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ORIGINAL ARTICLE

Assessment of heavy metal content in refined and unrefined salts obtained from Urmia, Iran

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Abstract

A heavy metal is any relatively dense metal that may be potentially toxic in a variety of foods. Heavy metals pollute and contaminate foods. These metals are usually toxic to human body. Heavy metals are the most important toxic metals which may cause health risks following the consumption of contaminated foods. The edible salt (NaCl) is a substance that has been used as a food additive since ancient times. Twenty samples of refined and unrefined edible salts produced in Iran were analyzed using flame atomic absorption spectrometry (FAAS) method for the presence of toxic heavy metals. The precision of the analysis was assured through the repeated analysis of the samples. The mean (±standard deviation) concentrations of toxic metals in dried samples of rock salt were as follows: Ni (1.870 ± 0.850), Cd (0.328 ± 0.143), Mn (0.184 ± 0.230) and Co (3.124 ± 0.880) mg/kg and in dry weights of samples obtained from Urmia market: Ni (1.982 ± 0.021), Cd (2.461 ± 0.036), Mn (0.192 ± 0.028) and Co (8.450 ± 0.025) mg/kg. There was a significant difference between the toxic metal concentrations and their guideline values. Therefore, it was important to assess the public health risks posed by the presence of toxic contaminants.

Keywords

Atomic absorption spectrometry, toxic metal, edible salt, safety, Iran

History

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Introduction

Although food-borne diseases are caused by food contaminated with microorganisms and may be caused by toxins including toxic metals, they can also have acute toxicity and chronic toxicity (Zhang et al., 1997). Therefore, it is very important to assess the related risks of dietary intake of these elements in food. The use of salt as a flavoring and preservative food has been concern from ancient times, although nowadays the consumption of salt is very high in developing countries (Ruusunen & Puolanne, 2005). The edible salt can be divided into two categories: refined and unrefined. The unrefined salts are prepared from salt lakes and are used for food by people of many countries including Turkey (Soylak et al., 2008). The absorption of toxic metals in the diet can be seen both in inorganic forms, through the corresponding salts, and as constituents of organic molecules (proteins, fats, carbohydrates and nucleic acids). Some toxic metals (i.e. iodine, manganese, copper, iron and selenium) are essential to health, whereas others (i.e. beryllium, mercury, lead, cadmium, aluminum) are toxic or have no known beneficial effects (Farahani et al., 2015; Fathabad et al., 2015; Kamkar et al., 2010; Mohammadpourfard et al., 2015). Even the toxic metals with beneficial effects are dangerous if consumed in large quantities (Coultate, 1990; Goyer, 1995). Some toxic metals may be natural and other toxic metals may be natural for human activity which is presented in foods (manuring practices, industrial emissions, exhausted gases, etc.).

Heavy metals may be present either naturally in foods, or by the result of human activities (e.g. manuring practices, industrial emissions, exhausted gases, etc.) or by contamination during industrial processes, preservation and cooking (Crosby, 1977). There is a relationship between the long-term effects of heavy metals on health and the presence of toxic metals in foods; thus, it is essential, in the interest of public health, to hold the toxicologically acceptable levels (Goyer, 1995). The most significant side effects for consuming the contaminated food with some toxic metals such as chromium are dermatitis and skin irritation to mucous membranes (Saracoglu et al., 2001). Nickel is a known haematotoxic, immunotoxic, neurotoxic, genotoxic, reproductive toxic, pulmonary toxic, nephrotoxic, hepatotoxic and carcinogenic agent (Aktas & Ibar, 2005).

Manganese toxicity can cause kidney failure, hallucinations, as well as diseases of the central nervous system (Yaman et al., 2004). Chronic exposure to cobalt by inhalation in humans results in effects on the respiratory system, such as respiratory irritation, wheezing, asthma, decreased lung function, pneumonia and fibrosis (Saulea et al., 2004). Cadmium exposure may cause bronchitis, chemical

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pneumonitis and pulmonary edema. For refining impurities in rock salt, usually ''minor or washing with water filtration and purification methods or complete recrystallization are used. In Iran, one of the most common ways for purifying salt is washing with water (Elsagh & Rabani, 2010a). Several researchers from Iran and other countries reported the presence of trace elements in the salt Heshmati et al (2014) from Iran reported that the heavy metal concentrations were generally higher in bakery refined salt. Also, the conducted study by Lugendo et al (2013) showed that the concentrations of all toxic elements found in the study samples were higher than their maximum tolerable limits set by international organizations. Furthermore, in Iran, from two differet cities of Tehran and Isfahan salts samples, zinc content was reported to be 6.50 μg/g (Jahed Khaniki et al., 2007) and 6.34 μg/g (Pourgheysari et al., 2012).

The aim of this study was to determine the contents of nickel, chromium, manganese and cobalt in refined salt and unrefined rock salt in Iran in the region of Urmia in 2013.

Materials and methods

Chemicals and sample collection

All chemicals substances (of analytical reagents) were purchased from Merck, Germany. All samples in this study were prepared twice with distilled water. The standard solutions of nickel, chromium, manganese, cobalt and sodium, HCl (1%), (2%) HNO_3 and water were made. Therefore, in order to eliminate any possible contamination, all glasswares were washed with 1:1 nitric acid and distilled water before use. Washing and drying were done for 4 h at 60 °C. Twenty samples from salt, including refined and unrefined salts, were prepacked. The samples were purchased directly from the shops around the city of Urmia. In fact, at least one of the samples of refined and unrefined salt was collected from each registered producer of the refined salt in the country.

Sample preparation

To prepare a saturated solution of salt, 36 g of the rock salt in ground temperature (30 °C) was dissolved in 100 mL of water (Lide, 1992; Weast, 1987). Then, rock salt powder with a certain amount of water was dissolved and stirred for 30 s. Next, the saturated salt solution was made and the resident was left for 20 min. Afterwards, the solution was centrifuged. Finally, the production of salt was kept in a dryer with hot air at 300 °C (Soylak et al., 2008).

Metals analysis

A Varian AA 240 atomic absorption spectrometer equipped with VGA 77, (Varian Atomic Absorption Spectrometers, Australia) was used to measure the amounts of the metals. For this purpose, the calibration curves for each metal were drawn separately by using the standard solutions of certain wavelengths of the maximum concentrations of the metals by an atomic absorption spectrometer (Elsagh & Rabani, 2010b; Soylak et al., 2008).

To determine the amount of any of the salts such as nickel, chromium, manganese and cobalt, certain volumes of each solution should be injected into the device. Salt solutions were prepared from each sample (for the initial solution of 2 g of each sample was dried salt in 50 mL volumetric flask and brought to volume in a two-stage dilution) to the device to absorb quantities and concentrations were read. The purity of the samples of salt sodium was calculated based on a percentage. In order to make an initial solution, 3 g of each sample was dried in 50 mL volumetric flask and the volume was completed in two steps of dilution. A sample was made by injection machine and the amount of the absorption and concentration was read. The read concentrations were converted by inserting the above relation to the actual concentration of the metal salt microgram per gram dry weight of salt (Berman, 1990).

$C_r = C_i \times V/W$ (Zhang et al., 1997)

C_r, the actual concentration of the sample (per mg, micrograms per gram dry weight); C_i, concentrations obtained from the device using a calibration curve (milligrams per liter); V, the sample size per ml (including dilution) and W, dry weight used (g).

Accuracy of tests

To ensure the accuracy of the tests, they were conducted three times for each sample and the average results were recorded. Also, to ensure the accuracy of the analytical methods of testing and measuring the correct amount of metals in samples obtained from the standard method of increasing, the percentage recovery of metals was used. In this study, 10 ml of the standard solution of Ni, Cd, Mn, Co and Na 1 mg per g of each sample were tested. It should be noted that the two samples for each metal, similar to the same conditions producing only one solution standard, was added. The concentration of each distinct (by extrapolation of the calibration) and the percentage recovery of the metals was calculated by the following formula (Association of Official Analytical Chemists, 1984).

$R = 100(A2–A1)/As$

R, percent recovery; A1, without standard samples (μg/g); A2, standard samples (μg/g) and As, the concentration of the standard solution (μg/g).

Sensitivity assay

The sensitivity of analytical methods can be described by limit of detection values (LOD) and limit of quantification (LOQ). The LOD is the lowest concentration of analyte that can be detected and reliably distinguished from zero, but not necessarily quantified. The LOQ can be defined as the lowest concentration of analyte that can be determined quantitatively with an acceptable level of precision (Gonzalez & Herrador, 2007).

Statistical analysis

All statistical analyzes were performed using SPSS software (Version 16; SPSS Inc., Chicago, IL) and the data were expressed as mean ± standard deviation (SD). We compared the metal contents of unrefined and refined salts using Mann–Whitney U-test. Significance level was set at $p < 0.05$.

Results and discussion

The results of the samples analysis have been summarized in Tables 1 and 2. Linearity was demonstrated by analyzing five different concentrations of Ni, Cr, Mn, Co and Na. The accuracy of the method was determined by calculating the recoveries of the metals. To ensure the accuracy of the analytical method, the recovery studies were carried out by adding a known quantity of Ni, Cr, Mn, Co and Na into the samples by the proposed method. Also, to check the accuracy of analytical method, the recovery studies were performed in order to confirm the losses of Ni, Cr, Mn, Co and Na or contamination during sample preparation and matrix interferences during the measurement step (Table 3). The mean concentrations of the tested tracer metals including Ni, Cd, Mn and Co have been summarized in Table 4 showing comparative analysis of the heavy metal contaminations of the unrefined and refined salts. No statistically significant difference was found between the two groups ($p > 0.05$). Table 4 shows the amount and purity of Na salts. It refers to the standardization and percentage increase for metal recovery. Table 5 shows the purity of the calculated samples. According to the percentages of metals recovery ($100 \pm 6\%$), it is concluded that the method used to determine the metals had enough confidence.

Heavy metals are the most important metals, which may cause health risks following the consumption of contaminated foods. Salt is one of the most used additive foods with a unique place in food consumption (Cheraghali et al., 2010). In our study, the mean concentrations of heavy metals of the refined salt table were Ni (1.870 ± 0.850), Mn (0.184 ± 0.230), Cd (0.328 ± 0.143) and Co (3.124 ± 0.880) mg/g in their dry weight, and for the unrefined salt they were Ni (1.982 ± 0.021), Cr (2.461 ± 0.036) and Co ($8.450 \pm 0/025$) mg/g based on their dry weight obtained, respectively. In the case of manganese salts, the lowest and highest values were accounted. Food contamination is a major route of human exposure and may represent a serious threat to human health. The presence of the heavy metals in salt can be harmful to human health (Silva et al., 2005).

Table 1. Terms device used to measure element method FAAS.

Ingredients	Wave length (nm)	Fission width (nm)	Flow (mA)
Ni	242.0	0.30	4.0
Cr	367.9	0.30	7.0
Mn	289.5	0.30	5.0
Co	250.7	0.30	7.0
Na	589.0	0.60	5.0

FAAS: flame atomic absorption spectroscopy.

Table 2. Standardization and percentage increase metal recovery.

Metal	Without standard samples (μg/g)	Standard concentration was added (μg/g)	Increasing concentrations of standard samples (μg/g)	Percent recovery
Ni	1.08	1	2.04	96
Cr	1.35	1	2.40	105
Mn	0.58	1	1.52	96
Co	2.42	1	3.45	103
Na	0.34	1	1.32	98

Table 3. Linearity range and LOD, LOQ.

Ingredients	LOD (μg/mL(g))	LOQ (μg/mL(g))	Linearity range (μg/mL(g))
Ni	0.022	0.075	0.2–1.6
Cr	0.025	0.075	0.3–1.8
Mn	0.02	0.06	0.1–1.7
Co	0.033	0.09	0.5–3
Na	0.02	0.65	0.1–1.4

Nickel is one of the most heavy metals that is accumulated in bodies of human and can cause serious illness and some studies indicate that its excessive intake has adverse effects on different organs such as skin, liver, gastrointestinal tract, kidneys, respiratory system, and neurological disorders (Eftekhari et al., 2014; Soylak et at al., 2008). In the present study, nickel concentrations were 1.982 μg/g and 1.870 μg/g in unrefined and refined salt samples, respectively. Moreover, in another study that was performed by Eftekhari et al. (2014) in Iran, they reported that the concentrations of nickel in recrystallized and washed salt samples were 0.15 and 0.13 μg/g, respectively. In the previously conducted survey in Iran, by Elsagh (2012), the nickel concentration was shown to be 1.860 μg/g in refined table salt. However, several studies have been performed in different countries for investigating the heavy metals contamination in food chain especially in salts. The previous study by Soylak et al. (2008) from Turkey, Egypt and Greece revealed that the nickel content of refined table salt samples were 0.16–1.57 μg/g, but our results were higher than this.

Cobalt is a usually beneficial element compared with other metals. Accordingly, the toxicity is low and is an essential trace element which is used in the treatment of anemia. However, increasing its level has side effects on the fetus growth (Eftekhari et al., 2014; Soylak et al., 2008). Elsagh (2012) from Iran reported that the cobalt content of refined table salts samples was in the range of 0.83–3.74 μg/g. Therewith, the similar study by Eftekhari et al. (2014) revealed that in recrystallized and washed samples, the cobalt concentration was 0.008 and 0.037 μg/g, respectively. Furthermore, in our unrefined and refined samples, the cobalt concentrations were reported between 8.450 and 3.214 μg/g, respectively. The previous study by Soylak et al. (2008) found that the cobalt content was in the range of 0.22–0.48 μg/g, but our results were higher than this.

Cadmium is a widespread environmental pollutant. Its exposure induces the causes of several illnesses such as bone damage, osteoporosis and renal tubular dysfunction that lead to renal failure in the long run. In addition, it has a serious problem in relation with several cancers (Eftekhari et al., 2014; Soylak et al., 2008).

In the present results of unrefined and refined samples, cadmium contents were found to be 2.461 and 0.328 μg/g, respectively. The previous survey from Iran by Jahed Khaniki et al (2007) reported that the cadmium content of table salts was in the range of 0.65–0.91 and the similar study by Zarei et al. (2011) from Iran found the 0.01–0.4 range. In another

Table 4. Toxic metals concentrations in micrograms per gram dry weight of salt samples.

	Unrefined salt				Refined salt			
Metal	Mean	SE	Min	Max	Mean	SE	Min	Max
Ni	1.982	0.021	1.970	2.011	1.870	0.850	0.590	2.214
Cr	2.461	0.036	2.420	2.492	0.328	0.143	0.195	0.482
Mn	0.192	0.028	0.175	0.215	0.184	0.230	0.027	0.352
Co	8.450	0.160	8.375	8.682	3.214	0.880	1.83	3.462

N denotes the number of repetitions ($N = 9$).

Table 5. The amount and purity of Na salts.

Quantity	Refined Salt (% w/w; $n = 6$[a])	Unrefined salt (% w/w; $n = 6$[a])
Na	38.873 ± 0.037	38.140 ± 0.024
Purity	98.830 ± 0.016	96.985 ± 0.053

[a]The occurrence (mean ± standard error), $p < 0.05$.

report by Soylak et al. (2008), it was revealed that cadmium content was 0.014–0.030 in refined and unrefined table salt samples from Turkey, Egypt and Greece. Cadmium level of table salts from Brazil was found to be in the range of 0.01–0.03 by Amorim and Ferreira (2005). The concentration of cadmium of table salts from Iran was reported to be in the range of 0.112–0.195 by Pourgheysari et al. (2012). Moreover, these results showed that the refined table salt samples had the highest level. According to Iranian food standards and Codex legislation, the maximum permitted levels of cadmium in food grade salt are 0.2 and 0.5 μg/g, respectively.

Manganese is a ubiquitous element in the environment and is widely used throughout the industry. Although manganese has a relatively low toxicity, in chronic overdose or prolonged occupational exposure, it has been associated with neuropsychological and neurological squeal in exposed workers (Bowler et al., 2016; Soylak et al., 2008). In the present study, the manganese concentrations were found to be 0.192 and 0.184 μg/g in unrefined and refined salt samples, respectively. The previous investigated results by Soylak et al. (2008) reported that the manganese content of 28 of refined and unrefined table salt samples from Turkey, Egypt and Greece was 0.26–4.68 μg/g.

Conclusion

It was found that the content of the toxic metals in refined salt was lower than the maximum amount set by Codex. Moreover, the public health authorities do not allow the salt with high levels of heavy metals to be consumed. Impurities and/or insoluble substances, which have adverse effects on human health, may be present in unrefined salts because of the presence of higher concentrations of toxic metals in the salts. Since high consumption of refined salt by consumers in dormitories, restaurants, hospitals, etc. as a food additive is costly, the use of unrefined salt must be checked and controlled by the Iranian health authorities because it may be considered as a harmful breach.

Declaration of interest

The authors report no conflict of interest. The authors alone are responsible for the content and writing of this article.

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