0.57	<0.24
L-SA	1.5	3.0	4.5	28.94	80.37	71.64	5.04	<0.24
M-SE	1.0	6.0	6.0	68.58	180.00	1.50	3.72	<0.24
N-SE	0.5	2.0	1.0	8.57	31.43	3.04	2.90	<0.24
O-SE	0.6	2.0	1.2	7.72	44.58	0.40	0.73	0.93
P-SD	0.4	6.0	2.4	12.00	42.86	1.56	1.30	<0.24
Q-SD	0.5	3.0	1.5	6.44	25.71	3.74	4.20	<0.24
R-SD	0.1	6.0	0.6	6.00	21.00	0.79	0.64	<0.24
S-SD	0.1	3.0	0.3	0.64	3.21	0.60	0.67	<0.24
T-SD	0.4	6.0	2.4	15.43	59.14	5.42	5.09	<0.24
U-SD	0.6	3.0	1.8	1.93	16.70	11.09	3.96	<0.24
V-SA	0.9	6.0	5.4	50.17	142.72	1.78	2.27	<0.24
W-SE	0.7	4.0	2.8	10.00	77.00	23.24	5.68	2.25
X-SA	1.0	3.0	3.0	<0.19	45.00	198.96	4.08	0.86
Range	–	–	–	<0.19–68.93	<2.83–71 178.57	<0.003–198.96	0.39–5.68	<0.24–2.25
TDI				10000	40000	240	68	49

TW: Tablet weight; RP: recommended portion indicate in the label; DSI: dietary supplements intake; TDI: tolerable daily intake. TDI for Cu and Zn; recommended by the Institute of Medicine in µg/day for 19–>70 years male and female. TDI for Pb, Cd, and Hg; recommended by the WHO in µg/day for a body weight of 68 kg.

the risk of adverse effects resulting from excess zinc intake from these supplements appears to be low. Zn is a cofactor in the superoxide dismutase enzyme, which is involved in protection against oxidative processes (Olalla et al., 2004). This characteristic may explain the addition of Zn in dietary supplements. It is recognized that high intakes of zinc are due to the use of supplements. Therefore, information about potential toxicity of zinc as a dietary supplement needs study.

For Pb the daily intake ranged from 0.07 to 198.96 µg/day. The highest value was corresponded to X-SA supplement and represented the 82.9% of the TDI of 240 µg/day of the WHO (for a body weight of 68 kg). For Cd the daily intake ranged from 0.39 to 5.68 µg/day. The highest value was corresponded to W-SE supplement, and represented the 7.5% of the TDI of 68 µg/day of the WHO (for a body weight of 68 kg). For Hg the daily intake ranged from 0.86 to 2.25 µg/day. The highest value was corresponded to W-SD supplement, and represented the 4.7% of the TDI of 49 µg/day of the WHO (for a body weight of 68 kg). It is important to note that the presence of 30% and 83% of the TDI of Pb in both X-SA and L-SA supplements indicated that production processes need to be improved.

There are few reports related to metal intakes in dietary supplements. The levels of Cu and Zn found in this study were lower than those reported by Hight et al. (1993) where Cu and Zn daily intakes ranged from 4 to 3000 µg/day and 0.5 to 66000 µg/day respectively, while Pb and Cd ranges were higher (Pb 0.1–104 µg/day, Cd 0.021–1.16 µg/day). Cu and Zn are routinely added to various dietary supplements as health and nutritional substances for adults (FDA, 2001).

Since total daily intake is defined as the sum of the maximum daily intake from total diet, it is noteworthy to mention that the daily intake calculated values of each metal represented the concentration only by the intake of one supplement. The food is the principal way of human exposure to metals, accounting for >90% if compared to other exposure ways like inhalation and dermal contact (Goyer and Clarkson, 2001; Bin et al., 2001). According to the FDA, consumers of dietary supplements generally used more than one product. In the present study was estimated the intake of metals by three products, Cu and Zn presented little increased concentration. However, Pb, Cd, and Hg concentrations increase 1.5–3 times. Therefore, the ingestion of metals such a Pb and Cd may increase significantly due the supplement consumptions. Also, the intake of combined supplements plus fortified foods may cause health problems.

The levels of Cu, Zn, Cd, Pb, and Hg and their daily intake were determined in 24 dietary supplements. The estimated daily intakes of metals were below those recommended by WHO and the Institute of Medicine, showing that little intake of metals is associated with the consumption of the dietary supplements analyzed. However is important to note that daily intake of metals like Pb may increase due to the number of consumed dietary supple-

ments. Therefore, the study of toxicological implications of metals in dietary supplements is needed. Also, it is important to establish regulatory limits regarding the presence of metals as contaminants in dietary supplements in order to safeguard human health.

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