András S. Szabó[1]

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Investigation of the strontium content of foods and the biological role of strontium

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1. Summary

From a physiological point of view, strontium is a microelement, having a common transport system with calcium. In plants and soils, there is usually a difference of two orders of magnitude between the concentrations of calcium and strontium. However, in the case of humans and animals, the difference can be as high as three orders of magnitude. The reason for this is the strong ability to discriminate, as a result of which strontium is absorbed from the alimentary canal of warm-blooded creatures in significantly lower amounts. Thus, the Sr:Ca ratio in the human body, as well as its radiostrontium contamination ($^{90}Sr/Ca$) is significantly lower than the values that can be measured in the foods consumed. It is well known that calcium can be partially replaced by strontium, but large amounts of strontium are toxic to the body. At the same time, strontium is used in medicine to treat osteoporosis.

Clarification of the plant, animal and human physiological role of strontium, and proving of its possible essential role requires further investigation. Since the concentration distribution of essential and non-essential microelements shows significant differences within a healthy organism, this knowledge will be of great help in the determination of the biological role of the given microelement by examining the concentration distributions in healthy body tissues, because essential elements are present in the tissues in a narrow concentration range.

2. Introduction

In an earlier paper, published in the Journal of Food Investigations, there is a comprehensive overview of the grouping possibilities of microelements, of the relationship between the essential and non-essential nature and the concentration distribution, and of the analytical measurement techniques that can be applied for the analysis of trace elements **[1]**. In this paper, the physiological role of strontium is discussed. With regard to its concentration in foods, this metal is typically a microelement. It should be noted that several details of its physiological role need further clarification.

Research on microelements has a history of roughly 100 years, and it is closely related to the development of the performance and sensitivity of analytical methods. Nowadays, such analyses are performed almost exclusively using instrumental analytical techniques, by the application of measurement techniques based on chemical or physical methods.

In the 1950s, the great importance of strontium-related microelement research was due to the fact that, among the isotopes causing long-term radioactive contamination of the biosphere, because of the environmental impact of frequent nuclear weapons tests, the two isotopes of strontium, radionuclides ^{89}Sr and ^{90}Sr played a major role. Radiochemical, radiobiological and radioecological research focused on the physiological significance of the microelement as well.

It is known that, of the 92 elements that occur in nature (and, obviously, in the human body), not considering the so-called transuranium elements produced artificially, 75 can be classified as microelements, including strontium, which is the 20th most common element in the Earth's crust. It was named by its discoverers,

[1] Public Benefit Foundation of Food Physics

Crawford and Cruickshank in 1790 (the name of the element refers to the village of Strontian in Scotland **[32]**.)

Chemically, strontium is an alkaline earth metal, found in group 2 of the periodic table. It is in period 5, located in the table below calcium and above barium. Regarding its chemical and physical properties, it is similar to calcium, however, its concentration in surface waters, soils and plants is usually 2 orders of magnitude, and in animal and human tissues 3 orders of magnitude lower than that of calcium **[23]**. In drinking water, apart from extreme values at certain geographic locations, its typical values are below or well below 1.0 mg/liter. According to our flame photometric measurements, the strontium contents of the so-called indicator plants (sorrel, spinach, lettuce) and cereals in Hungary was typically 1 to 3% of the calcium contents. The measurable calcium content was related to the plant type and the strontium content of the soil. During the ICP analysis of the composition of vegetables (peas, tomatoes, radish and peppers) it was found that the calcium content was 100 to 200 times higher than the amount of strontium **[2]**, **[3]**, **[4]**, **[5]**.

Concentration distribution studies, based on the results of which it can be assumed that strontium is an essential microelement for plants have already been presented in previous lectures and papers **[6]**, **[1]**, **[7]**, **[8].**

The study of trace element contents and their distributions in foods can help to clarify the biological role of the microelements in question **[9]**. The objective of this paper is to present the presumed biological role of strontium. The question is whether strontium can be considered an essential microelement or should be classified as a non-essential microelement. It is likely that strontium plays an important biological role not only in plant physiology, but also in animal and human physiology. In all likelihood, this alkaline earth metal is a determining factor in ensuring the hardness of bones and teeth, and it may also have a role in preventing the development of osteoporosis.

3. Radioactive strontium isotopes

It was mentioned above that the study of the physiological role of strontium was important in the 1950s and 1960s because long-term environmental contamination of nuclear weapons tests was mainly due to the strontium isotope with a mass number of 90. This isotope undergoes beta decay, with a physical half-life of 28 years. This means that environmental radioactivity due to ^{90}Sr decreases only very slowly. In addition, since strontium is very similar to calcium, ^{90}Sr is also deposited in bones, where it causes long-term radiation exposure. ^{90}Sr is one of the most significant fission products, playing a major role in the development of the radioactive contamination of the affected area. The generation and decay scheme after the fission of the uranium nucleus, due to neutrons, is as follows:

$$^{235}U \text{ or } ^{236}U \text{ fission} \rightarrow {}^{90}Kr - {}^{90}Rb - {}^{90}Sr - {}^{90}Y - {}^{90}Zr$$

The individual isotopes are transformed to elements of the same mass number, but having increasing atomic numbers, by successive beta decays. The decay chain is concluded by the no longer radioactive zirconium with an atomic number of 40. Due to the relatively short half-life of ^{90}Y, ^{90}Sr and ^{90}Y isotopes are in radioactive equilibrium with each other.

In addition to the ^{90}Sr isotope, the also beta-decaying isotope ^{89}Sr is well-known among polluting radionuclides. Although the ^{89}Sr isotope is also formed in significant amounts following the fission of the uranium nucleus, but because its half-life is significantly shorter than that of the ^{90}Sr nuclide, only 50 days, it does not play a role in long-term contaminations. Due to its relatively rapid decay, its presence and radiation exposure effect have to be taken into consideration in the biological chain in the first few months of the contamination.

In animals and humans, because of its so-called bone seeking property, similar to that of calcium, radiostrontium is primarily found in the high calcium content skeleton. If the contamination level (the contamination effect of nuclear weapons tests or nuclear reactor accidents) is high, then bone cancer can be caused by the radiation exposure due to the long half-life of the ^{90}Sr isotope (or to its so-called effective half-life calculated from the biological and physical half-lives). Due to the long biological half-life, the biggest threat to bones, in terms of radiation exposure, is the ^{90}Sr isotope **[10]**. However, in Hungary, the measurable ^{90}Sr contamination level was not significant in the early 1960s due to nuclear weapons tests and then in 1986 because of the Chernobyl nuclear power plant accident **[11], [3], [12], [13]**.

4. Importance of the discrimination factors

The essence of the discriminating ability of the individual organisms is that the relative concentrations (compared to other elements) of certain elements can differ significantly between the elements of the biological chain process.

In the case of Sr, it is advisable to express the strontium value on a calcium content basis, because calcium and strontium form a common transport system, due to their physical, chemical and biological similarities. Calculation of the calcium/strontium ratios can also be carried out on the basis of radiometric measurements, when not strontium, but radiostrontium concentrations (activities) are compared to the calcium values. It can be stated that calcium is the inactive carrier of strontium and radiostrontium.

The discrimination factor for strontium, in the case of the animal or human body, can be calculated as follows:

$$D_{Sr} = \frac{\frac{Sr}{Ca} \text{ a tejben}}{\frac{Sr}{Ca} \text{ a takarmányban, élelmiszerben}}$$

The discrimination factor of strontium can also be calculated based on the activity of ^{90}Sr:

$$D_{Sr} = \frac{\frac{^{90}Sr}{Ca} \text{ a tejben}}{\frac{^{90}Sr}{Ca} \text{ a takarmányban, élelmiszerben}}$$

Of course, the determination of the discrimination factors, indicating the discriminating abilities of the different organisms, can also be based on the analysis of other body tissues (e.g., bone, muscle, egg). According to our measurements **[14]**, no difference could be detected between the strontium contents of bones coming from the same animal, but from different areas of the body. In the case of sheep, there was no significant difference between the different bones (e.g., femur, metacarpus), based on the analysis of 96 samples each and taking into consideration the standard deviations, the average value was around 0.70 mg/g ash, with a standard deviation of roughly 20%.

5. Materials and methods

In our studies, a large number of sample of biological origin (feed and food crops, diets, cow's milk, muscle, egg, bone) were measured. In the case of milk and feed samples coming from a given dairy farm, parallel sampling was always carried out, i.e., the feed consumed by the dairy animals according to the period was sampled.

Determination of the strontium and calcium content was carried out using a flame photometric method, or an X-ray fluorescence procedure (XRF or REA). Flame photometric measurements were performed on the hydrochloric acid solution of ashes after ashing of the samples at 461 and 554 nm. For X-ray fluorescence measurements, an ^{125}I ion source and a Si(Li) detector were used. This technique is mainly suitable for the determination of elements with atomic numbers larger than ten. In the course of our work, analyses were also performed using the ICP-AES technique, with a Thermo Jarrell Ash 9000 instrument, determining the elemental composition of a 0.1 n HNO_3 solution of samples ashed at 550°C. For the measurement of radioactivity, halogen-filled GM tubes and scintillation detectors (plastic and NaI) were used. During the measurements, beta activity was determined, measuring the total activity and the activity of the so-called metal ion fraction. The latter indicates the contamination of the sample with radioactive strontium isotopes.

6. Results and evaluation

Table 1 shows the average strontium and radiostrontium content of parallel milk and feed samples on a calcium content basis. Calculation of the discrimination factors can be performed on the basis of the relative values, calculated on calcium content. The data in ***Table 1*** show that there is an order of magnitude difference between the specific strontium and radiostrontium concentrations. Levels that can be measured in milk constitute only 10 and 13% of the values in feed, respectively. This ratio proved to be somewhere between 8 and 15% for the individual samples, based on the data of a large number of measurements over the years. This suggests that the absorption of strontium is significantly lower than that of calcium and, therefore, the relative stable strontium and ^{90}Sr contents in milk, and other body tissues, are significantly lower than the calcium content.

Regarding discriminating ability, ***Table 2*** contains similar results. Based on the results of bone analyses, the concentration difference of one order of magnitude can be seen easily also. It is well known that the strontium content of feeds, in this case, feed crops of plant origin, depends on the plant species and soil characteristics. Radiostrontium activity, on the other hand, is related to external contamination effects. Influencing factors include geographic characteristics (e.g., precipitation volume) and the agrotechnical processes applied. The Sr and ^{90}Sr contents of animal tissues are a function of the discriminating ability and the chemical composition of the given feed, as well as the age of the animal. According to our measurements, the radioactivity of calf bones was significantly lower than that of young cow bones. This is related to the fact that, in the diet of the calves, the mother's milk is a major factor in the first 1 or 2 months, and its strontium and radiostrontium contents are orders of magnitude lower than those of feeds of plant origin. The rate of substitution (incorporation, accumulation, elimination) for the Ca-Sr system is quite low, it is a function of the biological half-life, and this is the cause of the significantly different strontium and radiostrontium contents of the tissues of calves, young and adult cows **[15]**, **[16]**.

It should be noted here that there are significant differences between the different animal species regarding radiostrontium contamination, partly because of the different discriminating abilities and partly because of the different nutrition and feeding conditions. Based on our measurements, the following ascending order was obtained regarding the strontium content: pigs, poultry, cattle, sheep, fish. In the case of herbivorous fish, the radiostrontium contamination exceeded significantly the ^{90}Sr concentration that could be measured in the bones of predatory fish. The reason for this is that, in the case of predatory fish, basically there is a double discrimination effect due to the same factors mentioned in the case of calf feeding.

7. Calcification in the skeleton

Most of the mineral content of bones and teeth consists of calcium orthophosphate, characterized by the composition $Ca_3(PO_4)_2$. However, in addition to calcium and in a ratio that depends on the diet, the presence of magnesium, strontium, barium and radium can always be detected among the cations. Magnesium is an essential macroelement. However, barium and radium are probably non-essential microelements **[17]**, **[18]**, **[19]**. For plants, the role of strontium is controversial, but based on concentration distribution studies, its essentiality is probable **[5]**, **[7]**. In the case of higher animals (vertebrates) and humans, the biological role of strontium has not yet been clarified, so it is not clear whether strontium should be classified as an essential or a non-essential microelement.

For essential elements, the physiological need and its optimum can be found at the saturation range, over the deficiency symptoms. The first section of this range means an adequate supply, while the second section means overconsumption. After the optimal range, there is a toxicity threshold and a toxicity range. For non-essential elements, in the absence of a biological need and a mechanism regulating the concentrations of the ions, in addition to possible antagonistic effects, only the toxicity threshold is meaningful. In other words, in the case of non-essential elements, an intake not exceeding the toxicity threshold value is not expected to have a significant biological effect.

Certain compounds of strontium, e.g., the strontium salt of ranelic acid, are used as drugs in the treatment of osteoporosis, so the biological role of the metal cannot be disputed. Administration of strontium ranelate promotes bone formation and increases the calcium content of the bones **[20]**. It was demonstrated that the administration of strontium ranelate ($C_{12}H_{10}N_2O_8Sr$) reduced significantly the occurrence of fractures in the vertebrae and the hip bone in menopausal women suffering from osteoporosis, it reduced bone resorption and increased bone density **[30]**. The latter manifested partly in the actual increase in bone mass, and partly in the increased incorporation of strontium, with a higher atomic number and density than those of calcium. Other strontium compounds, for example, strontium citrate, are also used in dietary supplements **[21]**.

Strontium plays a role in bone metabolism, and has an anabolic effect on the skeleton (osteoblast and osteoclast). It can be assumed that its passive diffusion and Ca carrier roles are decisive in ion transport. Its effects manifest through Ca-sensing receptors. The presence of strontium inhibits resorption in the bone and stimulates bone formation **[22]**.

8. Strontium in the human body

Like calcium, barium and radium, the vast majority of strontium in the human body is found in the bones and teeth. According to literature data **[23], [24]**, the total amount of strontium in the human body is about 350 to 400 mg. The average daily strontium intake is estimated at 1.5 to 2.0 mg. Strontium passes through the mucous membrane of the digestive tract in an ionic form relatively rapidly, and is excreted in urine or even breast milk. However, most of the strontium taken up is excreted in the faeces. Since the placenta is not an obstacle, strontium can also be detected in the blood of newborns. The largest absorption occurs in the duodenum, while it is most effective in the ileum.

However, the rate of absorption of strontium is significantly lower than that of calcium, so the mother's body protects the infant from strontium and the radiation exposure of radiostrontium with breast milk, since the discrimination factor in the Sr-Ca system can be considered roughly 0.25, according to human physiological measurements. Of course, we can also say that calcium is enriched significantly in the human body compared to strontium **[10]**.

According to our own measurements involving many foods, a daily strontium intake of around 2 mg can only be achieved if a significant part of the food is of animal origin, where the discriminating effect strongly reducing the concentration of strontium relative to calcium prevails. If the food is mainly of plant origin and the plants consumed have a high calcium content (e.g., spinach, sorrel, nuts), then daily strontium amounts exceeding even 10 mg can be incorporated. Thus, the amount of strontium entering the body depends primarily on our diet. According to Pais **[25]**, the strontium content of the human body increases with age.

Most of the strontium entering the human body with food and, to a lesser extent, drinking water, is excreted with the faeces. Takács **[18]** reported that, in an eight-day study, it was determined that the average daily amount of strontium excreted with urine was 0.39 mg, while the amount in faeces was 1.58. According to the studies of Anke et al. **[26]**, roughly 86% of the strontium entering the adult human body is excreted with the faeces, and only 14% is excreted with the urine. According to data from Germany, 32% of the strontium entering the human body came from milk and dairy products, 22% from vegetables, 18% from fruits and 17% from bread and pasta. The amount of strontium entering the body with meats and meat products was essentially negligible **[26].**

One should not be vary of the absence of strontium even when consuming products of animal origin, even if it proves to be an essential element. In some areas of our planet, in East Siberia, North China, Tibet or North Korea, it occurs in the soil and plants at such high concentrations that it can interfere with human and animal metabolism or, by inducing a relative shortage of calcium, it can cause bone formation disorders and joint disorders **[27]**, **[28]**. This is the so-called Urov disease or Kashin-Beck disease (KBD), which has been known for more than 150 years. The cause of the disease, affecting several millions of people, especially those between the ages of 5 and 15, is probably complex: in addition to the intake of large amounts of strontium, the absence of other microelements, the occurrence of mycotoxins in the food, and the contamination of drinking water with phenolic compounds may all play a role.

It has been known since the famous Swiss physician-chemist-botanist Paracelsus (Philippus Theophrastus Aureolus Bombastus von Hohenheim, 1493-1541), i.e., for five centuries, that all microelements are toxic above a certain concentration or dose. Paracelsus wrote: *„Alle Ding' sind Gift und nichts ohn' Gift; allein die Dosis macht, das ein Ding kein Gift ist"*(All things are poison and nothing is without poison; only the dose makes that a thing is no poison) **[31].**

Strontium is considered weakly toxic, since its toxicity level in the diet is around 150 mg/kg.

9. Is strontium an essential microelement?

Numerous test results indicate that for higher plants strontium could be an essential element. Calcium and strontium have a common ion transport in plants. The biochemical significance of strontium is also suggested by the fact that, in the case of a calcium deficiency, the effect of excess magnesium can be prevented by the administration of strontium. At the same time, from the point of view of animal and human physiology, the theory of a common transport system is only valid with certain limitations, since the rate of absorption in the case of strontium is far lower than in the case of calcium, and in the case of strontium, the values of the so-called discrimination factors (the relative Sr/Ca ratio in two successive segments of the metabolic process) is roughly 0.10 between feed and milk and approximately 0.25 between food and breast milk. Compared to the Sr/Ca ratio in the food or feed consumed, the Sr/Ca ratio is reduced significantly in human and animal body tissues.

The time has come to ask the question: is strontium an essential microelement or not? Takács says **[18]**: there are no incontrovertible data that strontium is an essential element. He notes that experiments suggest that, in the case of a strontium-deficient diet, growth inhibition, bone forming disorders and an increasing incidence of caries occurs. Iyengar et al. **[29]** reported data on strontium concentrations measured in different human tissues. Ratios of the maximum and minimum values measured are shown in ***Table 3***.

In a paper published earlier **[1]** it was already discussed that there is a characteristic of non-essential elements that is significantly different from those typical of essential elements. This characteristic is the concentration distribution. For healthy individuals and typical essential elements, the distribution of a certain macro- or microelement in a given body tissue (e.g., blood plasma) or organ (e.g., the heart) shows a normal distribution, and its concentration falls within a relatively narrow range **[9]**, **[6]**, **[1]**. Under different external conditions, due to the control mechanisms, measured concentrations of the microelement in question show relatively small differences, and the vast majority of the measured values will remain within the narrow range calculated for the arithmetic average and characterized by the f=1.58 (i.e., logf=0.2) factor. Parameter f is a factor indicating the standard deviation of the data, characterizing the range by the extreme values obtained by multiplying and dividing the average value. As a result, there is no big difference between the arithmetic and geometric mean values and the median, the concentration range is not wide. Data in **Table 3** indicate that there is only a 2 to 3-fold difference between the highest and lowest measured strontium concentrations, suggesting the the concentration range is quite narrow. As mentioned above, a narrow concentration range is characteristic of essential microelements.

Naturally, the analysis of concentration distribution can only be regarded as decisive when it comes to essentiality, if a large number of samples are analyzed, the healthy individuals participating in the study live in quite different geographic conditions, and their dietary habits differ significantly. To prove that strontium is an essential microelement for the human body, further research is needed, since it is also characteristic of essential microelements that they are components or activators of enzymes that play a decisive role in biochemical processes. Based on this, the existence of enzymes cooperating with strontium has to be verified.

10. Conclusions

Strontium is a microelement that is present in the water-soil-plant-animal-human biological system in significant concentrations, having a common transport system with calcium. In plants and soils, the Ca/Sr ratio is around 100:1, so there is a difference in occurrence concentration of two orders of magnitude. However, the ratio of calcium to strontium in animal and human bodies is roughly 1000:1, so the difference is three orders of magnitude. The reason for this is a strong discriminating ability, meaning that strontium is absorbed from the intestinal tract in much smaller amounts than calcium. Our analyses demonstrated that the value of the Sr/Ca ratio in the human body and its radiostrontium contamination (^{90}Sr/Ca) are much lower than the values that can be measured in the foods consumed.

Strontium may partially replace calcium, but too much strontium is toxic and causes diseases in the body. However, in order to form an opinion on the role of strontium in animal and human physiology, and to prove its possible essential nature, further studies are needed. At the same time, the essential nature of strontium in plant physiology is quite likely. We believe that to clarify its role in human physiology, the analysis of the concentration distribution of healthy human tissues could be helpful, even in the case of significantly different dietary habits.

11. References

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