# Alternative ways of enriching the human diet with iodine

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# Abstract

Based on the literature, this study presents the main problems associated with introducing iodine into food. The problem of iodization of table salt and its use in the production of processed food is analysed. This method of enriching the human diet with iodine is linked to the over-consumption of salt, and thus to the risk of hypertension and kidney disease. Because of the need to reduce consumption of table salt, alternative methods of supplying the body with iodine are gaining in popularity. Described are attempts to fortify foods of plant origin with iodine. One alternative method of enriching food with iodine is the cultivation of plants fertilized with iodine. The results are presented of experiments aimed at the cultivation of vegetables enriched with iodine and analysed methods of iodine fertilization: foliar application, soil application and hydroponic cultures. Also discussed are the problem of the effects of iodine, not only on yield, but also on selected physiological processes taking place in plants which are responsible for the biological quality of the crop.

# Key words

micronutrients, prevention, fertilizers, iodine biofortification, plants

# THE IMPORTANCE OF IODINE

Iodine is an essential micronutrient for the normal growth and development of both humans and animals (mainly mammals). It is a basic component of the thyroid hormones thyroxine (T4) and triiodothyronine (T3), which participate in metabolism regulation and are required for skeletal and central nervous system development in the foetus and infant [1]. Iodine deficiency is associated with impaired production of thyroid hormones [2]. Insufficient iodine in the organism can cause hypothyroidism, whose symptoms include increased body weight, overall weakness, and an enlarged thyroid gland. Iodine deficiency is of particular significance for pregnant women, infants, and small children. Prolonged iodine deficiency in the development period leads to irreversible brain damage and mental retardation [3].

Iodine is delivered to the organism mainly in food and drinking water. The amount present in foods depends mainly on its quantity in the soil and water. Iodine is also taken in by the respiratory system and through the skin. The highest iodine levels are found in sea water and in products originating in the sea, i.e. sea algae and seafood. Other good sources of iodine are dairy products, mainly owing to iodized animal feed [4], and eggs [5]. Iodine is eliminated from the organism by the kidneys and iodine deficiency can be diagnosed by determining its content in the urine [6].

In scientific publications, the conviction predominates that iodine fertilization is not essential for the development of plants. Optimum iodine levels vary for different crops, from 0.025–0.2 mg iodine kg<sup>-1</sup> sand [7]. In a few crops, however, it has been found to be as high as 0.6 mg iodine kg<sup>-1</sup> sand. Land

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plants can take up iodine from the soil through their roots or directly from the atmosphere, e.g. through the leaves. The physiology of iodine in plants is poorly understood. Plants can absorb iodide anion from the soil [8]; they are better able to tolerate high levels of iodine in the form of iodate ( $IO_3^{-1}$ ) than as iodide ( $I^-$ ) in the root environment [9]. In root cells,  $I^-$  follows the chloride ( $CI^-$ ) transport pathway with H<sup>+</sup>/anion symporters across membranes into the xylem. The molecular identities of transporters catalysing these fluxes are not firmly established. Putative H<sup>+</sup>/halide transporters belong to the chloride channel (CLC) transporter family and a subset of the ATP binding cassette (ABC) protein superfamily, while Na:K/ Cl symporters belong to the cation chloride co-transporter (CCC) gene family [10].

A study on the development of genetically- engineered Arabidopsis plants harbouring the human sodium-iodide symporter (hNIS) gene showed that stable expression of the transgenic lines produced plants capable of absorbing more iodine. This improved uptake was even more significant when hNIS plants were grown at 30 °C, while nitrate fertilization had an abating effect. The efficacy of iodine uptake in hNIS plants was limited by the activity of an endogenous halide methyltransferase enzyme which methylates free iodide to produce volatile methyl iodide [11]. Once inside the plant, a xylem flux of iodine seems to be largely predominant, but generally the absorbed iodine is not uniformly distributed among plant tissues, ranking as follows: root > leaf > stem [12]. Plants rich in iodine include sea algae, lichens, cardamine, garlic, spinach, and herbs. Iodine present in the plant is known to be well assimilated by the human body. Novel studies have demonstrated that some vegetables can store iodine in their edible tissues, making them good candidates for iodine biofortification programmes. The concentrations of iodate in soil have been shown to have a significant effect on the biomass of edible parts of pakchoi and spinach, but no significant effect on that of carrots, water spinach, celery,

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or onion [13].

### **IODINE SUPPLEMENTATION OF FOOD PRODUCTS**

Nearly 38% of the human population live in areas deficient in iodine, and thus at the risk of thyroid disease [14]. For this reason, many countries practice iodine prophylaxis involving the introduction of iodine into the diet via iodized salt. The effectiveness of this method is mainly due to excess consumption of salt, which has led to a global increase in cardiovascular disease, including hypertension, and certain cancers. In accordance with recommendations by the World Health Organization (WHO), attempts are being made to reduce daily consumption of table salt by one half. This could lead to a substantial reduction in the iodine supply in the diet [15, 16]. In the 1990s, iodized salt programmes were introduced in many countries. Progress has been made over the past two decades in the global effort to eliminate iodine deficiency. In 1993, 110 countries were iodine-deficient, and between 2003-2011, the number of iodine-deficient countries decreased from 54-32, and the number of countries with adequate iodine intake increased from 67 to 105. Currently, 71% of the global population has access to iodized salt [17]. Nevertheless, over 1.9 billion people worldwide are affected by an insufficient supply of iodine, including 285 million school-age children. In 2007, insufficient iodine consumption by children aged 6 –12 (< 100  $\mu$ g  $\cdot$  dm<sup>-3</sup> in morning urine) was noted in 52.45% of cases in Europe and 10% in the United States [18].

Since 1994, the WHO and the United Nations Children's Fund (UNICEF) have recommended universal salt iodization (USI) as a safe, cost-effective, and sustainable strategy to ensure sufficient intake of iodine by all individuals. In industrialized countries, people are consuming the majority of their salt through processed foods, in which iodized salt is generally not used, rather than through iodized table salt. Iodized salt appears to be primarily used in food products only when required by legislation, and companies do not appear to use iodized salt in product categories that do not require it [19]. West and Merx [20] cited reports that salt iodized with potassium iodide and used for cheese manufacturing, particularly Emmentaler and Gruyere, had no discernible impact on flavour or quality.

The Programme for Elimination of Iodine Deficiency in Poland, financed by the Ministry of Health, involves obligatory iodization of table salt (20–40 mg KI  $\cdot$  kg<sup>-1</sup>) and infant formula ( $10 \,\mu\text{g} \cdot 100 \,\text{ml}^{-1}$  milk), as well as supplements of 150–200 µg per day for pregnant and breast-feeding women. Iodine-rich foods (saltwater fish, eggs, milk, vegetables and fruit) are recommended in the daily diet. Also planned are measures aimed at enriching cows' milk with iodine (to 100–150  $\mu$ g · dm<sup>-3</sup>) by iodizing fodder [15]. The measures undertaken are achieving the desired effects. Populationbased studies confirm the elimination of endemic goitre in schoolchildren and a marked reduction in the frequency of thyroid cancer in women over 40, goitre in pregnant women (from 80% – 19%), and transient hypothyroidism in infants (from 2.0% – 0.16%) [21]. Recent research has brought to light another aspect of iodine prophylaxis, i.e. the concurrence of symptoms of iodine deficiency with a high incidence of stomach cancer. Epidemiological data from many parts of the world indicate that a high incidence of this type of cancer coincides with endemic goitre, while the incidence of stomach cancer decreases with the decreasing prevalence of endemic goitre due to iodine prophylaxis. Epidemiological data in Poland seem to confirm this. In 1996, obligatory iodization of table salt was introduced in Poland, at a level of  $2.3 \pm 0.77$  mg of iodine per 100 g table salt, equivalent to  $30 \pm 10$  mg of potassium iodide or  $39.13 \pm 13$  mg potassium iodate) per kg of table salt [18].

The main source of iodine in the prophylaxis system is iodized table salt, but research shows that the values for iodine content in salt are at the lower end of the norm [22]. On average, four-fifths of samples tested were found to fulfil the norm for iodine. The percentage of samples with satisfactory iodine content varied among the Polish provinces. In 2000, the lowest percentage of salt samples with satisfactory iodine content was noted in the West Pomeranian Province – 40%; in 2001, the lowest percentage was noted in the Lower Silesian Province - 69%; and in 2002, the lowest percentage was in the Lublin Province – 60%. In 2001–2002, there were some provinces in which all salt samples tested met the requirements for iodine content (Lubusz and Warmian-Masurian Provinces in 2001, and the Lesser Poland and Greater Poland Provinces in 2002). It was also determined that 20% of the iodized salt produced still exhibited low levels of potassium iodide [22].

Medical research indicates that salt is the main cause of hypertension and its secondary effects, including arterial sclerosis, infarctions and arterial embolisms, but it is also a source of iodine preventing thyroid disease (goitre, thyroid cancer) [23].

Consumption of sodium chloride in Poland exceeds the recommended daily requirement (5.0–6.0 g) by nearly two-fold [15]. According to recommendations by the WHO, the positive effects of iodine prophylaxis should be maintained while salt consumption should be reduced. A programme for reducing consumption of table salt inevitably entails a significant decrease in the iodine supply in the population, and thus may reduce the effectiveness of iodine prophylaxis. This means that additional sources of iodine must be introduced into the food market in the form of iodine-rich foods.

Current dietary recommendations indicate the need to increase consumption of iodine-rich foods (saltwater fish, eggs, milk, vegetables and fruit). Consumption of milk and dairy products may meet the requirement not only for calcium, but also to a significant degree for iodine. Due to high consumption levels, dairy products are a good source of this nutrient. A study conducted on a group of pregnant women showed that their average consumption of dairy products provided nearly one-sixth of the recommended iodine consumption for this group. Iodine-rich hard cheese is the largest contributor of iodine in the diet of pregnant women, followed by curd cheeses, yoghurt and milk [24]. Research on women in the Greater Poland Province indicates that their food consumption met 93% of the daily iodine requirement [24]. In Poland, iodine in milk does not exceed 0.10–0.15 mg  $\cdot$  dm<sup>-3</sup> [25]. Saltwater fish and seafood have a high natural iodine content, but their contribution to the overall dietary iodine intake is modest unless consumed every day. Fish consumption in Poland is still unsatisfactory. Children and teenagers are at risk of iodine deficiency, while consumption of fish and fish products among young people aged 16-19 meets only 10% of the requirement for iodine [26].

Drinking water drawn from certain aquifers or water disinfected with iodine can also be rich in iodine. Tap water in a few locations in Europe contains up to 139 µg dm<sup>-3</sup> of iodine, mostly bound to humic substances, probably leaching from marine sediments in the aquifers. Even higher iodine contents have been found in villages along Bohai Bay, China [27]. In Poland, water rich in chlorine and sodium ions, with varied content of minerals such as iodine, bromine, magnesium and calcium, are found in the Carpathian Mountains, and in the Polish Lowland water rich in chlorine and sodium ions, with smaller amounts of other minerals (iodine, bromine), predominates. The bottled water 'Jodica' from the town of Nieszawa, on the River Vistula in Kujawsko-Pomorskie Province, is one of the few examples of iodine-enriched water available in Poland. One dm<sup>3</sup> of 'Jodica' contains 150 µg I, which is the daily requirement for adults. Water of the brands 'Zuber', 'Franciszek', 'Jozef' and 'Henryk' contain a substantial amount of iodine and can be used in the diet of those at risk of iodine deficiency [28]. Attempts have been made to create a new and popular source of iodized food for children and teenagers. Iodide was used in the production of salty snacks, but the results obtained were not satisfactory due to iodine losses [29].

The use of sugar as a vehicle for iodine supplementation was explored in a study on iodine deficiency in the Sudan. Iodinated sugar was produced by adding iodine salts to a sugar solution prior to crystallization in an evapocrystallizer or spraying them on the conveyor of cured sugar before it entered the dryers. The results indicate that fortification of sugar with iodine may serve as a new alternative approach in attempts to eradicate disorders related to iodine deficiency in endemic areas [30].

Iodine-rich foods are an essential element of the diet, but during food processing iodine content is reduced. Iodized salt (KI, KIO<sub>2</sub>) was added to pork and the iodine loss during roasting, refrigeration and freezing of meat dishes was determined. Just 15 minutes of roasting caused a 5% loss of iodine in meatballs, and 30 minutes of heat treatment caused iodine losses that were three times greater [31]. Iodine loss during cooking can be prevented by adding iodine-binding compounds to the food. Heat treatment of meatballs caused a 18-30% reduction in total inorganic iodine. Adding soy hydrolysate decreased inorganic and total iodine losses by about 7% [32]. Iodine in the form of potassium iodide or potassium iodate was added to pickling mixes used in the production of meat products, but the iodine losses in these products following heat treatment and refrigeration were high. In a similar experiment, iodine was added to wheat and rye bread during production [4]. This research indicates that it would be beneficial to eat iodine-rich products raw, in order to limit iodine losses.

### **PROBLEMS WITH IODINE BIOFORTIFICATION**

One of the problems encountered by researchers attempting to fortify plants with iodine involves the method of applying it, and its redistribution in the plant. The literature provides many examples of iodine fertilization. Studies on iodine biofortification of cabbage, lettuce, tomato and spinach, alfalfa, pakchoi, celery, pepper and radish [8, 33, 34, 35] have generally focused on optimizing plant enrichment with this element. One strategy is soil fertilization with iodine [10]. Ledwożyw-Smoleń et al. [36] noted a significant decrease in the weight of lettuce heads in plants growing in nutrient solution containing 10 mg I  $\cdot$  dm<sup>-3</sup> in the form of iodate, but the plants did not exhibit symptoms of toxic effects of iodine. Application of KI as a source of iodine had a stronger inhibitory effect on the growth of the plants. As iodine content in the form of iodide increased in the nutrient solution (0, 1.0 and 10 mg I  $\cdot$  dm<sup>-3</sup>), the weight of the lettuce heads decreased, and plants growing in the nutrient solution with the highest iodine content were damaged by it to a considerable degree [36].

The diverse reactions taking place in the soil-plant system, particularly high sorption of iodine by organic matter and microorganisms, may reduce the effectiveness of iodine biofortification of plants. Plant uptake of iodine is dependent on its availability, which is governed by the adsorptiondesorption characteristics of soils [37]. The effectiveness of soil fertilization with iodine is limited by its high sorption in the soil [38]. Humic acids contained in the soil cause conversion of I<sup>-</sup> into I<sub>2</sub>, resulting in volatilization of iodine from the soil into the atmosphere [39]. Methylation of iodine also occurs, in which it is volatilized from plants into the atmosphere in the form of CH<sub>2</sub>I. High sorption of iodine in the soil with organic matter (thiol groups and polyphenols) and with hydrated iron and aluminium oxides substantially reduce the availability of iodine to plants [40]. Iodine desorption from the soil is low, taking place mainly when the reduction potential  $(E_{\mu})$  of the soil is reduced to negative  $E_{\mu}$  values. This situation occurs in agricultural soil only in the case of excessive, prolonged moisture, when anaerobic conditions arise [38]. Studies conducted by Muramatsu et al. [38] revealed that after 60-day soil incubation with  $5g \cdot dm^{-3}$ of sucrose (laboratory test),  $E_h$  decreased in the soil from positive (app. +580 mV) to negative values (app. -200 mV).

A solution to the problem of high iodine sorption by the soil is hydroponics. In a hydroponic culture of lettuce, biofortification with iodine during foliar fertilization in the form of IO<sub>2</sub> was found to be more effective than adding it to the nutrient solution [36]. Plant roots have little capability to take up iodate ions. The root systems of most plant species take up iodine more easily in the from of I<sup>-</sup> than IO<sub>2</sub>. Smoleń et al. [41] postulate that in a hydroponic tomato culture both in the vegetative and the generative growth stages - the iodide form (I) is better taken up by plants than the iodate form (IO<sub>3</sub><sup>-</sup>). Ledwożyw-Smoleń et al. [36] investigated how different means of applying KIO<sub>3</sub> affect iodine accumulation and the nutritional quality of lettuce grown hydroponically. They determined that foliar application with iodate was a more effective means of enriching lettuce with iodine than adding it to the nutrient solution, but the lettuce accumulated iodine in both methods. The results of a study by Bai et al. [33] with hydroponic cultures of two varieties of lettuce – 'Red-fire' and 'Rakuten' - also showed that plants treated with KI contained more iodine than those treated with KIO<sub>3</sub>. Smoleń et al. [14] investigated the efficiency of iodine uptake and nutrient content in the leaves and fruit of tomato plants. Iodine content in the fruit of plants fertilized with KI was nearly four times higher than in the control, and over three times higher than in the case of fertilization with KIO<sub>2</sub>. Ledwożyw-Smoleń et al. [36] found that foliar application of KIO<sub>3</sub> was more effective than adding it to the nutrient solution. A substantial increase in iodine content was obtained in lettuce plants. However, this method of iodine application (particularly at higher concentrations) caused a significant decrease in soluble sugars, ascorbic acid and free amino acids in the lettuce. Foliar application of KIO<sub>3</sub> was more effective than adding it to the nutrient solution, but reduced the biological quality of the lettuce. This calls into question the suitability of such conditions for enriching plants with iodine.

A problem often discussed in the literature is the effect of different forms of iodine on the mineral composition of plants. Ledwożyw-Smoleń et al. [36] postulate that IO<sub>2</sub><sup>-</sup> can interfere to some degree with nitrogen metabolism pathways. Iodine is better taken up from the soil in the presence of other nutrients. In a pot culture of carrot on heavy soil, varied combinations of iodine (in the form of I and IO, at 1 mg  $I \cdot dm^{-3}$  soil) and nitrogen (in the form of NH<sup>+</sup> and NO<sub>2</sub>) were applied to the soil [42]. Fertilization with iodine in the form of KIO<sub>2</sub> (in comparison with KI) increased the total nitrogen content in the storage roots of the carrots, but did not affect its content in the leaves. Fertilization with iodine in the form of KI and nitrogen in the form of NO, was found to substantially increase iodine content in the carrot leaves (158.5 mg  $\cdot$  kg<sup>-1</sup> d.w. in comparison with the control plants - 120 mg  $\cdot$  kg<sup>-1</sup> d.w.), while none of the variants of iodine fertilization affected its accumulation in the roots (results in the samples were similar to the control, 0.86 mg  $\cdot$  kg<sup>-1</sup> d.w.). This method of biofortification was also used in field cultivation of carrot [43]. Varied combinations of iodine in the form of I<sup> $\circ$ </sup> or IO<sub>3</sub><sup>-</sup> and nitrogen in the form of NO<sub>3</sub><sup>-</sup> or NH<sub>4</sub><sup>+</sup> were applied to the soil. Iodine was applied before sowing, and nitrogen before sowing and as top dressing. Iodine in the form of KI produced better effects, particularly when applied together with ammonium sulfate. Iodine concentration in the carrot roots was 5 mg  $\cdot$  kg<sup>-1</sup>, compared to 2 mg  $\cdot$  kg<sup>-1</sup> d.w. in the control sample.

Application of iodine in the form of IO<sub>2</sub><sup>-</sup> increased the efficiency of mineral nitrogen utilization by the plants, but lower iodine content was noted in the carrot roots in comparison with fertilization with I<sup>-</sup>. Neither form of iodine fertilizer affected the content in the carrot roots of Mg, S, Cu, Zn, Mo, Al and Pb, but they both increased P, K and Ca content and decreased Fe content. Spinach Spinacia oleracea L. of the variety 'Olbrzym zimowy' was grown in a pot experiment on mineral soil. Different concentrations of iodine were applied to the soil (1 mg or 2 mg  $\cdot$  dm<sup>-3</sup> soil in the form of KI), as well as iodine and nutrient solution enriched with sucrose [44]. Iodine synergistically improved the uptake of Mg, Na, Ca and Fe (for Fe only in the case of higher iodine concentrations) but antagonistically affected Cr uptake by the spinach plants. Following application of iodine at 2 mg I  $\cdot$  dm<sup>-3</sup> soil, higher accumulation of Na, Fe, Zn and Al was observed along with reduced concentration of P, S, Cu and Ba in the spinach plants in comparison with the control. Simultaneous application of iodine and sucrose significantly increased the accumulation of K, S and Mo in the spinach plants in comparison with the control or plants fertilized only with iodine, while decreasing the content of Mg, Fe, Ba, Co and La.

In a hydroponic experiment, Smoleń et al. [14] investigated the efficiency of iodine uptake and nutrient content in the leaves and fruit of tomato plants. Application of potassium iodide to the nutrient solution increased Na, B and Mn content, while application of potassium iodate caused it to decrease with respect to the control. KI added to the nutrient solution decreased molybdenum content in the tomato fruit in comparison with the control, while KIO<sub>3</sub> increased it. The highest content of Cu and Fe in the fruit was noted in the control. No statistically significant influence was noted for either form of iodine on the Zn content in the fruit. Adding potassium iodate to the nutrient solution decreased the content of P, K, Ca, S, Na, B, Cu and Fe in the tomato fruit with respect to the control and KI fertilization.

One aspect of research on the influence of iodine on plants is the biological quality of the fertilized plants. Application of varied iodine and nitrogen fertilizer combinations slightly reduced the dry weight of carrot roots, but increased the content of carotenoids and ascorbic acid, while it did not cause significant changes in the content of soluble sugars [42].

Tomatoes (Lycopersicon esculentum Mill.) of the variety 'Rambozo F<sub>1</sub>' were grown in a greenhouse in the Nutrient Film Technique (NFT) hydroponic system with recirculation of the nutrient solution. Iodine was applied at a concentration of 1 mg I · dm<sup>-3</sup> in the form of KI or KIO<sub>2</sub>. Iodine fertilization was applied throughout the growth of the plants, beginning at the stage when flowers appeared in the first cluster. A significant decrease in lycopene and increase in ascorbic acid were observed in the tomato fruit from the third cluster in comparison to the control. Iodine application in the form of KIO, had a positive effect on the biological quality of the tomato fruit due to a significant increase in the total acidity of the extract and total content of soluble sugars (including glucose and fructose). The factors had no significant influence on the content of dry matter, carotene, phenolic compounds, phenylpropanoids, flavonols, or anthocyanins, or on total fruit yield [45]. The literature data presented in the current study concerning the importance of iodine in plant nutrition substantiate the need for research on its effect on the growth and development of plants, and the identification of species most suitable for biofortification with this nutrient.

### CONCLUSIONS

Iodine is an essential microelement, which plays a irreplaceable role in metabolism process in human and animals organisms. The diet of many populations around the world contains an insufficient amount of these elements. Two main sources of iodine supplementation are seafood and iodized salt. Iodine biofortification of vegetables could become an alternative (to iodized salt) method of introducing this element into the human diet. A review of the literature provided a great deal of information about biofortification of plants with iodine. However, the subject of most of this research is adult plants. Iodine biofortification of plants through soil fertilization has a relatively low effect, which is caused by strong iodine sorption in the soil. Iodine is not included among essential plant nutrients, but literature data indicate that it can have a beneficial effect on their biological quality. Several methods of iodine plant enrichment have been proposed, but none of these can be considered as optimal and each species requires a careful and specific evaluation. The positive results obtained in trials carried out with some leafy vegetables (e.g., spinach, lettuce), particularly in hydroponic culture, have suggested that they are good candidates for iodine biofortification programmes. There is therefore a need for research on the effect of iodine biofortification on the growth and development of plants, and on selected aspects of their biochemistry and metabolism.

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