Vegetables Collected in the Cultivated Andean Area of Northern Chile: Total and Inorganic Arsenic Contents in Raw Vegetables

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High levels of arsenic are found in the soil and water of the Second Region in Chile as a result of natural causes. Total and inorganic arsenic contents were analyzed in the edible part of 16 agricultural products (roots, stems, leaves, inflorescences, and fruits) grown in this area. The total arsenic contents varied in the range 0.008–0.604 μg g^{-1} of wet weight (ww), below the maximum level allowed by Chilean legislation (1 μg g^{-1} of ww). Inorganic arsenic contents (range = 0.008–0.613 μg g^{-1} of ww) represented between 28 and 114% of total arsenic. The concentrations of total and inorganic arsenic found in edible roots and leaves were higher than those found in fruit. The highest concentrations were found in a sample of spinach. High quantities of this vegetable would have to consumed (250 g/day) to reach the Provisional Tolerable Weekly Intake for inorganic arsenic. The vegetable group may make a considerable contribution to the total intake of inorganic arsenic.

KEYWORDS: Total arsenic; inorganic arsenic; vegetables; Chile

INTRODUCTION

Because of geological factors, the Second Region in Chile, located in the pre-Andean area in the north of the country, presents an environment with high concentrations of arsenic. The volcanic bedrock in this area has a high content of arsenic associated with pyrite minerals (*1*), and subsequent processes of disintegration and lixiviation distribute the arsenic in the atmosphere, soil, and water. In this region, concentrations of arsenic reaching 1099 mg kg^{-1} have been detected in the soil (*2*), and levels attaining 11.25 mg L^{-1} have been found in aquifers (*3*).

This contamination of natural origin has had a negative effect on the health of the inhabitants of the Second Region in Chile. In this respect, the data provided by various Chilean organizations and reported by Smith et al. (*4*) showed that during the period 1955–1970 a large part of the population of the Second Region consumed drinking water with arsenic concentrations >0.05 mg L^{-1}, the maximum value permitted by Chilean legislation. They found that bladder and lung cancer mortality was markedly elevated and estimated that arsenic might account for 7% of all deaths among those aged 30 years and over. The towns in the Second Region with the highest populations now receive water with an arsenic content that conforms to Chilean regulations, but the financial cost makes it difficult to apply decontamination processes in smaller population centers.

Besides water, food is another source of arsenic for humans. Many total diet studies have shown that fish and shellfish make the largest contribution of arsenic to the diet (*5*, *6*). However, food groups such as vegetables and cereals can become a path by which arsenic may enter the food chain, because they can reflect the levels of arsenic that exist in the environment in which they are cultivated (soil, irrigation water, and atmosphere). Greenhouse experiments have revealed that an increase in arsenic in cultivated soils leads to an increase in levels of arsenic in edible vegetables (*7–9*), with many complex factors affecting bioavailability, uptake, and phytotoxicity of arsenic (*8*). Several scattered villages situated in the Second Region in Chile are dedicated to agricultural activities and cultivate 2193 ha, with agriculture accounting for ~25–76% of income, and 81–99% of its production is for self-consumption (*10*).

In the literature there are data for total arsenic contents in cereals and vegetables (*10–12*), but few values have been reported for inorganic arsenic (*9*, *13–15*). However, knowledge of inorganic arsenic contents is essential for an evaluation of

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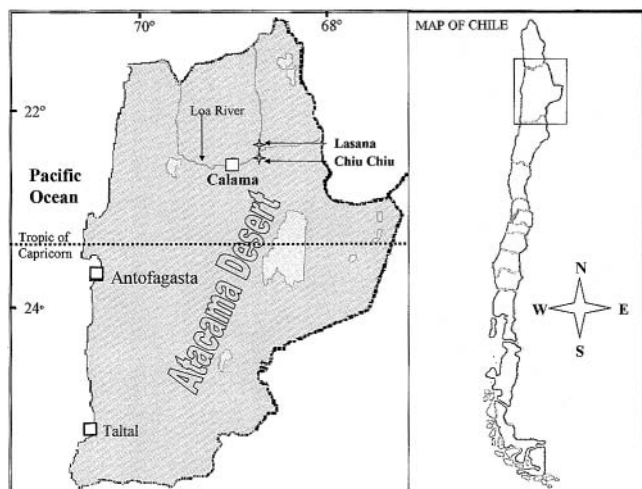
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Figure 1. Map of the Second Region in Chile, showing the locations where samples were collected, Lasana and Chiu Chiu, and the major towns in the region.

health risks; inorganic arsenic includes the most toxic species of arsenic [As(III) + As(V)], and the toxicological reference value for arsenic is established in terms of intake of inorganic arsenic [Provisional Tolerable Weekly Intake (PTWI) = 15 μg of inorganic arsenic/kg of body weight/week (*16*)].

Agriculture is an important resource in the Second Region, both commercially and in terms of domestic consumption, and the omnipresence of arsenic in the environment makes it of paramount importance to discover the levels of inorganic arsenic in vegetables produced in the area. The aim of the present study is to determine total and inorganic arsenic contents in agricultural products grown in the Second Region in soils naturally contaminated with arsenic and also to examine the possible commercial and health implications associated with distribution and consumption of those products.

MATERIALS AND METHODS

Study Area. The vegetables were collected in the agricultural smallholdings corresponding to the communities of Chiu Chiu and Lasana, located in the interior of the Second Region in Chile, in the geographical area known as the Desert of Atacama (**Figure 1**).

Chiu Chiu is a village of 1061 inhabitants, situated in the Loa area. Agriculture is the inhabitants' main economic activity. Their diet consists principally of vegetables and animal meat. The soil and water in the Chiu Chiu valley show a high accumulation of arsenic (*2*). In preliminary studies, agricultural surface soil (0−20 cm) collected in Chiu Chiu and analyzed in a Spanish laboratory (IATA-CSIC) showed total arsenic contents ranging between 50 and 70 mg kg^{-1} (unpublished data). Further preliminary studies carried out in a Chilean laboratory analyzed water obtained from the River Loa, which is used to irrigate the smallholdings of cultivated land, and detected concentrations of total arsenic in the range of 0.07−0.28 mg L^{-1} (unpublished data).

In Lasana, a village close to Chiu Chiu, there is a population of 135 people, and agriculture is their main work activity. The characteristics of the soil and water in Lasana are similar to those of Chiu Chiu.

Samples. The vegetables were selected on the basis of two criteria, production and consumption within the community, and were collected in the field with a random collection procedure (*17*). Sixteen agricultural products were analyzed: garlic, onion, potato, carrot, beetroot, asparagus, lettuce, chard, spinach, cabbage, broccoli, cauliflower, corn, tomato, beans, and pumpkin. For each species of vegetable, samples were collected from various agricultural smallholdings until the quantity required for subsequent analysis was obtained. They were collected by hand and afterward carefully packed into polyethylene bags. The cleaning of the samples was performed first by shaking them to remove the earth. Afterward, the samples were precleaned with deionized water.

They were then stored in a cold box at 4 °C and transported to the laboratory. All of the samples were freeze-dried. They were then ground in a domestic apparatus, and the powder was vacuum-packed and kept at 4 °C until analysis.

Instrumentation. Determination of inorganic and total arsenic was performed with a Perkin-Elmer (PE) 3300 atomic absorption spectrometer (Perkin-Elmer Hispania, S.A., Madrid, Spain) equipped with a flow injection system (PE FIAS-400) coupled with an autosampler (PE AS-90) to provide hydride generation in a flow injection mode. An electrothermally heated quartz cell was employed.

Other equipment used included a Genesis SQ 25 Super ES lyophilizer (Virtis, Gardiner, NY), a PL 5125 sand bath (Raypa, Scharlau, Barcelona, Spain), a K 1253 muffle furnace equipped with a Eurotherm Controls 902 control program (Heraeus, Madrid, Spain), a KS 125 Basic mechanical shaker (IKA Labortechnik, Merck Farma y Química, Barcelona, Spain), and an Eppendorf 5810 centrifuge (Merck Farma y Química).

Reagents. Deionized water, 18 MΩ cm, was used for the preparation of reagents and standards. All chemicals were of pro analysi quality or better. A standard solution of As(V) (1000 mg L^{-1}) was prepared by dilution of a Titrisol standard (Merck, Darmstadt, Germany). Standard solutions of As(III) were prepared from a reduced standard solution of As(V), using 5 mL of HCl (6 mol L^{-1}) and 5 mL of a reducing solution [5% (m/v) KI and 5% (m/v) ascorbic acid] and allowed to stand at room temperature for 30 min. All glassware was treated with 10% v/v HNO$_3$ for 24 h and then rinsed three times with Milli-Q water before being used.

Two reference samples certified for total arsenic contents [National Institute of Standards and Technology (NIST; Gaithersburg, MD)] were employed for QA/QC of analytical data: tomato leaves (SRM 1573) and rice flour (SRM 1568a).

Total Arsenic Determination. The lyophilized sample (0.25 ± 0.01 g) was weighed, and 1 mL of ashing aid suspension (20% m/v MgNO$_3$ + 2% m/v MgO) and 5 mL of 50% v/v HNO$_3$ were added. The mixture was evaporated on a sand bath until total dryness and placed in a muffle furnace at an initial temperature not exceeding 150 °C. The temperature was increased progressively to 425 ± 25 °C at the rate of 50 °C/h, and the maximum temperature was maintained for 12 h. Once the ash has cooled, 5 mL of 10% HNO$_3$ was added, the mixture was evaporated in the sand bath, and the calcination process was repeated until white ash was obtained. The white ash was dissolved in 5 mL of 50% v/v HCl and 5 mL of reducing solution (5% m/v KI + 5% m/v ascorbic acid). After 30 min, the resulting solution was diluted to volume with 50% v/v HCl and filtered through Whatman No. 1 filter paper into a 25-mL volumetric flask. The instrumental conditions used for arsenic determination by flow injection hydride generation atomic absorption spectrometer (FI-HG-AAS) were the following: loop sample, 0.5 mL; reducing agent, 0.2% (m/v) NaBH$_4$ in 0.05% (m/v) NaOH, 5 mL min^{-1} flow rate; HCl solution 10% (v/v), 10 mL min^{-1} flow rate; carrier gas, argon, 100 mL min^{-1} flow rate; wavelength, 193.7 nm; spectral bandpass, 0.7 nm; electrodeless discharge lamp system 2, lamp current setting, 400 mA; cell temperature, 900 °C.

The analytical characteristics for the total arsenic methodology are as follows: detection limit, 0.006 μg g^{-1} of ww; precision, 2%; accuracy for tomato leaves, found value = 0.28 ± 0.02 mg kg^{-1} (certified value = 0.27 ± 0.05 mg kg^{-1}); accuracy for rice flour, found value = 0.29 ± 0.03 mg kg^{-1} (certified value = 0.29 ± 0.03 mg kg^{-1}).

Inorganic Arsenic Determination. The method used has been described in a previous paper (*18*). The lyophilized sample (0.50 ± 0.01 g) was weighed into a 50 mL screw-top centrifuge tube, 4.1 mL of water was added, and the sample was agitated until it was completely moistened. Then 18.4 mL of concentrated HCl was added, and the sample was agitated again for 1 h and then left to stand for 12−15 h (overnight). The reducing agent (1 mL of 1.5% m/v hydrazine sulfate solution and 2 mL of HBr) was added, and the sample was agitated for 30 s. Then 10 mL of CHCl$_3$ was added, and the sample was agitated for 10 min. The phases were separated by centrifugation at 2000 rpm for 5 min. The chloroform phase was separated by aspiration and poured into another tube. The extraction process was repeated two more times. The chloroform phases were combined and centrifuged again. The remnants of the acid phase were completely eliminated by aspiration

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Table 1. Levels of Total Arsenic and Inorganic Arsenic, Percentages of Inorganic Arsenic with Respect to Total Arsenic, and Humidity in Roots of Vegetables Grown in the Second Region in Chile[a]

roots		μg of As g^{-1} of wet wt		% inorganic As	humidity (%)
		total	inorganic		
garlic	a	0.368 ± 0.005	0.378 ± 0.008	100	61
	b	0.317 ± 0.009	0.267 ± 0.010	84	59
	c	0.030 ± 0.002	0.030 ± 0.001	100	69
onion		0.067 ± 0.001	0.075 ± 0.006	114	88
potato	a	0.133 ± 0.004	0.098 ± 0.002	77	76
	b	0.102 ± 0.002	0.090 ± 0.003	90	80
	c	0.021 ± 0.001	0.024 ± 0.002	100	77
carrot	a	0.138 ± 0.002	0.126 ± 0.001	93	87
	b	0.078 ± 0.003	0.060 ± 0.002	75	89
	c	0.138 ± 0.002	0.128 ± 0.003	93	88
beetroot	a	0.268 ± 0.030	0.090 ± 0.004	33	85
	b	0.129 ± 0.002	0.121 ± 0.005	92	86
	c	0.168 ± 0.001	0.160 ± 0.027	94	83

[a] Each value corresponds to a sample collection period (a = April 1998; b = April 1999; c = December 1999). Each value is the mean ± SD of three replicates.

(acid phase remnants in the chloroform phase cause substantial overestimates of inorganic arsenic). Possible remnants of solid material in the chloroform phase were eliminated by passing it through Whatman GD/X syringe filters with a 25 mm PTFE membrane (Merck Farma y Química S.A., Barcelona, Spain).

The inorganic As in the chloroform phase was back-extracted by agitation for 10 min with 10 mL of 1 mol L^{-1} HCl. The phases were separated by centrifugation at 2000 rpm, and the acid phase was then aspirated and poured into a beaker. This stage was repeated once again, and the back-extraction phases obtained were combined. The determination of inorganic arsenic in the back-extraction phase was performed by means of the following procedure: 2.5 mL of ashing aid suspension and 10 mL of concentrated HNO$_3$ were added to the combined back-extraction phases. The resulting solution was evaporated to dryness and then treated in the same way as for total arsenic (dry ashing mineralization and quantification by FI-HG-AAS).

For the inorganic arsenic methodology the detection limit is 0.003 μg g^{-1} of ww and the precision 4%. There are no reference materials with certified contents of inorganic arsenic, and therefore the accuracy of the method was evaluated by using the sample of rice flour (SRM 1568a). The inorganic arsenic concentration found (0.110 ± 0.006 μg g^{-1}) is close to the only existing previous reference (0.092 μg g^{-1}) (*15*), obtained by the U.S. Food and Drug Administration using trifluoroacetic acid extraction, ion chromatography, and inductively coupled plasma mass spectrometry quantification.

RESULTS AND DISCUSSION

The samples were collected during three different periods: April 1998, April 1999, and December 1999. For some of the samples it was not possible to collect specimens during the three periods. The concentrations of total and inorganic arsenic found in the edible portions of the vegetables are shown in **Tables 1−4**. Each of the values shown in the tables is the mean of three replicates determined in samples obtained in one collecting period. To facilitate discussion of the results, the samples have been classified into five groups on the basis of the edible parts analyzed (root, stem, leaf, inflorescence, and fruit).

Total Arsenic Contents. The total arsenic contents found in the samples of garlic, onion, potato, carrot, and beetroot (**Table 1**), all tubers, were in the range of 0.021−0.368 μg g^{-1} of ww. The highest levels were found in garlic, with a mean value for the three sample collection periods of 0.238 ± 0.182 μg g^{-1} of ww (mean ± SD), and the lowest levels were found in samples of potato (0.085 ± 0.058 μg g^{-1} of ww).

Table 2. Levels of Total Arsenic and Inorganic Arsenic in the Skin and Edible Part of Roots of Vegetables Grown in the Second Region in Chile

root[a]	total As			inorganic As		
	skin (μg g^{-1} of ww)	root (μg g^{-1} of ww)	skin/ root	skin (μg g^{-1} of ww)	root (μg g^{-1} of ww)	skin/ root
garlic	2.1	0.030	70	1.93	0.030	64
potato	0.058	0.021	3	0.050	0.024	2
carrot	0.364	0.138	3	0.314	0.128	2
beetroot	0.877	0.168	5	0.749	0.160	5

[a] Samples were collected in December 1999.

Table 3. Levels of Total Arsenic and Inorganic Arsenic, Percentages of Inorganic Arsenic with Respect to Total Arsenic, and Humidity in Stems and Leaves of Vegetables Grown in the Second Region in Chile[a]

sample		μg of As g^{-1} of wet wt		% inorganic As	humidity (%)
		total	inorganic		
stems					
asparagus	a	0.132 ± 0.002	0.124 ± 0.001	92	88
	b	0.130 ± 0.007	0.109 ± 0.005	85	85
	c	0.081 ± 0.002	0.064 ± 0.009	88	91
leaves					
lettuce	a	0.234 ± 0.008	0.218 ± 0.006	96	89
	b	0.068 ± 0.001	0.061 ± 0.002	86	94
	c	0.448 ± 0.001	0.394 ± 0.099	87	91
chard	a	0.195 ± 0.031	0.161 ± 0.012	84	88
	b	0.247 ± 0.017	0.190 ± 0.003	76	92
	c	0.266 ± 0.001	0.187 ± 0.016	70	90
spinach	a	0.604 ± 0.010	0.613 ± 0.020	102	84
	c	0.121 ± 0.004	0.087 ± 0.001	75	91
cabbage	c	0.016 ± 0.002	0.013 ± 0.001	50	93

[a] Each value corresponds to a sample collection period (a = April 1998; b = April 1999; c = December 1999). Each value is the mean ± SD of three replicates.

Table 4. Levels of Total Arsenic and Inorganic Arsenic, Percentages of Inorganic Arsenic with Respect to Total Arsenic, and Humidity in Inflorescences and Fruits of Vegetables Grown in the Second Region in Chile[a]

sample		μg of As g^{-1} of wet wt		% inorganic As	humidity (%)
		total	inorganic		
inflorescence					
broccoli	a	0.318 ± 0.004	0.086 ± 0.001	28	83
cauliflower	a	0.060 ± 0.002	0.054 ± 0.004	83	87
	c	0.014 ± 0.001	0.010 ± 0.003	100	91
fruit					
corn	a	0.404 ± 0.006	0.242 ± 0.001	60	10
	b	0.015 ± 0.003	0.010 ± 0.003	50	68
	c	0.152 ± 0.009	0.110 ± 0.023	73	6
tomato	a	0.037 ± 0.001	0.028 ± 0.002	75	93
	b	0.008 ± 0.002	0.008 ± 0.002	100	92
	c	0.011 ± 0.001	0.009 ± 0.001	100	93
beans	c	0.022 ± 0.003	0.023 ± 0.013	100	86
pumpkin	c	nd	nd		92

[a] Each value corresponds to a sample collection period (a = April 1998; b = April 1999; c = December 1999). Each value is the mean ± SD of three replicates. nd, not detectable.

In the samples of garlic, potato, carrot, and beetroot, the arsenic present in the skin was quantified (Table 2). Comparison with the contents found in the edible part shows concentrations of arsenic in the skin between 3 and 70 times greater than the levels in the edible part. The concentration was particularly high in the skin of the garlic samples (2.1 μg g^{-1} of ww). Studies

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performed by others showed the same behavior in turnip (*8*) and carrots (*9*), with the total arsenic in the skin being ~2–7 times greater than in the inner part of the root. From the viewpoint of the consumer, given the low weight of the skin with respect to the total weight of the edible portion, consumption of the product with or without skin would not seem to have a substantial effect on the intake of total arsenic.

Table 3 shows the total arsenic contents in vegetables for which the edible portion consists of the stem and leaves. Asparagus, the only stem analyzed, presented a mean value for the three sample collection periods of 0.114 ± 0.029 μg g^{-1} of ww (mean $\pm$ SD). In leaves, the contents of total arsenic lay within the range 0.016–0.604 μg g^{-1} of ww, with the highest level being found in a sample of spinach and the lowest level in a sample of cabbage. Lettuce and chard presented similar mean values: lettuce, 0.250 ± 0.191 μg g^{-1} of ww; chard, 0.236 ± 0.037 μg g^{-1} of ww.

The contents of total arsenic in inflorescences and fruits are shown in **Table 4**, and in general they are the lowest levels found in the whole study. An exception is the sample of broccoli, with a total arsenic content of 0.318 μg g^{-1} of ww, close to the level found in some roots and stems. With respect to the samples of corn, the disparity in the values found in each of the subsamples is largely attributable to differences in humidity. The two subsamples with the highest arsenic levels were dry samples in which there might have been a concentration of analyte resulting from loss of humidity. In fact, if we take the same humidity value for all of the samples (68%, humidity of fresh sample), the total arsenic contents would be of the same order for all samples of corn.

A comparison of the results obtained for all of the vegetables (**Tables 1**, **3**, and **4**) shows that, with the exception of spinach, roots presented the highest levels of arsenic, whereas stems and leaves presented middling values, and the lowest levels corresponded to fruits. This has already been reported by Carbonell-Barrachina et al. (*19*), who established that arsenic tends to accumulate in the vegetable root and decreases from the root to the fruit, which always shows the lowest total arsenic contents.

The data in the literature for total arsenic contents in food products of vegetable origin analyzed in total diet and/or market basket survey methods show that the vegetables and cereals customarily consumed present contents in the range of <0.004–0.303 μg g^{-1} of As, ww (*6, 11, 13, 20*), which is lower than the range of values found in the present study for vegetables in the Second Region in Chile (0.008–0.604 μg g^{-1} of ww). This might indicate a higher base level of arsenic in vegetable products in the Second Region of Chile than in areas without contamination of natural origin.

Inorganic Arsenic Contents. The concentrations of inorganic arsenic found in the vegetables analyzed ranged from 0.008 μg g^{-1} of ww for a sample of tomato to 0.613 μg g^{-1} of ww for a sample of spinach (**Tables 1**, **3**, and **4**).

There are very few data in the literature for inorganic arsenic contents in vegetables. Schoof et al. (*13*), using the market basket survey method in a study of vegetables collected in the United States, detected concentrations of inorganic arsenic in the range of 0.002–0.074 μg g^{-1} of ww, values far below the levels found in the present study. Recently, in long-grain white rice samples from the United States, inorganic arsenic contents varying between 0.091 and 0.095 μg g^{-1} have been detected (*15*).

In most of the samples studied the inorganic arsenic contents were very close to the total arsenic contents, which may indicate

very low methylation of inorganic arsenic absorbed from the soil. Another possibility is low absorption of methylated compounds that may exist in the soil or the absence of such compounds in the soils used for growing the samples analyzed. The percentages of inorganic arsenic with respect to total arsenic varied from 28% for a sample of broccoli to values close to 100% for garlic, potato, onion, spinach, cauliflower, tomato, and beans. Consequently, arsenic appears mainly in the form of highly toxic inorganic species, which is very different from the situation found in seafood products, where there is a preponderance of organoarsenical species. Seafood products are the food group for which there is most data about inorganic arsenic contents (*18, 21–24*). The highest inorganic arsenic content reported in fish and shellfish was 1.19 μg g^{-1} of ww in a sample of blue whiting (*24*), followed by a sample of peppery furrow, 1.05 μg g^{-1} of ww (*23*). Without taking into account these high values, the other contents mentioned in the bibliography are <1 μg g^{-1} of ww. The scarce difference between seafood and vegetables in terms of the levels of inorganic arsenic introduces new considerations in connection with the control of this contaminant in vegetables cultivated in areas with chronic arsenic poisoning.

Legislation and Health Considerations. With respect to legislation, Chile establishes two maximum limits for arsenic for the products analyzed in the present study. For cereals, legumes, and leguminous plants the maximum content permitted is 0.5 mg kg^{-1} of ww (*25*), a value not attained by the samples of beans and corn analyzed. The other vegetables must be compared with the value established by Chilean legislation for the food group "other solid products" (1 mg kg^{-1} of ww) (*25*), a value not exceeded by any of the vegetables considered in the present study. Consequently, all of the samples of vegetable products analyzed conform to Chilean regulations, and their sale should not be restricted. This conclusion has important social repercussions for the communities of Chiu Chiu and Lasana. The inhabitants obtain a considerable income from growing vegetables, but in recent years the problems of arsenic in the area have frequently led to questions being raised about the health safety of products grown in these villages and sold in other parts of the country.

The legislation that exists in the rest of the world concerning maximum permitted levels of total arsenic in vegetables varies considerably (*26*). Many countries have no legislation on this point. Those that do have legislation include vegetables in the group "fruit, vegetables, and their products". Various countries fix concentrations for all fresh vegetables, with the strictest levels being found in Poland (0.2 mg kg^{-1}), where some of the samples obtained from the Second Region in Chile could not be sold. For the legislation in Bulgaria, the Czech Republic, the Slovak Republic, and Hungary (0.5 mg kg^{-1}), only one sample of spinach leaf analyzed in the present study exceeded the maximum allowed. Other systems of legislation permit higher levels: 1 mg kg^{-1} in Guyana, Jamaica, Trinidad and Tobago, Kenya, Zambia, Malaysia, Singapore, and the United Kingdom; 1.5 mg kg^{-1} in Papua New Guinea. In some cases, specific controls have been established for certain fresh vegetables such as potatoes (0.2 mg kg^{-1} in Poland; 0.3 mg kg^{-1} in the Czech and Slovak Republics and Hungary; 1 mg kg^{-1} in Japan), root vegetables and leaf vegetables (0.2 mg kg^{-1} in Poland); and spinach, tomatoes, and cucumbers (1 mg kg^{-1} in Japan). In Spain, the maximum content of arsenic in canned vegetables is fixed by regulation (1 mg kg^{-1}). With respect to cereals, few countries set maximum limits, whereas a few data exist for corn flakes, flour, and other cereal meal products. For example, Polish

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legislation establishes a maximum level of 0.2 mg kg^{-1} for cereals and cereal meal products. Within the international context, Chilean legislation occupies a middle position in terms of its legislative requirements.

As far as inorganic arsenic is concerned, the absence of legislation in vegetable food products makes it impossible to compare the data found in the present study with established regulations. It is possible, however, to establish certain points of a toxicological nature. In the case of spinach, the vegetable with the highest inorganic arsenic content in the study, it would be necessary to consume a quantity in excess of 1.7 kg a week (250 g/day) in order to reach the Tolerable Daily Intake (TDI) recommended by the WHO for a person with a body weight of 68 kg (0.146 mg of inorganic arsenic/day). The high consumption required does not make the products analyzed harmful, in principle. However, considered as part of a food diet, the "vegetable" group may make a considerable contribution to the intake of inorganic arsenic in populations such as those considered in the present study. An adequate estimate of intake would require evaluation of inorganic arsenic contents in cooked products, which may differ substantially from the levels in raw products. In this connection, the water used in these areas to cook such products contains arsenic, and studies on seafood products performed by our laboratory (*27*) have shown variations in concentrations of inorganic arsenic during cooking. To estimate intake it would also be necessary to carry out nutrition surveys to discover eating habits and the consumption of each product.

It is generally admitted that total arsenic intake depends on the quantity of seafood products in the diet, which seems to make seafood products the target for research connected with arsenic. However, as has been shown, the situation for inorganic arsenic is different. Populations in certain regions, including those in the Second Region in Chile, that consume a diet low in fish, may present a relatively high intake of inorganic arsenic, derived first from drinking water and second from high consumption of vegetables and cereals. Research into inorganic arsenic should therefore be extended to food matrices other than fish. It would be extremely useful to have data concerning the baselines of inorganic arsenic in vegetables from areas such as the one studied. Such data would provide the first step for research that, by calculations of intake and epidemiological studies, could lead to specific legislative control of inorganic arsenic in vegetables produced or consumed in those areas.

Conclusions. All of the vegetables analyzed presented total arsenic contents below the maximum limits permitted by Chilean legislation. The values found for inorganic arsenic were close to the levels of total arsenic, which indicates the predominant presence in the vegetables analyzed of highly toxic species of arsenic. When considered as part of a food diet, these vegetables may make a considerable contribution to the intake of inorganic arsenic. For a more realistic evaluation of the toxicological risks for populations exposed to high levels of arsenic in the environment, there is an urgent need to quantify the inorganic arsenic contributed by food products, with special reference to products of vegetable origin.

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