original article

Heavy metal content in edible salts in Isfahan and estimation of their daily intake via salt consumption

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ABSTRACT

Aims: In this study, the heavy metal contamination of consumable table salt, both unrefined and refined, was investigated. The provisional tolerable weekly intake (PTWI) of heavy metals just by edible salt, of the Isfahan population, was also estimated.
Materials and Methods: Twenty samples of salt, including 15 refined and five unrefined were analyzed. Precision of the analysis was assured through repeated analysis of the five samples, which had a great demand in the city. The heavy metal content in the samples was analyzed with Flame Atomic Absorption Spectrometry (AAS) and Inductively Coupled Plasma (ICP). The PTWI of the metals was calculated by a formula and by using the Iranian average body weight.
Results: The mean and standard deviations of cadmium (Cd), lead (Pb), arsenic (As), mercury (Hg), copper (Cu), and zinc (Zn), in refined table salts were 0.15 ± 0.02, 0.57 ± 0.1, 0.69 ± 0.09, 0.061 ± 0.008, 0.87 ± 0.11, and 6.34 ± 1.08 μg/g, and those in the unrefined ones were 0.16 ± 0.02, 0.61 ± 0.13, 0.63 ± 0.07, 0.058 ± 0.004, 0.86 ± 0.06, and 7.53 ± 2.93 μg/g, respectively. A PTWI via salt consumption was in the range of 0.8 – 3.1 percent.
Conclusions: There was a significant difference between the heavy metal concentrations and their guideline values. Estimation of the health risk due to heavy metals was not possible as PTWI showed total intake of a metal by total food consumption during a week. Therefore, it was important to assess the public health risks arising from the presence of these toxic contaminants in the foods consumed by the population of Iran.

Key words: Contamination, dietary exposure, heavy metals, PTWI, refined/unrefined salts

INTRODUCTION

The effects of environmental pollution on food contamination and its safety for human consumption are serious worldwide public issues and widely addressed.[1] Heavy metals, an inorganic heterogeneous group of compounds, which arise from different environmental sources, are significant in nutrition.[2-4] The food chain and diet are major exposure paths for trace metals in most people compared to other ways of exposure, such as, inhalation and dermal contact.[4-7] Owing to their non-biodegradable and persistent nature, heavy metals accumulate in the vital organs of the human body such as kidneys, bones, and liver, and are associated with numerous serious health disorders.[6] As there is no good mechanism for heavy metal elimination, the chronic intake of it, in low-levels, has a damaging effect on human beings and other animals.[5] The most widely distributed environmental poisons include lead, cadmium, mercury, and the metalloid arsenic, which are persistent contaminants that have been suggested to be toxic (acute, chronic or sub-chronic), neurotoxic, carcinogenic,

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mutagenic, and teratogenic.[3,6] Moreover, they have been associated with gastrointestinal disorders, ataxia, paralysis and convulsion, depression, and pneumonia.[6] Some of them, such as, copper and zinc are toxic only at high concentrations, whereas, they are essential for normal body functions.[4,8]

One of the biological necessities for humans is salt, which is added to a majority of foods, not only for improving the taste, but also as a preservative; generally to canned, salted, and pickled or fresh foods.[1,9] The average daily salt intake in most countries around the world is approximately 6 g/day per capita.[10,11] However, its intake in children older than five years is commonly more than that and increases with age.[12]

There are two types of edible salts used for food processing and cooking present in Iran's market: table salt, a fine-ground, refined, crystallized rock salt, with nearly pure sodium chloride, and kitchen salt, a packed salt obtained from unrefined ground rock salt.[1] As edible salt is prepared from rock salt, the concentration of heavy metals in table salt should be rigorously controlled.[9] Several researches have reported the presence of trace elements in the salts consumed by humans, which include: Aluminum (Al), Arsenic (As), Barium (Ba), Bromine (Br), Cadmium (Cd), Cerium (Ce), Chlorine (Cl), Cobalt (Co), Chromium (Cr), Cesium (Cs), Copper (Cu), Europium (Eu), Iron (Fe), Hafnium (Hf), Lanthanum (La), Manganese (Mn), Sodium (Na), Nickel (Ni), Lead (Pb), Rubidium (Rb), Antimony (Sb), Scandium (Sc), Samarium (Sm), Strontium (Sr), Tantalum (Ta), Terbium (Tb), Thorium (Th), and Zinc (Zn).[1,4,9,13-15] Although, direct human use of rock salt has been discouraged by the health authorities, its existence in the markets indicates that this type of salt is still being used by the consumers, especially in developing countries.[4,9]

Trace heavy metal ions in environmental samples including natural water, air, soil, fertilizers, and table salts have been repeatedly analyzed by several investigators such as Soylak *et al*., Cheraghali *et al*., Amorim *et al*., Steinhauser *et al*., and Peker *et al*.[4,9,13-15]

The aim of this study was to determine the heavy metal contamination of table salt consumed in both rock salt and refined salt. The dietary exposure level of the Isfahan population to heavy metals just by edible salt was also estimated.

MATERIALS AND METHODS

Sampling

Fifteen refined pre-packed salt (RFS) samples were purchased directly from shops in the city of Isfahan and five samples of rock salt (unrefined) (URS) were obtained from vendors around the city. The Pb, Cd, Hg, As, Cu, and Zn contents of all the samples were analyzed using the atomic absorption spectrometry (AAS) and inductively coupled plasma (ICP) systems.

Reagents

All materials used were purchased from the Merck Company and had analytical purities. Nitric acid was used for the digesting of samples. 4-methyl-2 pentanone (Methyl Iso-Butyl Ketone: MIBK) and ammonia pyrolidine dithiocarbamate (APDC) were applied for analyzing Pb and Cd. Sodium borohydride ($NaBH_4$) and Stannous Chloride or Tin Chloride ($SnCl_2$) were also used for Hg analysis. All solutions were prepared using double-deionized water (DDW).

Instruments

Analyzing of Hg was performed using a Perkin Elmer 4100 atomic absorption spectrometer equipped with a hydride generation (HG) system and a halo cathode lamp (HCL) as a source of the determination cathode ray tube. Lead and Cd were also determined by the same atomic absorption spectroscopy, equipped with a graphic furnace and a proper hollow cathode lamp (HCL). Determination of As, Cu, and Zn was implemented by ICP model 2 (Jobin Yvon Ultima), with an ICP V5.10 software.

Analyses of Mercury, Arsenic, Copper, and Zinc

Each salt sample of 4 g was mixed with 5 ml of nitric acid (HNO_3) (1 N) and heated on a hot plate to 150°C for 10 minutes, for digestion. The digested samples were then diluted with 100 ml double-deionized water. For reducing mercury to metallic Hg, $NaBH_4$ and $SnCl_2$ were added to the mixture after adding nitric acid. As Cu and Zn were measured using the ICP system, Hg was measured using cold vapor AAS.[1,9]

Analysis of cadmium and lead

As direct determination of Cd and Pb using flame atomic absorption spectrometry (FAAS) was not possible, a reported extraction method was used for analysis of the salt samples.[9] Firstly, 2 ml of HNO_3 was added to 0.5 g of each salt sample and then diluted to 50 ml using DDW. Following that, MIBK was saturated with DDW. Next, 2 g of APDC were dissolved in 50 ml of DDW, and 20 ml of MIBK solution was added to the APDC solution and mixed until the two phases were separated. Separation with 20 ml of MIBK was repeated and the phases were separated again. The organic phases of the two separation stages were mixed together. Finally, 2 ml of the organic solutions were mixed with 4 ml of the prepared acidified salt solution, and the organic phase was separated again and measured with an AAS equipped with a graphic furnace.[16]

Quality assurance

Precision of analysis was assured through repeated analysis of the five samples. The standard deviations of results for Cd, Pb, As, Hg, Cu, and Zn were found to be ±(0.002, 0.003, 0.008, 0.065, 0.023, and 0.004) (μg/g) of the averages of the previous analysis value, respectively.

Estimation of dietary exposure to heavy metals

The dietary intakes of the studied heavy metals were estimated as follows, and their associated risks were evaluated by comparing with the provisional tolerable weekly intakes (PTWIs). The exposure was expressed per kilogram of body weight by dividing the total dietary exposure by the average body weight of the population.[3,17]

Daily intake of heavy metals = ∑[concentration of heavy metals in both the studied salts × mean salt intake (g/person/day)]

Weekly intake of heavy metals = daily intake × seven days/week

Weekly intake per body weight (kg) (PTWIs) = weekly intake ÷ reference body weight (60 kg)[17]

RESULTS

The concentration of the desired metals in the refined and unrefined salt samples and their comparison with standards are shown in Table 1, and the dietary exposure of the Iranian population to heavy metals via salt consumption is summarized in Table 2.

DISCUSSION

Heavy metal concentration in salts (RFS and URS)

The results indicated that no statistically significant difference was found between the studied heavy metals' mean concentrations, in refined and unrefined salts ($P_{value} > 0.05$). As shown in Table 1, the cadmium level of the samples was found to be in the range of 0.112 – 0.195 μg/g with the RFS samples having the highest level. However, its mean concentration was lower than the guidelines regulated by the Iranian Institute of Standards and Industrial Research and Codex[18,19] ($P_{value} < 0.001$). Soylak *et al.*, reported that the cadmium content of table salts was in the range of 0.14 – 0.3 μg/g.[4] Peker *et al.*, reported that its concentration was up to 0.5 μg/g.[15] Narin *et al.* found it to be in the range of 0.1 – 0.12 μg/g.[24] Similar researches conducted in Tehran found it to be in the range of 0.65 – 0.91 μg/g,[1] and 0.024 μg/g.[9]

Lead level was found to be in the range of 0.386 – 0.853 μg/g, which was lower than the guidelines. However, the average of its concentration in the refined salts was slightly higher than that in the unrefined ones[18,19] ($P_{value} < 0.001$). Amorim *et al.* reported that the lead content of the salts was up to 0.106 μg/g.[13] In Tehran salt it was reported to be in the range of 0.87 – 1.6 μg/g[1] and 0.438 μg/g.[9]

The arsenic and mercury levels of the investigated salts were in the range of 0.511 – 0.894 μg/g and 0.05 – 0.08 μg/g, respectively. Their highest level was found in a refined sample, however, its mean concentration was lower than the guidelines[18,19] ($P_{value} < 0.001$). Cheraghali *et al.*, has reported 0.094 μg/g of arsenic and a concentration 0.021 μg/g Hg in table salt.[9] The copper level in the studied table salt was in the range of 0.654 – 1.08 μg/g. Its concentration in the RFS was the highest, but its average in the samples was lower than in the guidelines[18,19] ($P_{value} < 0.001$) [Table 1]. Soylak *et al.*, reported it to be in the range of 0.14 – 0.3 μg/g.[4] A range of 0.7 – 0.8 μg/g was also reported by Narin *et al.*,[24] up to 0.3 μg/g by Peker *et al.*,[15] and in the range of 1.21 – 1.24 μg/g by Khaniki *et al.*[1]

Zinc like Cu is an essential element in low concentrations and it is regulated by the physiological mechanisms in most organisms. However, in an excessive concentration it is known to be a potential hazard that can endanger both animal and human health.[25] The determined range of this heavy metal was 4.52 – 11.9 μg/g. The highest amount was in URS, but its mean in the samples was lower than the guideline value[18,19] ($P_{value} < 0.001$) [Table 1]. Cheraghali *et al.*, reported the Zn concentration of salts to be in the range of 6.02 – 6.5 μg/g.[9]

Evaluation of dietary exposure level to heavy metals via salt consumption

Intake of heavy metals via the food chain is widely addressed, therefore, understanding their health risks via salt consumption, as it is one of the essential additives in all food content, seems to be absolutely essential. Then, the dietary intake of each heavy metal was estimated, and the associated risk was evaluated by comparing the intake with the provisional tolerable weekly intake (PTWI). Actually, as the PTWI inferred, the provisional tolerable weekly intake of the metal during a week through total diet, and a real comparison between the results obtained and the PTWI was not possible. Therefore, calculating the percentage of the heavy metals' PTWI, via salt consumption — or any other content — seemed to be feasible.

The calculated PTWI for Cd was 0.11 μg/kg body weight/week, which was lower than the recommended one by

Table 1: Concentration of heavy metals in the investigated salts

Metals	Range(μg/g)		Mean ± SD(μg/g)		Iran(*) (μg/g)	Codex(**) (μg/g)
	RFS	URS	Refined	Unrefined		
Cd	0.112 – 0.195	0.135 – 0.183	0.15 ± 0.02	0.16 ± 0.02	0.5	0.2(a)
Pb	0.386 – 0.853	0.478 – 0.787	0.57 ± 0.1	0.61 ± 0.13	1	1
As	0.511 – 0.894	0.527 – 0.715	0.69 ± 0.09	0.63 ± 0.07	0.5	0.5
Hg	0.05 – 0.08	0.05 – 0.06	0.061 ± 0.008	0.058 ± 0.004	0.1	0.05
Cu	0.654 – 1.08	0.796 – 0.964	0.87 ± 0.11	0.86 ± 0.06	2	2
Zn	4.67 – 8.95	4.52 – 11.9	6.34 ± 1.08	7.53 ± 2.93	-	-

*Adapted from ISIRI, Food and Feed — Maximum limit of heavy metals.[18] **Adapted from, Codex alimentarius [19]

Table 2: Summary of dietary exposure of Iranian population to heavy metals due to edible salt consumption

Heavy metals	Daily intake (µg/ person)	Weekly intake(WI)		PTWI (µg/kg BW)	(WI/PTWI) × 100 (% intake via salt)
		(µg/ person)	(µg/kg BW)		
Cd	0.93	6.51	0.11	7*	1.57
Pb	3.54	24.78	0.41	25**	1.64
As	3.96	27.72	0.46	15***	3.07
Hg	0.36	2.52	0.04	5**	0.8

BW = Body weight (estimated mean body weight of the Iranian population: 60 kg was used[20]. *Adapted from FAO/WHO, 2003[21], **FAO/WHO, 1993[22], ***FAO/WHO, 1989

the Joint FAO/WHO Expert Committee on Food Additives (JECFA).[21] Thus, human intake of Cd by the edible salt consumption was only 1.57 percent of the PTWI [Table 2]. Similarly, the calculated PTWI for lead, arsenic, and mercury was 0.41, 0.46, and 0.04 µg/kg body weight/week, respectively, which was lower than the recommended ones.[22,23] Moreover, as shown in Table 2, they included 1.64, 3.07, and 0.8 percent of the PTWI values. There was no recommended PTWI by the related organization for zinc and copper, therefore, their PTWI was not calculated. Considering the percentage of PTWI, the level of heavy metals in the salt samples seemed to be in a suitable range.

The results showed that the concentration of arsenic in some of the unrefined and refined edible salts in Isfahan, Iran, were high and exceeded the maximum allowance concentration of metals recommended by the Codex and Iranian Institute of Standards and Industrial Research. However, their mean concentration was lower than that in the guidelines. The highest level of the contents of trace elements in the investigated table salts was found in refined table salt, and the importance of the fact appeared when we found out that the salt that had the highest metal content had the most consumers in Isfahan. This showed that it needed more deliberation by a responsible organization to use a suitable method for purification during salt processing.

In this study, the percent of the heavy metals with regard to the PTWI a person receives, found just by salt consumption, was in the range of 0.8 – 3.1 percent. Actually the estimation of the health risk due to the four studied heavy metals (Cd, Pb, As, and Hg) comprise of only a percent of the total PTWI. An investigation of the Iranian food basket, to assess public health arises for intake of all heavy metals seems to be necessary.

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