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Food Salt Characterization in Terms of Radioactivity and Metals Contamination

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Abstract: The analysis of food salt is very important because of its high consumption by the population, for both medicinal and nutritional use. In this study, nine different samples of food salt (Cyprus black, Himalayan pink, Hawaii red, iodized, hyposodic iodized, Maldon smoked sea, common sea, Breton sea and Persia blue), coming from large Italian retailers and employed by people for different cooking food purposes, were investigated through High Purity Germanium (HPGe) Gamma Spectrometry in order to evaluate the anthropogenic (^{137}Cs) and natural (^{40}K) radioisotopes activity concentration, and used Inductively Coupled Plasma Mass Spectrometry (ICP-MS) in order to assess any possible metals contamination by a comparison between Cu, As, Cd, Hg and Pb concentrations and the limits set by the Italian Legislation. The evaluation of dose levels due to the salt ingestion for the age category higher than 17 years was performed taking into account the human body daily need of about 10 g of salt, and in the precautionary hypothesis, this need was satisfied from a single type of salt. All obtained results are under allowable levels (1 mSv/year), thus excluding the risk of ionizing radiation effects on humans. Regarding to the metals concentration, experimental results show that it is lower than the contamination threshold values, thus excluding their presence as pollutants.

Keywords: food salt; radioactivity concentration; metals contamination; high purity germanium (HPGe) gamma spectrometry; inductively coupled plasma mass spectrometry (icp-ms); effective dose; ingestion

1. Introduction

Human beings are subjected to radiations coming from natural and artificial sources in their living environments [1]. Natural radioactivity is due to the presence of cosmogenic and primordial radionuclides in the Earth's crust [2]. Artificial fallout radionuclides, such as ^{137}Cs, are derived mainly from global nuclear tests conducted between the mid 1940s and the 1980s, as well as from nuclear accidents [3]. In addition to its ionizing effects, ^{137}Cs may be toxic; it can undergo bioconcentration and bioaccumulation and adversely impact human and ecosystem health [4].

There are three ways of exposure to ionizing radiations for humans: external gamma rays, inhalation of radon and other radioactive nuclides and ingestion of radioisotopes through food and water [5]. For the latter, in particular, the natural radioactivity in food comes mainly from ^{40}K; uranium and thorium daughter products are usually present in traces [6–8]. Ingested or inhaled radionuclides are distributed among body organs (according to the metabolism of the element involved and the organs), normally exhibiting varying radio sensitivities [9].

Salt is one of the most important compound in the life of organisms as it is necessary for humans, animals and plants; due to its importance for large cells of the body and metabolism in the cell, it is a

necessity for survival. It can be extracted from the sea and from mines and, with its derivative, it is largely used in many industries [10]. The human body needs about 10 g of salt daily, so the analysis of food salt in terms of chemical and radioactive contamination is extremely important to safeguard the human health [11]. Consuming contaminated food increases the amount of radioactivity and chemical contamination inside an individual, and therefore, it increases the health risks associated with radiation exposure and metals pollution [12–14]. The exact health effects depend on the type and the quantity of ingested contaminants [15,16].

In this article, nine different samples of food salt (Cyprus black, Himalayan pink, Hawaii red, iodized, hyposodic iodized, Maldon smoked sea, common sea, Breton sea and Persia blue), available at the Italy markets for their wide use in the various dietary regimes typical of the Italian population, were chosen to be analyzed to identify and quantify natural (^{40}K) and artificial (^{137}Cs) gamma-emitting radionuclides with HPGe gamma spectrometry. In order to evaluate any possible radioactive contamination and to estimate the effective dose due to the salt ingestion, metals (Cu, As, Cd, Hg and Pb) with Inductively Coupled Plasma Mass Spectrometry (ICP-MS) were used to assess any possible chemical pollution.

Food salts available at Italy markets have never been analyzed in terms of radioisotopes and metals contamination; so, as a consequence, the health risks for the Italian population have never been evaluated. Thus, no similar reports are present in the literature.

2. Materials and Methods

2.1. Samples Description

Cyprus black, Himalayan pink, Hawaii red, iodized, hyposodic iodized, Maldon smoked sea, common sea, Breton sea and Persia blue food salts were analyzed in this study. They are shown in Figure 1.

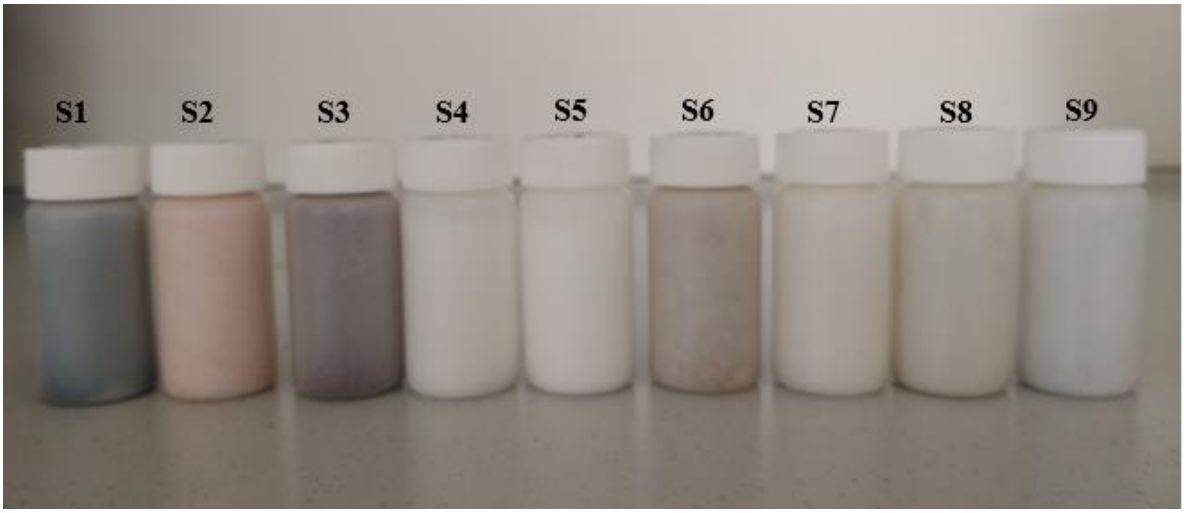

Figure 1. Photo of the investigated samples.

The first one is made by salt in flakes and charcoal. It has a light and delicate taste and is ideal for flavoring fish, potatoes, eggs and soups, and is used to make original decorations. The benefit to human health linked to its regular use consists in the absorption of intestinal gases and in the elimination of toxic waste [17].

The Himalayan pink is a particular type of food salt extracted from mines in the Himalayas. The pink color is due to the impurities present in it, especially iron oxide. Himalayan pink salt is perfect for use in the kitchen because it enhances the taste of food without covering its original flavor. This salt is

particularly suitable for seasoning fish, meat, raw and cooked vegetables, vinaigrettes and various types of sauces.

The Hawaii red is made by sea salt and purified red clay of volcanic origin. For its intense and lively flavor, it is perfect for grilled and roasted meats and it is also important for the supply of mineral salts to the human body [18].

The iodized one is a common food salt obtained from sea water or from the mines of rock salt. Then, artificially added iodine in the form of iodide or potassium iodate is added. The amounts added are chosen and standardized based on the nutritional status of the population; in Italy, for example, each kilogram of iodized salt contains 30 mg of iodine [19]. Iodized salt is the solution proposed by the world health organization to combat iodine deficiency disorders, which can cause very serious health problems.

The hyposodic iodized one is a dietary salt with low sodium (NaCl) and high potassium (KCl) content. It can be used as a substitute of common food salt in diets that require the consumption of small amounts of sodium (for example in cases of high blood pressure). Due to its iodine content, it is also indicated in diets that require an integration of this element.

The Maldon smoked sea salt is made by flakes gently smoked over oak to add a rich and sophisticated flavor to any dish. This particular type of salt is not used in cooking as much and it is used as a little touch to add when the dish is ready. It is recommended on white meats like chicken, turkey, rabbit and pork.

The Breton sea salt is obtained in the salt pans of Brittany from the natural evaporation of sea water. This sea salt is harvested in the traditional way and does not undergo further processing. It has a delicate and light taste and it is suitable for flavoring fish, salads and raw vegetables.

The Persia blue one comes from the ancient underground deposits of Iran. It is a rock salt with a pleasantly spicy taste. It is ideal for flavoring white meats and cooked vegetables.

All investigated samples are reported in Table 1, together with their ID code and country of origin.

Table 1. Investigated samples name, together their ID code and country of origin.

No	Sample ID	Sample Name	Country of Origin
1	S1	Cyprus black	Cyprus island
2	S2	Himalayan pink	Himalayas
3	S3	Hawaii red	Hawaii islands
4	S4	iodized	Italy
5	S5	Hyposodic iodized	Italy
6	S6	Maldon smoked sea	England
7	S7	Common sea	Italy
8	S8	Breton sea	England
9	S9	Persia blue	Iran

2.2. Gamma Spectrometry Analysis and Evaluation of Radiological Hazard Effects

In the laboratory, for the gamma spectrometry analysis, samples were crushed, sieved at 1 mm (for homogeneity) and kept moisture-free in an oven in order to reach a constant weight. They were packed in a 20 mL polyethylene plastic vial to reach geometric homogeneity around the detector; then, the respective net weights were measured and recorded.

Subsequently they were counted for 70,000 s and spectra were analyzed in order to obtain the activity concentration of ^{137}Cs and ^{40}K through the evaluation of their γ-lines at 661.66 keV and 1460.81 keV, respectively.

The experimental setup was composed by a positive biased Ortec HPGe detector (GEM) (FWHM of 1.85 keV; peak to Compton ratio of 64:1; relative efficiency of 40% at the 1.33 MeV ^{60}Co γ-line) placed inside lead wells to shield the background radiation environment. Efficiency and energy calibrations were performed using a multipeak Marinelli geometry gamma source (AK-5901) of 1 L capacity, covering the energy range 59.54 keV–1836 keV and customized to reproduce the exact geometries of

samples in a water-equivalent epoxy resin matrix. The ANGLE 4 code was employed for the efficiency transfer factors calculations to the 20 mL vial sample holder geometry [20]. The Gamma Vision (Ortec) software was used for data acquisition and analysis [21].

A photo of the gamma spectrometry experimental setup is reported in Figure 2a.

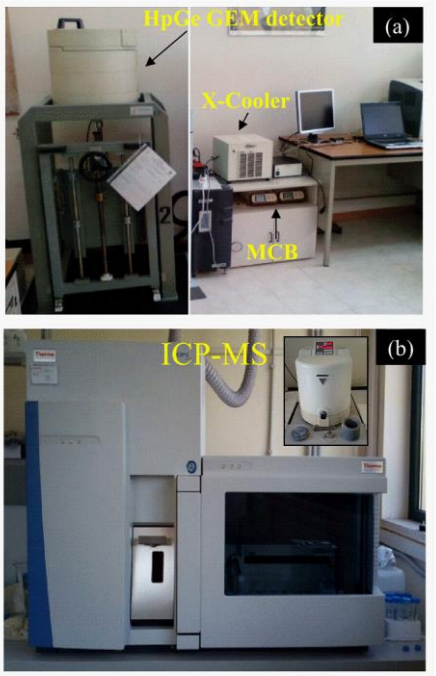

Figure 2. Photo of the gamma spectrometry (**a**) and of the ICP-MS (**b**) experimental setups.

The activity concentration of investigated radioisotopes was calculated using the following formula [22]:

$$C = \frac{N_E}{\varepsilon_E t \gamma_d M} \quad (1)$$

where N_E indicates the net area of the radioisotope photopeak, ε_E and γ_d are the efficiency and yield of the photopeak, M is the mass sample (g) and t is the live time (s).

The measurement result uncertainty, coverage factor k = 2, was calculated taking into account the following components: uncertainty of the counting estimation, of the calibration source, of the efficiency calibration, of the background subtraction and of the γ-branching ratio [21].

The evaluation of radiological hazard effects was made in terms of the annual effective dose for food salt ingestion. It was calculated by the following [23]:

$$D_{ing}\left(\frac{\text{Sv}}{\text{y}}\right) = h_{ing,K-40} \times J_{ing,\ K-40} \quad (2)$$

where $h_{ing,K-40}$ is the coefficient of effective dose for the insertion unit, for the ingestion of ^{40}K (Sv/Bq) and $J_{ing,K-40}$ is the intake of ^{40}K (Bq/year) [23]. The latter value is obtained by multiplying the annual salt food consumption for the activity concentration of the investigated radionuclide experimentally measured.

2.3. ICP-MS Analysis

The concentration of Cu, As, Cd, Hg and Pb was obtained through ICP-MS analysis using a Thermo Scientific iCAP Qc ICP-MS (Figure 2b). Approximately 0.1 g of the sample was dissolved, together with 0.5 mL of ultrapure (67–69%) HNO_3 in 50 mL of bidistilled water.

The sample introduction system consisted of a Peltier cooled (3 °C) baffled cyclonic spray chamber, PFA nebulizer and quartz torch with a 2.5 mm i.d. removable quartz injector. The instrument was

operated in a single collision cell mode, with kinetic energy discrimination (KED), using pure He as the collision gas. All samples were presented for analysis using a Cetac ASX-520. The iCAP Qc ICP-MS was operated in a single KED mode using the following parameters: 1550 W forward power, 0.98 L/min nebuliser gas, 0.8 L/min auxiliary gas, 14.0 L/min cool gas flow, 4.5 mL/min collision cell gas He, 45 s each for sample uptake/wash time, optimized dwell times per analyte (0.01 s, except 0.05 s for As) and one point per peak and three repeats per sample [24].

3. Results and Discussion

3.1. Radioactivity Analysis

The specific activity of ^{40}K in the investigated samples, as calculated by Equation (1), is reported in Table 2. It was found to be: (1.76 ± 0.16) Bq/g for Cyprus black, (0.89 ± 0.07) Bq/g for Himalayan pink, (0.79 ± 0.07) Bq/g for Hawaii red, (1.06 ± 0.09) Bq/g for iodized, (12.1 ± 1.1) Bq/g for hyposodic iodized, (1.47 ± 0.12) Bq/g for Maldon smoked sea, (0.93 ± 0.08) Bq/g for common sea, (1.51 ± 0.13) Bq/g for Breton sea and (4.55 ± 0.32) Bq/g for Persia blue, respectively.

Table 2. The specific activity of ^{40}K and ^{137}Cs in the nine investigated food salts and the annual effective dose due to their ingestion.

	Activity concentration (Bq/g)		D_{ing} (mSv/y)
Sample ID	**^{40}K**	**^{137}Cs**	
S1	1.76 ± 0.16	<1.6	0.040
S2	0.89 ± 0.07	<1.9	0.020
S3	0.79 ± 0.07	<1.6	0.018
S4	1.06 ± 0.09	<1.7	0.024
S5	12.1 ± 1.1	<1.8	0.27
S6	1.47 ± 0.12	<1.5	0.033
S7	0.93 ± 0.08	<1.7	0.021
S8	1.51 ± 0.13	<1.9	0.034
S9	4.55 ± 0.32	<1.8	0.10

In order to well understand these experimental results, we must think in terms of the potassium content, as an ion or compound, in the analyzed samples. The highest value of ^{40}K activity concentration refers to hyposodic iodized salt, which is characterized by a low sodium and a high potassium chloride, potassium citrate and potassium iodate total content (31.3%).

For Cyprus black, Himalayan pink, Hawaii red, iodized, Maldon smoked sea, common sea and Breton sea food salt samples, ^{40}K experimental specific activity is of about 1–2 Bq/g. This is due to their similar chemical composition in terms of potassium content (K is present in traces). The Persia blue sample has instead a ^{40}K activity concentration higher than the previous ones, because in this food salt there is a significant presence of potassium chloride (about 12%).

Regarding ^{137}Cs, its activity concentration in all analyzed samples is lower than the minimum detectable activity value, as reported in Table 1, thus excluding an anthropogenic radioactive contamination of the investigated samples.

3.2. The Annual Effective Dose for Food Ingestion

To evaluate the human health risk, the estimation of the annual effective dose due to the ingestion of investigated samples, as calculated by Equation (2), for the age category higher than 17 years, was performed and experimental results are reported in Table 2. They take into account the human body daily need of about 10 g of salt, and the precautionary hypothesis that this need was satisfied from a single type of salt and the effective ^{40}K half-life in a human being strictly correlated with the potassium metabolism.

Assuming a total yearly consumption of about 3.65 kg, the annual effective dose due to the food salt ingestion was found to be 0.040 mSv/year for Cyprus black, 0.020 mSv/year for Himalayan pink, 0.018 mSv/year for Hawaii red, 0.024 mSv/year for iodized, 0.27 mSv/year for hyposodic iodized, 0.033 mSv/year for Maldon smoked sea, 0.021 mSv/year for common sea, 0.034 mSv/year for Breton sea and 0.10 mSv/year Persia blue, respectively.

These values represent about the 1.6%, 0.8%, 0.7%, 1%, 11%, 1.4%, 0.9%, 1.4% and 4.3%, respectively, of the total natural radioactivity value (external + internal) for humans, expressed in terms of effective dose (2.4 mSv/year) [25].

The obtained annual effective dose is under allowable levels (1 mSv/year) [26] for all investigated samples and therefore, there is no risk of ionizing radiation effects on humans.

The evaluation of human health risk due to the ingestion of investigated samples for age categories lower than 17 years was not performed, because for the age category lower than 24 months, the ingestion of food salt is not recommended, and for the age category between 24 months and 17 years, the daily intake depends on the child needs, but regardless, it is lower than 10 g [27].

3.3. Metals Analysis

Copper, arsenic, cadmium, mercury and lead are heavy metals that bioaccumulate in the human body. They can only be partially excreted with the metabolism [28]. In particular, copper is an essential element in the right doses. Its excess can lead to excessive stimulation of the nervous system and to problems related to the organs where it tends to accumulate more: the liver, brain and reproductive organs [29]. Arsenic is a certain human carcinogen. Its accumulation beyond reference thresholds leads to bladder, lung and skin tumors [30]. Cadmium is a possible human carcinogen. The accumulation of it in excessive amount leads to considerable damage to the kidneys, liver, bones and reproductive system [31]. Mercury is another possible human carcinogen. Factors that determine the type and severity of the produced consequences are: its form (the organic form is more toxic than the inorganic one), the quantity, the duration and the way of exposure and the age of the exposed individual [32]. Regarding lead, the most affected apparatus for the accumulation of it in an excessive amount is the central nervous system, but the renal, gastrointestinal, hematopoietic and reproductive systems are also involved [33].

Table 3 reports metals concentration (mg/kg) in investigated food salt samples, as obtained through ICP-MS analysis. As reported, for Cu, As, Cd, Hg and Pb, the experimental values are lower than the contamination thresholds set by [34] (2.0 mg/kg, 0.5 mg/kg, 0.5 mg/kg, 0.1 mg/kg and 2.0 mg/kg, respectively), for all analyzed samples.

Table 3. Metals concentration (mg/kg) in investigated food salt samples.

Sample ID	Metals Concentration (mg/kg)				
	Cu	**As**	**Cd**	**Hg**	**Pb**
S1	1.7	0.03	0.01	0.03	0.5
S2	1.2	0.02	0.01	0.05	0.2
S3	1.8	0.04	0.005	0.02	0.2
S4	0.8	0.02	0.01	0.04	0.2
S5	0.6	0.01	0.01	0.02	0.5
S6	0.7	0.03	0.02	0.02	0.5
S7	1.3	0.01	0.02	0.09	0.3
S8	0.9	0.1	0.01	0.02	0.6
S9	1.5	0.01	0.01	0.02	0.3
Contamination threshold	*2.0*	*0.5*	*0.5*	*0.1*	*2.0*

Therefore, they can not be considered pollutants. As a consequence, they do not cause objectionable effects and do not constitute a risk to human health.

4. Conclusions

In this study, the radiation levels and metals contamination in nine different samples of food salt available at the Italy markets were assessed. This is extremely important because salt is a source of many elements and minerals essential and necessary for various biological processes in the human body.

The activity concentration of the main natural radionuclide present in food salt, ^{40}K, was measured using HpGe gamma spectrometry with the aim to estimate the health risk, for the age category higher than 17 years, by the effective dose due to their ingestion. The coefficient of the effective dose for ingestion was that reported by the Italian Legislative Decree 230/95, and then in successful modifications. The estimated annual effective dose was found to be in the range (0.018–0.27) mSv/year, about 0.7–11% of the total natural radioactivity value for humans (2.4 mSv/year) and under allowable levels (1 mSv/year), thus excluding the risk of ionizing radiation effects on humans.

The anthropogenic radioactivity was evaluated through the ^{137}Cs specific activity measurement. It was lower than the minimum detectable activity value, thus excluding its presence as a pollutant.

Regarding the metals concentration, it was lower than the contamination threshold value for Cu, As, Cd, Hg and Pb, thus excluding their presence as contaminants.

Results of this study can be useful within a broader look at the issue of food contamination, and there is a need for further research in the future for the Regulatory Authorities in the country to control public exposure to radioisotopes and metals due to the intake of salt.

Author Contributions: F.C. collected data, designed the study and drafted the manuscript; S.M. performed gamma-spectrometry analysis; M.D. and A.B. contributed to perform gamma-spectrometry analysis; M.M. performed ICP-MS analysis; L.S. contributed to perform ICP-MS analysis; G.B. supervised the study.

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