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Separation of strontium from calcium by the use of sodium hydroxide and its application for the determination of long-term background activity concentrations of ^{90}Sr in 100 km area around Kozloduy Nuclear Power Plant (Bulgaria)

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The method for the determination of ^{90}Sr which employs sodium hydroxide for the separation of strontium from calcium was further improved introducing the use of elevated temperatures. The results from 11-year study of background activity concentrations of ^{90}Sr in different environmental objects in 100 km zone around Kozloduy Nuclear Power Plant (Bulgaria) are presented as an application of the analytical method. The measured mean values are as follows: air precipitation – 0.0015±0.0009 Bq/(m^{2}·d), tap water – 0.0017±0.0012 Bq/L, soil – 1.90±1.26 Bq/kg, grass – 1.54±0.80 Bq/kg, milk – 0.023±0.012 Bq/L and for the Danube river: water – 0.0046±0.0026 Bq/L, bottom sediments – 0.64±0.60 Bq/kg, algae – 1.99±1.56 Bq/kg. The calculated transfer coefficients (soil-grass) are in the range of 0.33–0.84. Between 2 and 5 times reduction in actual background activities of ^{90}Sr is observed compared to 1972–1974.

Introduction

Strontium-90 (pure beta-emitter – $E_{\beta,max}$ = 546 keV, half-life = 28.78 years) has been released to the environment mainly by global fallout following atmospheric nuclear explosions and in lesser degree by nuclear waste discharges and fallout from the Chernobyl accident. Strontium-90 is one of the most radiological important radionuclide because of its chemical similarity with calcium, long half-life and high energy of its daughter nuclide ^{90}Y (pure beta-emitter, $E_{\beta,max}$ = 2280 keV). Strontium can be taken into the body by eating food, drinking water, or breathing air. About 30 to 40% of ingested strontium is absorbed into the bloodstream. About 15% of what enters the bloodstream is deposited in bone (bone surfaces and bone marrow), the remainder goes to soft tissue (kidneys) and it is excreted in urine. Effective biological half-life of ^{90}Sr remaining in the body is close to its physical half-life of 28.8 years.

Therefore, the determination of ^{90}Sr is among the priorities in all instructions for control on the environmental contamination with radionuclides during the last 50 years and it is carried out in huge number of laboratories all over the world.

Many analytical techniques for the determination of radiostrontium in various components of the environment have been developed. Most of them rely of the difference in solubility (precipitation reactions) and newer ones use selective cation-exchange (extraction with crown ethers, etc.). Unfortunately when analyzing radiostrontium in environmental materials with high calcium content (milk, bones, soils, sea water, etc.) only very limited number of procedures can be used. Still most widely used is the method with fumic or concentrated nitric acid for separation of radiostrontium from calcium.[1] Recently, cheaper and safer alternative method has been published which can overcome the problem with high calcium content in various environmental objects.[2,3] The method utilizes the difference in solubility of strontium and calcium hydroxides for separation of radiostrontium.

This work presents the results from additional study on best conditions for separation of strontium from calcium. The results from the last 11 years of determination of ^{90}Sr in different environmental components of 100 km area around Kozloduy Nuclear Power Plant (Bulgaria) are also presented, which are obtained by classical method with fumic nitric acid until 2002 and after that with the method with sodium hydroxide.

Experimental

Reagents and equipment

All chemicals (Merck, Fluka) used were of analytical grade. Hermle Z513 (Germany) centrifuge (with 100, 250 and 500-mL tubes) was used for the separation of the precipitates. Radiochemically separated ^{90}Y sources were measured on proportional gas-flow counter Pegase (Intertechnique, France) or liquid scintillation counter "Guardian" (Wallac, Finland).

Samples

All samples were collected in 100 km area of Kozloduy NPP (Bulgaria, KNPP). Sampling locations are shown in Fig. 1.

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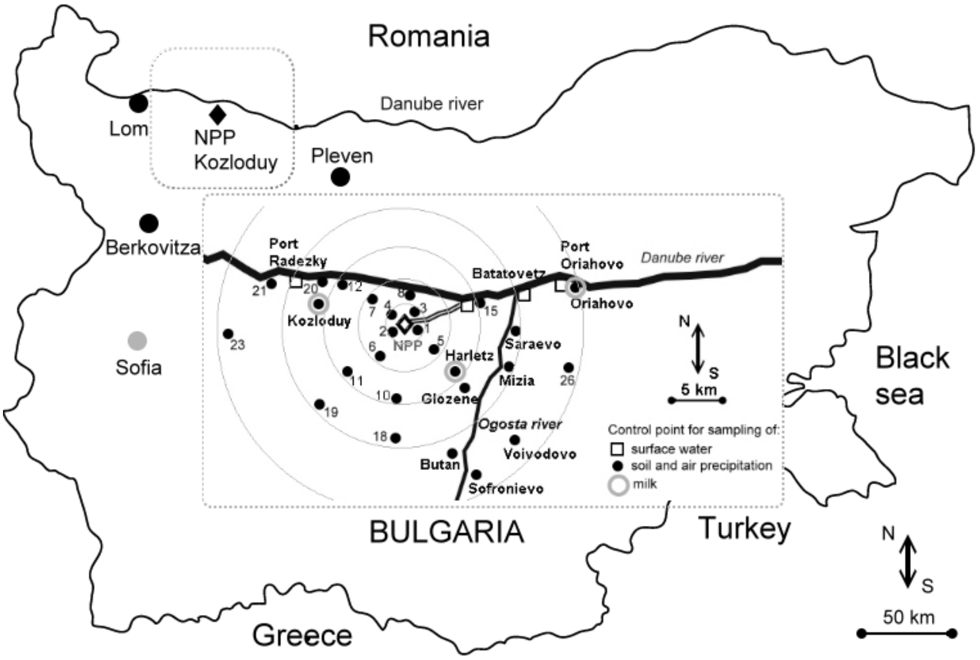

Fig. 1. Map of sampling locations around Kozloduy NPP

All 27 control points were combined in 5 radii around KNPP – radius 1 (0.5 km zone) consists of control points 1 to 4, radius 2 (1.5 km zone) – points 5 to 8, radius 3 (3 km zone) – points 9 to 12 (including village Harletz), radius 4 (6 km zone) – points 13 to 20 (including towns of Kozloduy and Mizia as well as villages Glozene and Saraevo) and radius 5 (12 km zone) – points 21 to 27 (including village of Butan and town of Oriahovo).

Water samples were collected weekly from the Danube river (upstream KNPP: port Radezky 704 km and after KNPP: warm channel 686 km, Batatovetz 682 km, port Oriahovo 678 km) and composite quaternary sample with volume of 40 L was analyzed twice per year after sampling (Harletz, Kozloduy and Oriahovo) in March and November.

9 L composite quaternary milk sample was collected for analysis after monthly sampling (Harletz, Kozloduy and Oriahovo).

Monthly collected filters of air precipitation (wet and dry) were combined for composite quaternary sample (radius 1 to 5).

0.2 kg dry soil composite samples (radius 1 to 5 and towns of Lom, Pleven and Berkovitza) were analyzed annually (in October) after collecting the surface 5 cm soil layer and removing roots and stones.

0.2 kg dry bottom sediments (from water channel connecting KNPP with the Danube river and locations at the Danube river upstream and downstream KNPP correspondingly: port Radezky, Batatovetz and port Oriahovo) were analyzed twice per year.

30 g ash of grass samples were analyzed monthly during vegetation period (March–September) at Harletz, Kozloduy, Oriahovo, Lom, Pleven and Berkovitza.

30 g ash of algae from the Danube river (upstream and downstream KNPP – port Radezky, Batatovetz and port Oriahovo) were analyzed annually.

30 g ash of fodder (sunflower and wheat) were analyzed annually after collecting samples in 3 km area of KNPP.

30 g ash of fish bones sample was analyzed after catch of fish from the Danube river (upstream and downstream KNPP).

Analytical procedure for the separation and determination of ^{90}Sr

Traditional method with fumic nitric acid for separation of strontium from calcium was used for purification of the strontium sources until year 2002.[1] Since 2003 new procedure has been applied for separation and determination of ^{90}Sr in the samples where NaOH instead of fumic HNO_3 was used.[2]

In 2005 the procedure was further modified and improved for the separation step of strontium from calcium. Precipitation of calcium hydroxide was performed under heating to boiling (at ~90 °C) and to concentration of 0.4–0.6M NaOH instead of 0.25M. The precipitate was washed only once with hot 0.2M NaOH. This guarantees minimization of loss of strontium and its better and faster separation from calcium.

Radiochemically separated ^{90}Sr sources were left for 3 weeks to allow ingrowth of ^{90}Y. Yttrium-90 was purified from isotopes of strontium, barium, radium and lead by sulfate and hydroxide precipitations. Finally ^{90}Y was measured as $Y_2(C_2O_4)_3$ on proportional gas-flow counter or as $Y(NO_3)_3$ on liquid scintillation spectrometer in Cherenkov counting mode (distilled water instead of scintillation cocktail).

Quality assurance

In order to ensure high quality of the analytical results, blank, duplicate and spiked samples were analyzed periodically according to internal quality assurance program. Weekly checks of the background and the efficiency were done of the measurement system. Calibration of the detectors was made twice per year. At least one participation took place annually in international interlaboratory comparisons (IAEA – Austria, BfS/PTB – Germany, etc.).

Results from the analysis of reference samples with different matrices were published elsewhere.[2]

Results and discussion

Radiochemical separation of ^{90}Sr

Environmental samples (milk, bones, soil, etc.) usually contain large amounts of calcium. Separation of strontium from such large amounts of calcium is a serious problem where most radiochemical procedures encounter difficulties. Traditional method with fumic nitric acid is often used to overcome the problem.[1] Unfortunately, fumic nitric acid is a very dangerous chemical for the health of the operator, laboratory equipment and the environment, not to mention its relative high price.

Separation of strontium from calcium by the use of NaOH is an easy and convenient way which was used by very limited number of researchers in the last 50 years although it relies on the well known difference in the solubility of the corresponding hydroxides.[2–4] According to our previous experiments it is critical to separate quantitatively strontium from calcium if relatively large amount (1000 mg or more) of strontium carrier is used. In order to minimize chemical losses of strontium 3 consequent washings with 0.2M NaOH combined with intensive stirring must be applied to the precipitates of hydroxides.[2] Finally, usually between 1 and 20 mg of calcium remain unprecipitated.[2]

Attempts were made to eliminate the washings with 0.2M NaOH and thus to decrease the amounts of remaining calcium together with strontium as well as to save time and effort. Initially precipitation of calcium hydroxide was performed at lower temperature (5–10 °C, cooling of the solution with ice), but the results were worse than at normal temperature (25 °C). The opposite approach was tested – precipitating at elevated temperatures. The solution was heated to boiling (~90 °C) and precipitation with hot 6M NaOH was done from 0.3M up to final 0.6M concentration. The results are presented in Table 1. It is obvious that strontium remains quantitatively in the solution for all range of concentrations of the final solution: 0.3–0.6M. Centrifugations of the precipitates were done at 3000 rpm for short time (3 minutes) in order to prevent undesirable cooling of the solution. Under these conditions chemical yield of strontium is practically 100% without additional washing of the precipitate to be required.

Unfortunately, when separating small amounts of strontium from large amounts of calcium (100 and more times difference in mole ratios) even at temperature of 90 °C, losses of strontium occur due to its inclusion in formed calcium precipitate. The only way to recover strontium is by the use of washing with hot (~90 °C) 0.2M NaOH once or twice, depending on the amount of formed precipitate of $Ca(OH)_2$. But in most cases where calcium is present in low concentration no washing is required as can be seen from the results presented in Table 2. The reduced number of separations (only 2) and washings (in worst case only 2, typically only 1) leads to faster analysis, combined with reduced effort and cost.

Table 1. Results for amount of strontium (1000 mg Sr^{2+}) remaining in the solution at 90 °C after addition of 6M NaOH to different final concentrations

Concentration of the solution of NaOH, M	Recovery of Sr, %
0.3	100.5
0.4	100.2
0.5	99.7
0.6	99.4

Table 2. Results for chemical yield of strontium when separations from calcium are done at 90 °C with 6M NaOH with up to 2 washings of the precipitate with hot 0.2M NaOH

$CaCl_2$, g	$Sr(NO_3)_2$, g	Number of separations	No washing, %	1 washing, %	2 washings, %
20	2.5190	2	89.8	95.4	92.3
50	2.4996	2	83.6	91.6	93.8
150	2.5681	2	82.0	90.1	89.1
20	0.5042	2	90.0	94.8	–
50	0.5024	2	86.2	91.7	–
150	0.5074	3	84.7	90.3	–

The amount of remaining Ca^{2+} in the solution is below 3 mg independently of final concentration of the solution (if no washing is applied), which is a better achievement than when hydroxide precipitation is performed at room temperatures – below 20 mg.[2] It seems that the amount of calcium is the lowest boundary of the method for separation of strontium from calcium. But the highest boundary has not been found so far although small quantities of strontium were separated even from 100 g Ca^{2+} or above.

The reason for better separation of strontium from calcium at elevated temperatures is due to the strange property of calcium hydroxide – to decrease its solubility if temperature is increased. At high temperatures most of the compounds including strontium hydroxide increase their solubility, which leads to the possibility for better separation of the two elements even in their relatively small difference in their solubility constants at room temperatures.

Another strange fact was observed in routine application of the method. When concentration of salts in the solution is high then it is almost impossible in practice to precipitate calcium hydroxide even when the calcium content is high. The only solution in that case is to precipitate the calcium-strontium as carbonates to get rid of nitrates, chlorides, etc. After filtration the carbonate precipitate should be dissolved in a minimum volume of nitric acid and then the method can be applied successively. The explanation of this fact lies in the increased ionic strength of the solution, due to increased concentration of other ions, thus preventing solubility constant to be reached and formation of the calcium hydroxide to occur.

Finally it should be mentioned again that the presence of carbonates in sodium hydroxide should be avoided because of formation of undesirable strontium carbonate. Distilled water also should be fresh as well as sodium hydroxide. To minimize unavoidable losses of strontium due to carbonates in water and sodium hydroxide relatively large amount of stable strontium carrier is used.

Background concentrations of ^{90}Sr in 100 km area of Kozloduy NPP

Levels of ^{90}Sr prior to commissioning. Annual discharges of ^{90}Sr from Kozloduy NPP

Kozloduy Nuclear Power Plant (KNPP), located in North Bulgaria, close to the Danube river and the border with Romania, is a potential source of ^{90}Sr. KNPP has 6 units with total installed power of 3760 MW, of which only units V and VI (WWER 1000 MW) are now in operation. Units I, II were decommissioned in December 2002 and units III, IV – in December 2006.

There is lack of data in the literature concerning actual background concentrations of ^{90}Sr in the environment of Bulgaria as well as Balkan Peninsula as a whole. Probably this is because of the difficulties and the intensive chemical labour associated when performing ^{90}Sr determinations.

In the period of 1972–1974, before commissioning, National Center for Radiobiology and Radiation Protection (NCRRP, Ministry of Health) had conducted a series of measurements of ^{90}Sr (and ^{137}Cs) in the environment of the future KNPP. These results are presented in Table 3.[5] The origin of ^{90}Sr found in different environmental objects is only due to atmospheric nuclear tests intensively occurring mainly between 1950 and 1960.[5] These levels of ^{90}Sr form typical background values. In the 30 years history of monitoring of the environment only in 1986 there was significant (1–10 times) increase in some objects (milk, etc.) in levels of ^{90}Sr as a result of the accident at Chernobyl NPP.

Typically liquid and gaseous emissions from KNPP are negligible as it can be seen from Table 4.[6] Normalized emissions of ^{90}Sr as well as of other radionuclides are similar to other nuclear power plants around the world with PWR-reactors.[7] It can be calculated that for the period 1998–2005 the total activity of ^{90}Sr deposited in 30 km area around KNPP was about 0.014 Bq/m^2. Such activities are difficult to be registered with instrumental methods and it is impossible to be distinguished from the typical background ^{90}Sr concentrations.

Table 3. Background activities of ^{90}Sr in 1972–1974 in the environment of the future Kozloduy Nuclear Power Plant[5]

Object	^{90}Sr (1972–1974)
Soil, Bq/kg	5.0 ± 0.4
Water from Danube river, Bq/L	0.012 ± 0.002
Bottom sediments from Danube river, Bq/kg	2.6 ± 0.6
Grass, Bq/kg	4.4 ± 0.3
Wheat (grain), Bq/kg	0.15 ± 0.01
Cow milk, Bq/L	0.111 ± 0.015
Fish bones, Bq/kg	1.48 ± 0.20
Air precipitation, Bq/m^2·d	0.030 ± 0.011

Table 4. Liquid and gaseous discharges (in MBq) of ^{90}Sr from Kozloduy NPP during 1998–2005[6]

Year	Liquid ^{90}Sr emissions, MBq	Gaseous ^{90}Sr emissions, MBq	Total ^{90}Sr emissions, MBq	Normalized ^{90}Sr emissions, MBq/GW·a
1998	2.74	1.51	4.25	2.20
1999	2.32	3.68	6.00	3.32
2000	1.32	2.72	4.04	1.95
2001	12.69	3.74	16.43	7.36
2002	1.22	3.25	4.47	1.94
2003	0.91	2.13	3.04	1.54
2004	0.62	0.51	1.13	0.59
2005	0.65	0.03	0.68	0.32

Table 5. Radioactivity of ^{90}Sr in soil (in Bq/kg, dry weight)

Sampling	Radius 1	Radius 2	Radius 3	Radius 4	Radius 5	Lom	Pleven	Berkovitza
Oct 1995	1.75 ± 0.11	2.24 ± 0.13	2.08 ± 0.11	1.49 ± 0.08	3.07 ± 0.11	0.87 ± 0.08	1.90 ± 0.10	9.17 ± 0.20
Oct 1996	1.04 ± 0.09	2.12 ± 0.11	2.84 ± 0.11	2.26 ± 0.09	3.71 ± 0.14	2.10 ± 0.11	1.55 ± 0.09	4.79 ± 0.11
Oct 1997	1.77 ± 0.09	1.66 ± 0.09	1.92 ± 0.11	2.36 ± 0.12	2.95 ± 0.12	1.59 ± 0.08	3.71 ± 0.17	5.13 ± 0.14
Oct 1998	1.05 ± 0.08	2.11 ± 0.14	2.27 ± 0.14	2.93 ± 0.14	2.52 ± 0.13	1.18 ± 0.08	0.91 ± 0.07	3.47 ± 0.10
Oct 1999	1.47 ± 0.09	1.45 ± 0.11	1.93 ± 0.13	2.72 ± 0.15	2.33 ± 0.12	1.53 ± 0.08	1.18 ± 0.10	5.81 ± 0.20
Oct 2000	1.81 ± 0.08	1.32 ± 0.09	1.14 ± 0.10	1.13 ± 0.08	1.10 ± 0.06	1.05 ± 0.07	0.91 ± 0.08	3.49 ± 0.11
Oct 2001	1.60 ± 0.09	1.16 ± 0.09	1.10 ± 0.10	1.65 ± 0.10	1.84 ± 0.11	2.06 ± 0.08	1.86 ± 0.08	4.10 ± 0.11
Oct 2002	1.93 ± 0.08	3.43 ± 0.12	1.01 ± 0.08	1.09 ± 0.07	1.70 ± 0.07	1.57 ± 0.07	1.86 ± 0.09	n.t.
Oct 2003	1.71 ± 0.08	1.08 ± 0.09	1.48 ± 0.07	2.38 ± 0.09	2.16 ± 0.09	2.35 ± 0.11	1.97 ± 0.11	5.39 ± 0.13
Oct 2004	1.75 ± 0.11	1.66 ± 0.08	2.17 ± 0.11	2.13 ± 0.09	2.07 ± 0.10	2.17 ± 0.09	2.33 ± 0.10	5.58 ± 0.13
Oct 2005	1.46 ± 0.08	1.96 ± 0.09	1.87 ± 0.09	1.97 ± 0.09	1.88 ± 0.09	1.84 ± 0.09	2.37 ± 0.10	6.06 ± 0.17
Average:	1.56 ± 0.31 (±1 SD; *n*=11)	1.87 ± 0.64 (±1 SD; *n*=11)	1.80 ± 0.57 (±1 SD; *n*=11)	2.01 ± 0.61 (±1 SD; *n*=11)	2.30 ± 0.73 (±1 SD; *n*=11)	1.66 ± 0.49 (±1 SD; *n*=11)	1.87 ± 0.79 (±1 SD; *n*=11)	5.30 ± 1.64 (±1 SD; *n*=10)

Soils

The results from the analysis of soils for the period 1995–2005 are presented in Table 5. Figure 2 shows ^{90}Sr activities for a longer period 1974–2005 in the 3 km area around KNPP. Mean activities of ^{90}Sr are in the range of 1.56–2.30 Bq/kg, excluding the town of Berkovitza where a higher mean value of 5.30±1.64 Bq/kg was determined. The reason is probably that Berkovitza is located at higher altitude than the rest of the sampling points and there is normally more rain fall during depositions of radiostrontium mainly in 1950–1960. There is a correlation between altitude and concentrations of ^{90}Sr in soil and for the rest of locations in Bulgaria.[8] The type of soil is also different, typical grey forest soil (Lessive Luvisols), which can better retain strontium, instead of Calcic Chernozems, Haplic Chernozems or Leached Chernozems for the other locations. Similar ^{90}Sr concentrations were measured at other locations in Bulgaria and in other countries:

Bulgaria – mean value of 3.6±1.6 Bq/kg were measured for soils in North Bulgaria (20 control points) and 9.4±4.3 Bq/kg for South Bulgaria (13 points) in 1996 by the National Center for Agricultural Sciences. For the whole territory of Bulgaria a spot distribution of ^{90}Sr for surface soil horizons was observed which explains the wide determined range of 2.9–12.8 Bq/kg.[8]

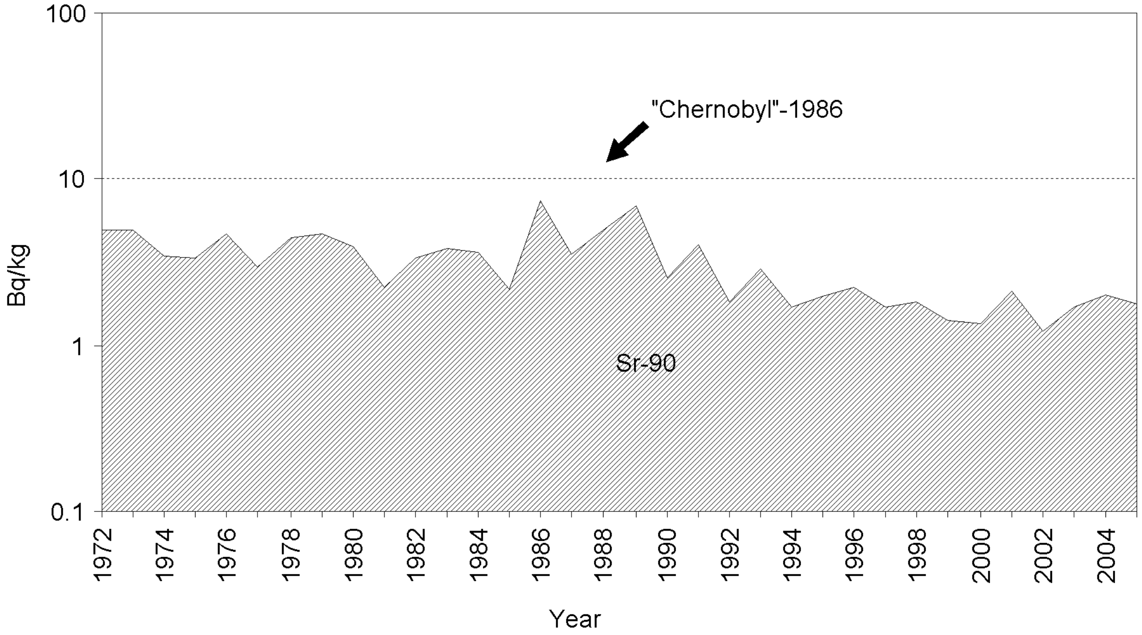

Fig. 2. Average activities of ^{90}Sr in soils from 3 km radius around Kozloduy NPP, 1972–2005[6]

HOLGYE et al.[9] determined a mean value of 2.5 Bq/kg and range of 0.5–6.5 Bq/kg for the soils near Prague, Czech Republic in 2002.

AL HAMARNEH et al.[10] for Jordanian soils in 2002 measured range of 2.8–11.4 Bq/kg.

Although sampling points are located on apparently undisturbed places it seems that mixing of soil horizons occurred 20–40 years ago due to agricultural deep plough. Results for depth vertical distribution of ^{90}Sr in soil of Berkovitza control point are presented in Table 6. ^{90}Sr is almost uniformly distributed in depth through soil horizons 0–25 cm. The same mixing also is observed at the rest sampling locations together with wind and water erosion of the Chernozems soils.

The decrease in the present activities of ^{90}Sr compared to 1972–1974 period is about 2–3 times which requires higher volumes of the soil (above 200 g dry weight) to be analyzed in order to get more reliable result.

Grass and fodder

Mean concentrations of ^{90}Sr in grass are in the range 1.39–1.74 Bq/kg (dry weight) (Table 7 and Fig. 3). Measured activities are similar to those determined for example in Denmark in 1992–1993 by AARKROG et al.[11] 0.4–0.43 Bq/kg, but in fresh weight, which if calculated for dry weight may result in 1–2 Bq/kg (dry weight). There is a good agreement between concentrations of ^{90}Sr in soil and grass. The exception is only Berkovitza where the determined ^{90}Sr activities in grass are the same as for other locations although for Berkovitza there is 2–3 times higher ^{90}Sr activity in soil. It should be mentioned that calcium content in the above soil is on the average 2–3 times higher than for the rest of the locations. This leads to "dilution" of radiostrontium in the soil and subsequently in the plant. So, the difference in soil types determines the difference in transfer coefficients, calculated on the ground of the presented data: 0.77 (for Harletz), 0.79 (for Kozloduy), 0.76 (for Oriahovo), 0.84 (for Lom), 0.84 (for Pleven) and 0.33 (for Berkovitza). Transfer coefficients were calculated according to the equation:

$$\text{TC (soil to grass, fodder)} = \frac{\text{Mean } ^{90}\text{Sr concentration (grass, fodder), Bq/kg}}{\text{Mean } ^{90}\text{Sr concentration (soil), Bq/kg}}$$

Table 6. Depth distribution of ^{90}Sr for soil – Berkovitza (June 2006)

Soil layer Berkovitza, cm	^{90}Sr activity, Bq/kg
0–5	4.07 ± 0.20
5–10	4.17 ± 0.22
10–15	4.57 ± 0.20
15–20	4.48 ± 0.19
20–25	4.19 ± 0.18

Table 7. Radioactivity of ^{90}Sr in grass (in Bq/kg, dry weight)

Sampling	Harletz	Kozloduy	Oriahovo	Lom	Pleven	Berkovitza
May 1995	0.26 ± 0.02	n.t.	n.t.			
Jun 1995	1.03 ± 0.05	n.t.	n.t.			
Jul 1995	1.56 ± 0.06	n.t.	n.t.			
Aug 1995	1.60 ± 0.10	n.t.	n.t.	0.84 ± 0.05	1.00 ± 0.06	0.83 ± 0.04
May 1996	1.49 ± 0.06	n.t.	n.t.			
Jun 1996	1.83 ± 0.13	n.t.	n.t.			
Jul 1996	1.64 ± 0.07	n.t.	n.t.			
Aug 1996	0.72 ± 0.09	n.t.	n.t.	0.82 ± 0.05	1.16 ± 0.05	1.92 ± 0.07
May 1997	2.03 ± 0.08	0.59 ± 0.04	0.90 ± 0.06			
Jun 1997	2.34 ± 0.09	1.03 ± 0.05	0.58 ± 0.04			
Jul 1997	1.76 ± 0.07	1.21 ± 0.06	2.20 ± 0.10			
Aug 1997	1.21 ± 0.07	0.75 ± 0.04	0.56 ± 0.05	0.53 ± 0.10	0.90 ± 0.06	1.87 ± 0.07
May 1998	1.61 ± 0.11	0.30 ± 0.04	2.36 ± 0.08			
Jun 1998	1.46 ± 0.05	1.57 ± 0.11	0.43 ± 0.03			
Jul 1998	3.11 ± 0.10	1.24 ± 0.10	2.37 ± 0.09			
Aug 1998	1.30 ± 0.08	0.94 ± 0.11	1.14 ± 0.06	1.53 ± 0.08	2.55 ± 0.13	0.83 ± 0.06
May 1999	1.05 ± 0.05	0.70 ± 0.07	2.78 ± 0.09			
Jun 1999	1.44 ± 0.06	0.98 ± 0.06	1.52 ± 0.11			
Jul 1999	1.47 ± 0.06	0.69 ± 0.05	2.50 ± 0.07			
Aug 1999	1.18 ± 0.06	0.69 ± 0.05	0.31 ± 0.04	0.62 ± 0.07	1.05 ± 0.07	2.51 ± 0.10
May 2000	2.50 ± 0.07	1.06 ± 0.07	1.51 ± 0.07			
Jun 2000	1.60 ± 0.09	1.90 ± 0.09	1.54 ± 0.06			
Jul 2000	1.53 ± 0.08	3.70 ± 0.13	0.61 ± 0.03			
Aug 2000	1.54 ± 0.09	1.62 ± 0.08	2.27 ± 0.08	0.98 ± 0.05	1.45 ± 0.08	4.03 ± 0.12
May 2001	2.02 ± 0.08	2.44 ± 0.11	1.48 ± 0.06			
Jun 2001	2.16 ± 0.12	1.62 ± 0.10	2.75 ± 0.08			
Jul 2001	0.25 ± 0.05	0.76 ± 0.06	1.69 ± 0.08			
Aug 2001	0.76 ± 0.05	2.46 ± 0.10	1.83 ± 0.12	1.77 ± 0.10	1.50 ± 0.05	2.80 ± 0.06
May 2002	1.19 ± 0.06	1.69 ± 0.09	2.82 ± 0.11			
Jun 2002	0.35 ± 0.03	2.28 ± 0.20	n.t.			
Jul 2002	0.49 ± 0.03	1.24 ± 0.06	n.t.			
Aug 2002	0.68 ± 0.06	0.84 ± 0.06	2.17 ± 0.09	1.99 ± 0.09	1.87 ± 0.09	n.t.
May 2003	0.26 ± 0.02	1.62 ± 0.09	0.66 ± 0.04			
Jun 2003	1.03 ± 0.05	3.63 ± 0.15	3.07 ± 0.10			
Jul 2003	1.56 ± 0.06	4.19 ± 0.17	1.07 ± 0.06			
Aug 2003	1.60 ± 0.10	2.86 ± 0.12	1.09 ± 0.06	1.82 ± 0.07	0.86 ± 0.05	1.16 ± 0.05
May 2004	1.49 ± 0.06	1.33 ± 0.08	3.01 ± 0.11			
Jun 2004	1.83 ± 0.13	0.78 ± 0.09	1.04 ± 0.05			
Jul 2004	1.64 ± 0.07	1.54 ± 0.07	2.18 ± 0.07			
Aug 2004	0.72 ± 0.09	1.60 ± 0.08	1.42 ± 0.08	0.17 ± 0.05	1.26 ± 0.12	0.83 ± 0.04
May 2005	1.77 ± 0.06	1.21 ± 0.09	2.49 ± 0.08			
Jun 2005	0.98 ± 0.06	1.49 ± 0.09	2.51 ± 0.06			
Jul 2005	1.31 ± 0.05	1.73 ± 0.09	3.08 ± 0.09			
Aug 2005	0.68 ± 0.05	2.49 ± 0.15	1.13 ± 0.06	2.31 ± 0.09	1.20 ± 0.07	1.36 ± 0.06
Average:	1.39 ± 0.63 (±1 SD; *n*=44)	1.58 ± 0.92 (±1 SD; *n*=36)	1.74 ± 0.85 (±1 SD; *n*=34)	1.22 ± 0.70 (±1 SD; *n*=11)	1.34 ± 0.50 (±1 SD; *n*=11)	1.92 ± 1.05 (±1 SD; *n*=10)

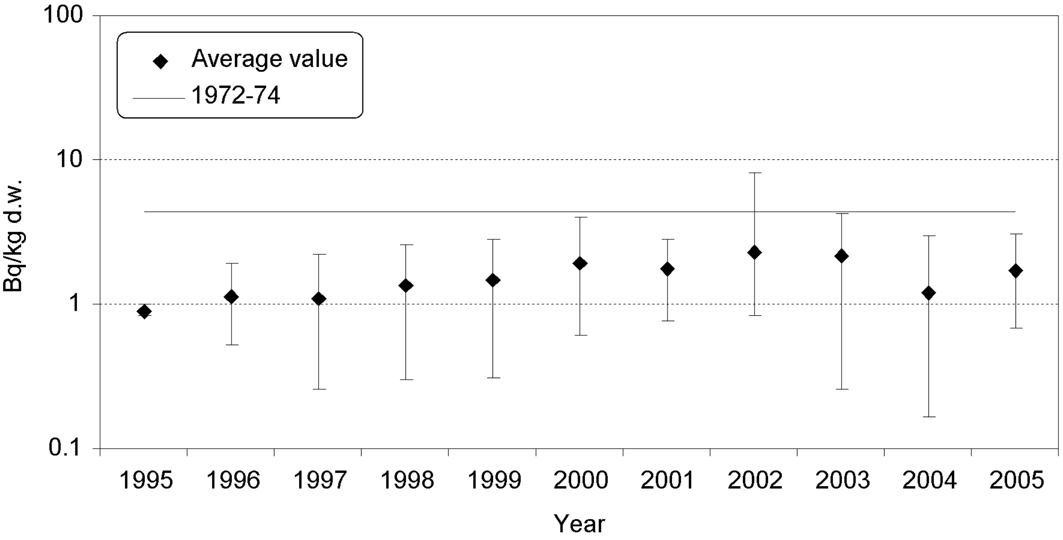

Fig. 3. Concentrations of ^{90}Sr in grass from 100 km zone around Kozloduy NPP [Bq/kg dry weight]

EHKEN and KIRCHNER[12] determined for North German soils transfer coefficients in the range of 0.3–4.0 depending on the soil types, seasons and whether areas are “growth” or “grazed”.

Similar to soils it can be seen a decrease in actual ^{90}Sr activities about 2–3 times compared to 1972–1974 period can be seen which again leads to a requirement for analysis of bigger samples (above 30 g ash).

Analyzed fodder samples were sunflower (seeds and floats), and wheat (grains and straws). Determined mean values for ^{90}Sr-content (in Bq/kg dry weight) were correspondingly: 0.22±0.16 for sunflower seeds, 1.82±0.57 for floats, 0.29±0.31 for wheat grains and 0.81±0.41 for straws (Table 8). Similar values were measured in 1987 for other locations in Bulgaria – Vratza and Yambol, correspondingly for wheat grains 0.13–0.15 Bq/kg and for wheat straws 0.39–0.69 Bq/kg.[6] Different plants (and their parts) accumulate ^{90}Sr from soil in different extent. From the results it can be calculated that the sunflower accumulate ^{90}Sr about 8–9 times higher in floats rather than in seeds and wheat 2–3 times higher in straws than in grains. Transfer coefficients of ^{90}Sr from soil to fodder are: 0.12 (soil – sunflower seeds), 0.97 (soil – sunflower floats), 0.16 (soil – wheat grains) and 0.43 (soil – wheat straws). KROUGLOV et al.[13] obtained transfer coefficients for wheat in the range of 0.1–1 (soil – wheat grains) and 1.5–7 (soil – wheat straws) for Chernobyl-contaminated soils.

Milk

The presence of ^{90}Sr in milk is determined by its transfer through the system soil to grass (plants) to cow due to chemical similarity of strontium with calcium and the inability of plants to distinguish the two elements. Mean concentrations of ^{90}Sr in milk are in a narrow range for the 3 sampling locations: Kozloduy – 0.023±0.014 Bq/L, Harletz – 0.020±0.009 Bq/L and Oriahovo – 0.024±0.010 Bq/L (Table 9 and Fig. 4). Similar mean values were determined by AARKROG et al.[11] in Denmark in 1992–0.042 Bq/L.[11] For Bulgaria, mean value of 1.39 Bq/L was measured by the National Center for Agricultural Sciences in May 1986 (Chernobyl accident) and the concentration of ^{90}Sr slowly have decreased until 1993 to before Chernobyl accident values of 0.03–0.06 Bq/L.[8]

If we assume the average feed intake of cows about 15 kg (dry weight) per day and average yield of milk 13 L per day, we can calculate the average transfer coefficients (grass – milk) for Kozloduy – 0.00096 d/L, Harletz – 0.00095 d/L and Oriahovo – 0.00092 d/L. There is an agreement between our results and the results of GASTBERGER et al.[14] who obtained a range of 0.0005–0.0012 d/L for transfer coefficients of ^{90}Sr (vegetation–milk) at two lowland and two Alpine pastures and 0.0006–0.0013 d/L for stable strontium. FABRI et al.[15] determined 0.0008 d/L value, although IAEA recommends 0.0028 d/L and range of 0.001–0.003 d/L.[15,16] The following formula was used for the calculation of transfer coefficients:

$$\text{TC (grass to milk)} = \frac{\text{Mean } ^{90}\text{Sr concentration (milk), Bq/L}}{\text{Daily consumption grass, kg/d} \cdot \text{Mean } ^{90}\text{Sr concentration (grass), Bq/kg}}, \text{ d/L}$$

Table 8. Radioactivity of ^{90}Sr in fodder around Kozloduy NPP (in Bq/kg, dry weight)

Sampling	Sunflower – seeds	Sunflower – floats	Wheat – grains	Wheat – straws
Jun 1995	0.084 ± 0.01	0.85 ± 0.06	1.03 ± 0.08	0.28 ± 0.06
Jun 1996	0.19 ± 0.02	2.08 ± 0.07	n.t.	n.t.
Sep 1997	0.30 ± 0.03	1.96 ± 0.09	0.62 ± 0.03	1.53 ± 0.06
Aug 1998	0.19 ± 0.03	2.16 ± 0.10	0.12 ± 0.02	0.46 ± 0.03
Sep 1999	0.59 ± 0.03	2.01 ± 0.11	0.22 ± 0.02	0.86 ± 0.06
Aug 2000	0.26 ± 0.03	1.60 ± 0.07	<0.13	0.90 ± 0.09
Aug 2001	n.t.	n.t.	0.20 ± 0.02	1.42 ± 0.05
Jun 2002	n.t.	n.t.	0.29 ± 0.04	0.93 ± 0.05
Jun 2003	0.058 ± 0.01	1.40 ± 0.07	0.08 ± 0.01	0.67 ± 0.05
Aug 2004	0.13 ± 0.02	1.50 ± 0.11	0.08 ± 0.01	0.38 ± 0.04
Jul 2005	0.16 ± 0.03	2.85 ± 0.10	0.11 ± 0.01	0.71 ± 0.04
Average:	0.22 ± 0.16 (±1 SD; n=9)	1.82 ± 0.57 (±1 SD; *n*=9)	0.29 ± 0.31 (±1 SD; *n*=10)	0.81 ± 0.41 (±1 SD; *n*=10)

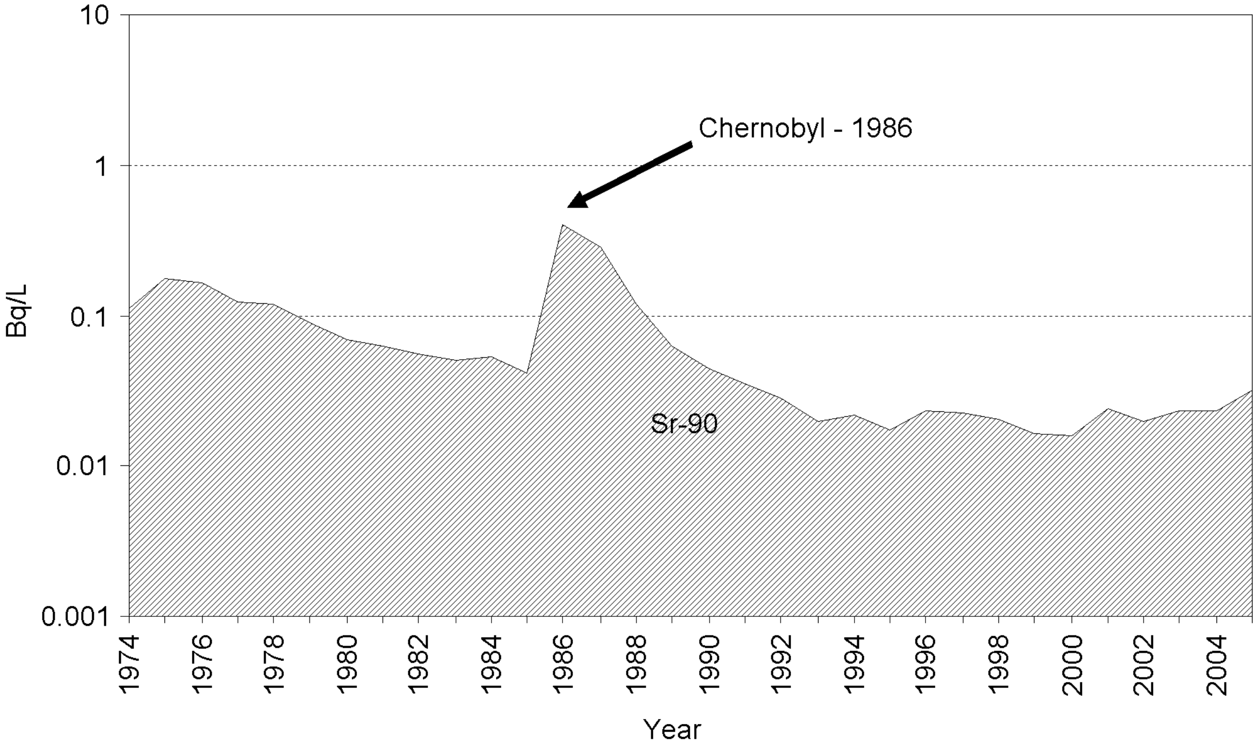

Fig. 4. Mean concentrations of ^{90}Sr in milk in 30 km area around Kozloduy NPP, 1974–2005[6]

Table 9. Radioactivity of ^{90}Sr in milk (in Bq/L, fresh weight)

Sampling	Milk – Kozloduy	Milk – Harletz	Milk – Oriahovo
1st qtr 1995	0.0143 ± 0.0020	0.0094 ± 0.0014	0.0235 ± 0.0019
2nd qtr 1995	0.0246 ± 0.0020	0.0141 ± 0.0013	0.0062 ± 0.0010
3rd qtr 1995	0.0160 ± 0.0022	0.0109 ± 0.0015	0.0095 ± 0.0017
4th qtr 1995	0.0752 ± 0.0035	0.0341 ± 0.0021	0.0303 ± 0.0016
1st qtr 1996	n.t.	0.0208 ± 0.0015	0.0261 ± 0.0016
2nd qtr 1996	0.0255 ± 0.0017	0.0114 ± 0.0019	0.0421 ± 0.0025
3rd qtr 1996	0.0159 ± 0.0018	0.0204 ± 0.0022	0.0292 ± 0.0015
4th qtr 1996	0.0044 ± 0.0011	0.0195 ± 0.0015	n.t.
1st qtr 1997	0.0182 ± 0.0026	0.0168 ± 0.0040	0.0356 ± 0.0031
2nd qtr 1997	0.0234 ± 0.0015	0.0233 ± 0.0018	0.0348 ± 0.0020
3rd qtr 1997	0.0225 ± 0.0018	0.0169 ± 0.0022	0.0258 ± 0.0017
4th qtr 1997	0.0150 ± 0.0016	0.0128 ± 0.0030	0.0277 ± 0.0029
1st qtr 1998	0.0250 ± 0.0017	0.0203 ± 0.0024	0.0117 ± 0.0020
2nd qtr 1998	0.0080 ± 0.0015	0.0204 ± 0.0014	0.0265 ± 0.0016
3rd qtr 1998	0.0349 ± 0.0023	0.0250 ± 0.0025	0.0345 ± 0.0024
4th qtr 1998	0.0530 ± 0.0028	0.0138 ± 0.0018	0.0077 ± 0.0017
1st qtr 1999	<0.0035	0.0211 ± 0.0020	0.0249 ± 0.0016
2nd qtr 1999	0.0166 ± 0.0016	0.0449 ± 0.0022	0.0127 ± 0.0016
3rd qtr 1999	0.0132 ± 0.0014	0.0096 ± 0.0013	0.0074 ± 0.0018
4th qtr 1999	0.0131 ± 0.0016	0.0126 ± 0.0018	0.0157 ± 0.0021
1st qtr 2000	0.0154 ± 0.0017	0.0146 ± 0.0018	0.0246 ± 0.0019
2nd qtr 2000	0.0132 ± 0.0023	0.0181 ± 0.0018	0.0148 ± 0.0017
3rd qtr 2000	0.0072 ± 0.0015	0.0158 ± 0.0017	0.0233 ± 0.0023
4th qtr 2000	0.0338 ± 0.0025	0.0112 ± 0.0021	0.0171 ± 0.0014
1st qtr 2001	0.0096 ± 0.0013	0.0242 ± 0.0014	0.0206 ± 0.0016
2nd qtr 2001	0.0219 ± 0.0020	0.0250 ± 0.0021	0.0138 ± 0.0021
3rd qtr 2001	0.0142 ± 0.0014	0.0138 ± 0.0016	0.0120 ± 0.0013
4th qtr 2001	0.0451 ± 0.0023	0.0397 ± 0.0028	0.0506 ± 0.0022
1st qtr 2002	<0.0037	<0.0032	0.0048 ± 0.0010
2nd qtr 2002	0.0127 ± 0.0012	0.0087 ± 0.0015	0.0115 ± 0.0014
3rd qtr 2002	0.0120 ± 0.0015	<0.0165	0.0347 ± 0.0022
4th qtr 2002	0.0553 ± 0.0021	0.0412 ± 0.0019	0.0340 ± 0.0017
1st qtr 2003	0.0244 ± 0.0016	0.0253 ± 0.0017	0.0155 ± 0.0015
2nd qtr 2003	0.0160 ± 0.0014	0.0152 ± 0.0017	0.0318 ± 0.0025
3rd qtr 2003	0.0304 ± 0.0023	0.0241 ± 0.0017	0.0279 ± 0.0018
4th qtr 2003	0.0229 ± 0.0016	0.0228 ± 0.0016	0.0248 ± 0.0015
1st qtr 2004	0.0263 ± 0.0016	0.0256 ± 0.0016	0.0258 ± 0.0015
2nd qtr 2004	0.0250 ± 0.0018	0.0245 ± 0.0017	0.0210 ± 0.0020
3rd qtr 2004	0.0249 ± 0.0021	0.0121 ± 0.0016	0.0316 ± 0.0023
4th qtr 2004	0.0237 ± 0.0026	0.0188 ± 0.0019	0.0233 ± 0.0022
1st qtr 2005	0.0341 ± 0.0017	0.0131 ± 0.0014	0.0250 ± 0.0017
2nd qtr 2005	0.0206 ± 0.0018	0.0189 ± 0.0014	0.0209 ± 0.0017
3rd qtr 2005	0.0353 ± 0.0020	0.0272 ± 0.0018	0.0293 ± 0.0020
4th qtr 2005	0.0452 ± 0.0021	0.0251 ± 0.0020	0.0395 ± 0.0024
Average:	0.0231 ± 0.0147 (±1 SD; *n*=43)	0.0196 ± 0.0087 (±1 SD; *n*=44)	0.0238 ± 0.0104 (±1 SD; *n*=43)

Table 10. Radioactivity of ^{90}Sr in air precipitation (in Bq/m^{2}·d)

Sampling	Radius 1	Radius 2	Radius 3	Radius 4	Radius 5
1st qtr 1995	<0.0004	<0.0004	<0.0016	<0.0002	<0.0003
2nd qtr 1995	0.0017 ± 0.0003	<0.0007	<0.0011	0.0007 ± 0.0001	0.0008 ± 0.0001
3rd qtr 1995	<0.0005	<0.0005	0.0005 ± 0.0002	0.0006 ± 0.0001	0.0005 ± 0.0001
4th qtr 1995	0.0006 ± 0.0001	0.0010 ± 0.0002	0.0012 ± 0.0001	n.t.	0.0007 ± 0.0001
1st qtr 1996	0.0004 ± 0.0001	<0.0003	0.0003 ± 0.0001	0.0003 ± 0.0001	0.0015 ± 0.0002
2nd qtr 1996	0.0031 ± 0.0002	0.0008 ± 0.0001	0.0008 ± 0.0001	0.0007 ± 0.0001	0.0008 ± 0.0001
3rd qtr 1996	0.0017 ± 0.0001	0.0010 ± 0.0002	0.0010 ± 0.0002	0.0008 ± 0.0001	0.0016 ± 0.0001
4th qtr 1996	0.0015 ± 0.0002	0.0008 ± 0.0001	0.0010 ± 0.0001	0.0006 ± 0.0001	0.0010 ± 0.0001
1st qtr 1997	<0.0007	<0.0007	<0.0007	<0.0005	<0.0003
2nd qtr 1997	0.0013 ± 0.0001	0.0007 ± 0.0001	0.0007 ± 0.0001	0.0008 ± 0.0001	0.0009 ± 0.0001
3rd qtr 1997	0.0007 ± 0.0002	0.0015 ± 0.0002	n.t.	0.0011 ± 0.0001	0.0009 ± 0.0001
4th qtr 1997	0.0014 ± 0.0002	0.0008 ± 0.0002	0.0025 ± 0.0002	0.0011 ± 0.0001	0.0013 ± 0.0001
1st qtr 1998	0.0025 ± 0.0002	0.0008 ± 0.0001	0.0006 ± 0.0001	0.0004 ± 0.0001	0.0004 ± 0.0001
2nd qtr 1998	0.0035 ± 0.0002	0.0008 ± 0.0001	0.0016 ± 0.0002	0.0011 ± 0.0001	0.0011 ± 0.0001
3rd qtr 1998	0.0019 ± 0.0002	0.0031 ± 0.0002	0.0018 ± 0.0002	0.0027 ± 0.0002	0.0016 ± 0.0001
4th qtr 1998	0.0025 ± 0.0002	0.0023 ± 0.0002	0.0026 ± 0.0002	0.0047 ± 0.0002	0.0017 ± 0.0001
1st qtr 1999	0.0020 ± 0.0002	0.0012 ± 0.0002	0.0011 ± 0.0003	0.0013 ± 0.0001	0.0021 ± 0.0002
2nd qtr 1999	n.t.	0.0022 ± 0.0002	0.0027 ± 0.0002	0.0013 ± 0.0001	0.0015 ± 0.0001
3rd qtr 1999	0.0018 ± 0.0002	0.0010 ± 0.0001	0.0015 ± 0.0002	0.0010 ± 0.0001	0.0020 ± 0.0001
4th qtr 1999	0.0025 ± 0.0001	0.0021 ± 0.0001	0.0010 ± 0.0001	0.0015 ± 0.0001	0.0028 ± 0.0001
1st qtr 2000	0.0015 ± 0.0002	0.0016 ± 0.0002	0.0014 ± 0.0002	0.0009 ± 0.0001	0.0014 ± 0.0001
2nd qtr 2000	0.0019 ± 0.0002	0.0023 ± 0.0002	0.0011 ± 0.0001	0.0019 ± 0.0001	0.0019 ± 0.0001
3rd qtr 2000	0.0027 ± 0.0002	0.0020 ± 0.0002	0.0022 ± 0.0002	0.0023 ± 0.0001	0.0022 ± 0.0001
4th qtr 2000	0.0038 ± 0.0003	0.0063 ± 0.0004	0.0029 ± 0.0003	0.0013 ± 0.0002	0.0016 ± 0.0002
1st qtr 2001	0.0019 ± 0.0001	0.0017 ± 0.0001	0.0018 ± 0.0002	0.0011 ± 0.0001	0.0013 ± 0.0001
2nd qtr 2001	0.0027 ± 0.0002	0.0017 ± 0.0001	0.0032 ± 0.0002	0.0023 ± 0.0001	0.0017 ± 0.0001
3rd qtr 2001	0.0044 ± 0.0002	0.0030 ± 0.0002	0.0033 ± 0.0002	0.0024 ± 0.0001	0.0021 ± 0.0001
4th qtr 2001	0.0017 ± 0.0002	0.0024 ± 0.0002	0.0022 ± 0.0002	0.0017 ± 0.0001	0.0013 ± 0.0001
1st qtr 2002	0.0013 ± 0.0001	0.0008 ± 0.0001	0.0009 ± 0.0002	0.0016 ± 0.0001	0.0010 ± 0.0001
2nd qtr 2002	0.0005 ± 0.0001	0.0010 ± 0.0001	0.0009 ± 0.0002	0.0009 ± 0.0001	0.0008 ± 0.0001
3rd qtr 2002	0.0034 ± 0.0002	0.0009 ± 0.0001	0.0017 ± 0.0002	0.0017 ± 0.0001	0.0013 ± 0.0001
4th qtr 2002	0.0010 ± 0.0002	0.0028 ± 0.0002	0.0026 ± 0.0002	0.0030 ± 0.0002	0.0031 ± 0.0001
1st qtr 2003	0.0016 ± 0.0002	0.0024 ± 0.0002	0.0023 ± 0.0002	0.0019 ± 0.0002	0.0009 ± 0.0002
2nd qtr 2003	0.0032 ± 0.0002	0.0013 ± 0.0002	0.0018 ± 0.0002	0.0007 ± 0.0001	0.0011 ± 0.0002
3rd qtr 2003	0.0012 ± 0.0002	0.0016 ± 0.0002	0.0030 ± 0.0002	0.0017 ± 0.0001	0.0019 ± 0.0001
4th qtr 2003	0.0014 ± 0.0001	0.0027 ± 0.0002	0.0022 ± 0.0002	0.0014 ± 0.0001	0.0018 ± 0.0001
1st qtr 2004	0.0007 ± 0.0001	0.0012 ± 0.0001	0.0007 ± 0.0001	0.0018 ± 0.0001	0.0014 ± 0.0002
2nd qtr 2004	0.0020 ± 0.0002	0.0017 ± 0.0002	0.0009 ± 0.0001	0.0012 ± 0.0001	0.0014 ± 0.0001
3rd qtr 2004	0.0008 ± 0.0001	0.0010 ± 0.0001	0.0009 ± 0.0001	0.0010 ± 0.0001	0.0013 ± 0.0001
4th qtr 2004	0.0012 ± 0.0001	0.0042 ± 0.0002	0.0021 ± 0.0002	0.0011 ± 0.0001	0.0014 ± 0.0001
1st qtr 2005	0.0012 ± 0.0001	0.0030 ± 0.0002	0.0016 ± 0.0002	0.0015 ± 0.0001	0.0012 ± 0.0001
2nd qtr 2005	0.0009 ± 0.0001	0.0012 ± 0.0002	0.0012 ± 0.0001	0.0012 ± 0.0001	0.0016 ± 0.0001
3rd qtr 2005	0.0013 ± 0.0002	0.0014 ± 0.0002	0.0007 ± 0.0002	0.0012 ± 0.0001	0.0006 ± 0.0001
4th qtr 2005	0.0014 ± 0.0002	0.0021 ± 0.0002	0.0017 ± 0.0002	0.0010 ± 0.0001	0.0007 ± 0.0001
Average:	0.0017 ± 0.0010 (±1 SD; *n*=43)	0.0017 ± 0.0011 (±1 SD; *n*=44)	0.0016 ± 0.0008 (±1 SD; *n*=43)	0.0014 ± 0.0008 (±1 SD; *n*=43)	0.0013 ± 0.0006 (±1 SD; *n*=44)

Air precipitation

Resuspension of the top soil layer, as well as inclusion, during vegetation period, of organic matter (small blossoms, leaves, etc.) in the sampling device, are the main reasons to measure in atmospheric precipitations concentrations of ^{90}Sr above MDA's. Registered mean activities are in narrow range of 0.0013–0.0017 Bq/(m^{2}·d) for all 5 control radii (Table 10). In Denmark were determined similar average values of 0.0010 and 0.0011 Bq/(m^{2}·d) for 1992 and 1993.[11]

There is a relationship between mass of the ashed samples and the measured absolute ^{90}Sr activities. If specific activity is expressed in Bq/kg (ash) instead of Bq/(m^2·d), then the calculated results will be similar to the specific activities of the surrounding soils. This leads to the conclusion that the measured ^{90}Sr activity in air precipitation is a result mainly of the resuspension of the top soil.

Probably aerosole filters used in the sampling device contain some low concentrations of ^{90}Sr, because they are made from celulose. And that should also be kept in mind when calculating final results.

Surface and tap water

Table 11 and Figure 5 present measured activities of ^{90}Sr in surface water of the Danube river upstream and downstream KNPP. There is no difference between mean concentrations upstream (port Radezky 704 km – 0.0046±0.0025 Bq/L) and downstream KNPP (warm channel 686 km – 0.0043±0.0025 Bq/L, Batatovetz 682 km – 0.0048±0.0024 Bq/L, port Oriahovo 678 km – 0.0049±0.0027 Bq/L). PENTSCHEV et al.[17] in 1964–1974 determined in the Bulgarian part of river Danube ^{90}Sr activities in the range of 0.0030–0.0100 Bq/L and PODEANU et al.[19] in Romanian part – 0.0004–0.0400 Bq/L.

Table 11. Radioactivity of ^{90}Sr in water from Danube river (in Bq/L)

Sampling	port Radezky, 704 km	channel KNPP, 686 km	Batatovetz, 682 km	port Oriahovo, 678 km
1st qtr 1995	n.t.	0.0006 ± 0.0001	n.t.	0.0031 ± 0.0003
2nd qtr 1995	n.t.	<0.0010	n.t.	0.0035 ± 0.0003
3rd qtr 1995	0.0013 ± 0.0002	0.0024 ± 0.0005	0.0027 ± 0.0004	0.0021 ± 0.0004
4th qtr 1995	0.0013 ± 0.0003	<0.0013	n.t.	0.0036 ± 0.0004
1st qtr 1996	n.t.	n.t.	n.t.	0.0038 ± 0.0003
2nd qtr 1996	n.t.	n.t.	n.t.	0.0048 ± 0.0003
3rd qtr 1996	0.0038 ± 0.0004	0.0038 ± 0.0003	0.0073 ± 0.0004	0.0067 ± 0.0004
4th qtr 1996	n.t.	0.0066 ± 0.0004	n.t.	0.0048 ± 0.0004
1st qtr 1997	0.0044 ± 0.0003	0.0068 ± 0.0004	n.t.	0.0028 ± 0.0003
2nd qtr 1997	0.0032 ± 0.0003	0.0025 ± 0.0003	0.0044 ± 0.0004	0.0046 ± 0.0004
3rd qtr 1997	0.0049 ± 0.0004	0.0106 ± 0.0006	0.0131 ± 0.0006	0.0032 ± 0.0004
4th qtr 1997	0.0011 ± 0.0003	0.0037 ± 0.0004	n.t.	0.0071 ± 0.0004
1st qtr 1998	0.0032 ± 0.0004	0.0036 ± 0.0003	n.t.	0.0048 ± 0.0004
2nd qtr 1998	0.0030 ± 0.0003	0.0030 ± 0.0003	0.0031 ± 0.0004	0.0056 ± 0.0004
3rd qtr 1998	0.0016 ± 0.0003	0.0029 ± 0.0004	0.0038 ± 0.0005	0.0052 ± 0.0005
4th qtr 1998	0.0067 ± 0.0005	0.0057 ± 0.0005	n.t.	0.0031 ± 0.0004
1st qtr 1999	0.0083 ± 0.0005	n.t.	n.t.	0.0045 ± 0.0004
2nd qtr 1999	0.0031 ± 0.0004	0.0108 ± 0.0005	0.0035 ± 0.0005	0.0040 ± 0.0003
3rd qtr 1999	0.0056 ± 0.0005	0.0053 ± 0.0005	0.0067 ± 0.0005	0.0149 ± 0.0010
4th qtr 1999	0.0053 ± 0.0004	0.0037 ± 0.0003	n.t.	0.0030 ± 0.0003
1st qtr 2000	0.0044 ± 0.0003	0.0044 ± 0.0004	n.t.	0.0029 ± 0.0004
2nd qtr 2000	0.0108 ± 0.0005	0.0038 ± 0.0004	0.0035 ± 0.0003	0.0026 ± 0.0003
3rd qtr 2000	0.0127 ± 0.0009	0.0134 ± 0.0011	0.0030 ± 0.0004	0.0101 ± 0.0005
4th qtr 2000	0.0027 ± 0.0004	0.0036 ± 0.0004	n.t.	0.0078 ± 0.0004
1st qtr 2001	0.0031 ± 0.0003	0.0032 ± 0.0003	n.t.	0.0029 ± 0.0003
2nd qtr 2001	0.0074 ± 0.0006	0.0041 ± 0.0004	0.0066 ± 0.0005	0.0025 ± 0.0003
3rd qtr 2001	0.0033 ± 0.0003	0.0033 ± 0.0003	0.0040 ± 0.0003	0.0064 ± 0.0006
4th qtr 2001	0.0029 ± 0.0003	0.0030 ± 0.0003	n.t.	0.0092 ± 0.0011
1st qtr 2002	0.0028 ± 0.0003	0.0031 ± 0.0003	n.t.	0.0121 ± 0.0005
2nd qtr 2002	0.0024 ± 0.0003	0.0021 ± 0.0003	0.0030 ± 0.0003	0.0036 ± 0.0003
3rd qtr 2002	0.0061 ± 0.0004	0.0045 ± 0.0003	0.0058 ± 0.0004	0.0046 ± 0.0003
4th qtr 2002	0.0081 ± 0.0012	0.0050 ± 0.0005	n.t.	0.0043 ± 0.0003
1st qtr 2003	0.0052 ± 0.0004	0.0044 ± 0.0004	n.t.	0.0027 ± 0.0003
2nd qtr 2003	0.0036 ± 0.0003	0.0034 ± 0.0003	0.0044 ± 0.0003	0.0032 ± 0.0003
3rd qtr 2003	0.0037 ± 0.0003	0.0032 ± 0.0003	0.0032 ± 0.0003	0.0036 ± 0.0003
4th qtr 2003	0.0067 ± 0.0004	0.0035 ± 0.0003	n.t.	0.0031 ± 0.0003
1st qtr 2004	0.0032 ± 0.0003	0.0052 ± 0.0003	n.t.	0.0055 ± 0.0004
2nd qtr 2004	0.0042 ± 0.0003	0.0036 ± 0.0003	0.0032 ± 0.0003	0.0031 ± 0.0003
3rd qtr 2004	0.0034 ± 0.0003	0.0014 ± 0.0003	0.0063 ± 0.0003	0.0035 ± 0.0003
4th qtr 2004	0.0027 ± 0.0003	0.0058 ± 0.0004	n.t.	0.0021 ± 0.0004
1st qtr 2005	0.0039 ± 0.0003	0.0038 ± 0.0003	n.t.	0.0049 ± 0.0003
2nd qtr 2005	0.0053 ± 0.0004	0.0039 ± 0.0003	0.0040 ± 0.0003	0.0042 ± 0.0004
3rd qtr 2005	0.0089 ± 0.0004	0.0068 ± 0.0004	0.0042 ± 0.0003	0.0052 ± 0.0004
4th qtr 2005	0.0054 ± 0.0004	0.0051 ± 0.0005	n.t.	0.0055 ± 0.0004
Average:	0.0046 ± 0.0025 (±1 SD; *n*=39)	0.0043 ± 0.0025 (±1 SD; *n*=41)	0.0048 ± 0.0024 (±1 SD; *n*=20)	0.0049 ± 0.0027 (±1 SD; *n*=44)

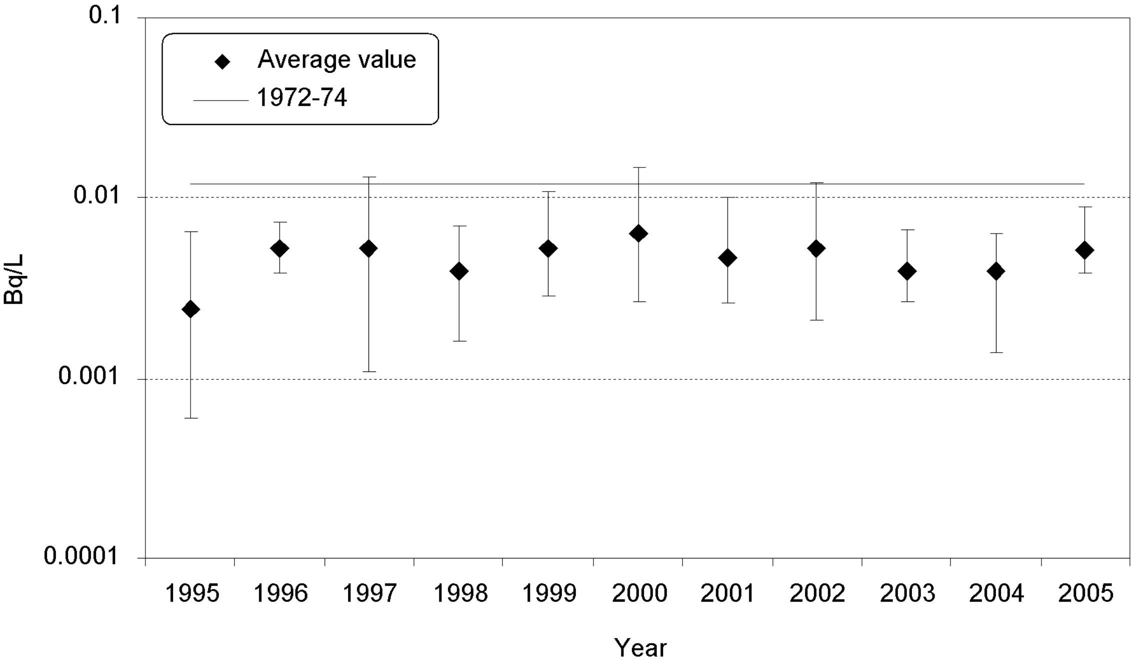

Fig. 5. Background activities of ^{90}Sr in surface water from Danube river around Kozloduy NPP

After the Chernobyl accident concentrations of ^{90}Sr in the Danube close to the Black Sea remain unchanged in the range of 0.0010–0.0040 Bq/L, according to the results of GEDEONOV et al.[21] for the period 1986–1990.

In other big European rivers such as Dnepr and Dnestar higher concentrations were determined than in the Danube, in the range of 0.0050–0.0410 Bq/L for the period of 1987–1995, probably due to higher activities of ^{90}Sr in the surrounding soils and the proximity of Chernobyl NPP.[19–21] AARKROG et al.[11] measured in surface waters (lakes and rivers) in Denmark 0.0008–0.0012 Bq/L for 1992–1993, where higher activities were registered in lakes. For the last 33 years it can be noted a decrease only about 2 times in ^{90}Sr activities in the Danube water. It seems that the self-cleaning of the water is difficult process and it is determined only by the physical half-life of ^{90}Sr – 28.8 years. The reason lies in the high solubility of strontium which constantly goes into water from the surrounding soils and further it is not possible to be retained by bottom sediments.

In tap waters around KNPP lower mean activities in the range 0.0015–0.0023 Bq/L were measured than in the surface waters of the Danube river (Table 12). Tap waters are of underground origin (from 5–10 m depth) and because of the relatively high mobility of strontium, ^{90}Sr from upper soil layers goes into underground tap waters. It should be mentioned that the measured activities are very close to MDA of 0.0007–0.0010 Bq/L and are associated with higher uncertainty. Thus 40 L volume of analyzed samples is insufficient and an increase to 100–200 L is required in order to lower further MDA. This also can be combined with an extension in counting times (currently 6000 s). AARKROG et al.[11] had determined lower activities of ^{90}Sr for tap water in Denmark with mean value of 0.0002 and range of 0.00002–0.0036 Bq/L.[11]

Algae

Table 13 presents the results for ^{90}Sr concentrations in algae from river Danube. Mean activities are correspondingly: 2.79±2.39 Bq/kg dry weight (d.w.) upstream KNPP (port Radezky) and downstream KNPP: 1.69±0.99 Bq/kg d.w. (Batatovetz) and 1.71±0.87 Bq/kg d.w. (port Oriahovo). Transfer coefficients of ^{90}Sr for the system water → algae can be calculated for the 3 sampling locations and are 607 L/kg for port Radezky, 352 L/kg for Batatovetz and 349 L/kg for port Oriahovo. It is obvious that algae are a good indicator for a presence of ^{90}Sr in water due to the high bioaccumulation. Sometimes increased activities can be measured in algae because of the inability to remove all mud contained in the sample even after good washing. AARKROG et al.[11] had measured in algae from Baltic sea (Roskilde fiord, Denmark) similar average values for ^{90}Sr in the range of 2.2–2.5 Bq/kg d.w. in 1992–1993.[11]

Table 12. Radioactivity of ^{90}Sr in tap water (in Bq/L)

Sampling	Kozloduy	Harletz	Oriahovo
Mar 1995	0.0010 ± 0.0003	<0.0009	<0.0010
Nov 1995	<0.0014	<0.0010	0.0020 ± 0.0003
Mar 1996	0.0016 ± 0.0003	0.0011 ± 0.0003	0.0028 ± 0.0003
Nov 1996	0.0040 ± 0.0003	0.0035 ± 0.0003	0.0032 ± 0.0006
Mar 1997	0.0025 ± 0.0003	<0.0008	0.0018 ± 0.0003
Nov 1997	0.0016 ± 0.0004	<0.0008	0.0028 ± 0.0004
Mar 1998	0.0039 ± 0.0004	0.0015 ± 0.0003	0.0021 ± 0.0003
Nov 1998	0.0053 ± 0.0005	0.0017 ± 0.0004	0.0038 ± 0.0003
Mar 1999	n.t.	0.0036 ± 0.0004	0.0010 ± 0.0003
Nov 1999	n.t.	0.0018 ± 0.0003	0.0020 ± 0.0003
Mar 2000	0.0018 ± 0.0003	<0.0009	0.0029 ± 0.0003
Nov 2000	0.0015 ± 0.0003	<0.0008	0.0019 ± 0.0003
Mar 2001	0.0034 ± 0.0003	0.0021 ± 0.0003	0.0011 ± 0.0002
Nov 2001	0.0014 ± 0.0002	0.0008 ± 0.0002	n.t.
Mar 2002	0.0015 ± 0.0003	0.0031 ± 0.0004	<0.0013
Nov 2002	0.0009 ± 0.0003	0.0008 ± 0.0002	0.0043 ± 0.0003
Mar 2003	<0.0010	<0.0009	0.0025 ± 0.0003
Nov 2003	0.0044 ± 0.0003	<0.0010	0.0021 ± 0.0003
Mar 2004	<0.0010	<0.0007	<0.0010
Nov 2004	0.0016 ± 0.0003	<0.0008	0.0020 ± 0.0003
Mar 2005	<0.0007	0.0026 ± 0.0004	0.0025 ± 0.0003
Nov 2005	0.0016 ± 0.0003	<0.0009	0.0022 ± 0.0003
Average:	0.0020 ± 0.0013 (±1 SD; *n*=20)	0.0015 ± 0.0009 (±1 SD; *n*=22)	0.0023 ± 0.0009 (±1 SD; *n*=21)

Fish bones

The results for ^{90}Sr activities in fish bones from the Danube river are presented in Table 14 and Fig. 6. With few exceptions only 3 types of fish were analyzed – *Barbus barbus*, *Hypophthalmichthys molitrix* and *Aristichthys nobilis*. There is correlation between total mass of the fish and the measured activities of ^{90}Sr in bones. The mean results upstream (1.39±0.67 Bq/kg fresh weight) and downstream KNPP (1.52±1.18 Bq/kg fresh weight) are similar.

Factors which affect the measured activities are the age and type of fish, and of less importance location where the fishing takes place because of relatively high mobility of fish.

Correlation between ^{90}Sr content in algae and fish bones should exists but due to mobility of fish it is difficult to be determined with the present data.

Bottom sediments

Concentrations of ^{90}Sr in bottom sediments in river Danube are presented in Table 15. It should be mentioned that sampling of the bottom sediments took place close to the river coasts due to sampling being unable to be performed in the middle of the river because of the high depth of the river and the relatively high speed of the stream. Dissolution of ^{90}Sr from the surrounding soil of the coast and transfer in water are the reasons for measuring activities of ^{90}Sr. Mean concentrations of ^{90}Sr are in the range of 0.57–0.69 Bq/kg dry weight for the period of 1995–2005. The measured values are almost 3–4 times lower than in 1972–1974.[5]

Table 13. Radioactivity of ^{90}Sr in algae from Danube river (in Bq/kg, dry weight)

Sampling	port Radezky, 704 km	Batatovetz, 682 km	port Oriahovo, 678 km
Jun 1995	<0.81	2.80 ± 0.19	n.t.
Jun 1996	4.61 ± 0.26	1.67 ± 0.13	n.t.
Sep 1997	1.10 ± 0.18	0.71 ± 0.15	0.95 ± 0.11
Aug 1998	2.02 ± 0.24	3.60 ± 0.37	1.20 ± 0.19
Sep 1999	n.t.	1.87 ± 0.21	n.t.
Aug 2000	0.70 ± 0.13	0.36 ± 0.11	1.48 ± 0.14
Aug 2001	1.53 ± 0.24	1.48 ± 0.24	1.33 ± 0.14
Jun 2002	1.55 ± 0.19	1.23 ± 0.12	3.58 ± 0.31
Jun 2003	5.44 ± 0.28	1.51 ± 0.26	1.72 ± 0.18
Aug 2004	7.16 ± 0.29	2.80 ± 0.19	1.72 ± 0.13
2005	n.t.	n.t.	n.t.
Average:	2.77 ± 2.39 (±1 SD; *n*=9)	1.69 ± 0.99 (±1 SD; *n*=10)	1.71 ± 0.87 (±1 SD; *n*=7)

Table 14. Radioactivity of ^{90}Sr in fish bones from Danube river (in Bq/kg, fresh weight)

Sampling	Before KNPP	Sampling	After KNPP
28.6.1995	1.78 ± 0.09	01.6.1995	<0.27
15.9.1995	1.15 ± 0.09	06.9.1995	2.42 ± 0.12
28.6.1996	1.37 ± 0.10	12.6.1996	2.09 ± 0.11
20.8.1996	2.44 ± 0.11	17.10.1996	1.87 ± 0.08
1997	n.t.	23.7.1997	3.81 ± 0.15
20.2.1998	1.71 ± 0.26	23.7.1997	3.81 ± 0.15
30.3.1998	0.85 ± 0.13	10.9.1997	1.54 ± 0.11
06.5.1998	1.09 ± 0.09	25.2.1998	3.02 ± 0.28
06.5.1998	1.04 ± 0.13	25.2.1998	1.15 ± 0.07
17.6.1998	0.31 ± 0.07	30.3.1998	<0.20
29.9.1998	1.03 ± 0.12	06.5.1998	3.60 ± 0.15
21.11.1999	0.68 ± 0.08	17.6.1998	1.34 ± 0.12
13.1.2000	1.43 ± 0.13	24.6.1998	5.33 ± 0.25
17.8.2000	2.03 ± 0.10	18.9.1998	0.81 ± 0.08
28.8.2000	1.12 ± 0.11	16.6.1999	<0.65
17.10.2000	0.38 ± 0.08	24.6.1999	1.48 ± 0.14
12.1.2001	2.10 ± 0.21	22.7.1999	1.52 ± 0.12
13.3.2001	2.32 ± 0.14	20.8.1999	3.36 ± 0.16
18.4.2001	0.39 ± 0.08	24.9.1999	0.55 ± 0.06
20.6.2001	0.52 ± 0.07	01.10.1999	1.64 ± 0.29
27.8.2002	1.72 ± 0.17	19.4.2000	0.97 ± 0.12
02.10.2002	0.99 ± 0.13	29.11.2000	0.59 ± 0.07
04.11.2003	0.98 ± 0.08	28.2.2001	1.38 ± 0.30
10.2.2004	1.73 ± 0.10	19.4.2001	1.28 ± 0.11
14.5.2004	1.28 ± 0.09	21.6.2001	0.49 ± 0.07
26.4.2005	3.02 ± 0.28	05.2.2002	0.93 ± 0.09
02.6.2005	1.15 ± 0.07	17.10.2003	1.07 ± 0.08
17.8.2005	<0.20	25.5.2004	1.30 ± 0.09
26.10.2005	3.60 ± 0.15	15.10.2004	1.34 ± 0.10
08.12.2005	1.34 ± 0.12	15.12.2004	0.64 ± 0.06
		24.2.2005	1.05 ± 0.05
Average:	1.39 ± 0.67 (±1 SD; *n*=29)	Average:	1.52 ± 1.18 (±1 SD; *n*=31)

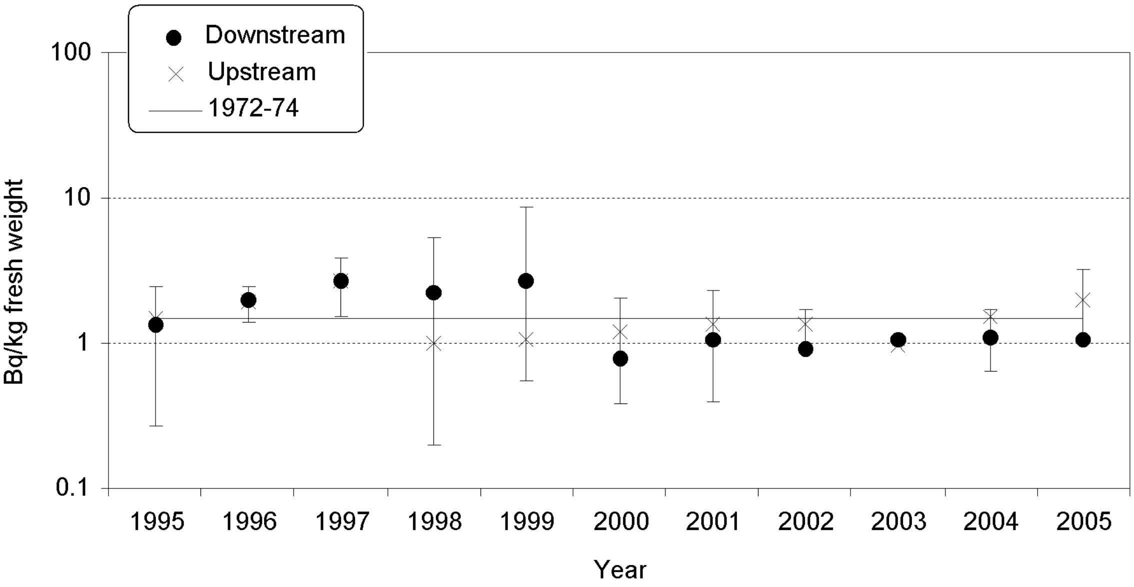

Fig. 6. Mean concentrations of ^{90}Sr in fish bones from Danube river

Table 15. Radioactivity of ^{90}Sr in bottom sediments from Danube river (in Bq/kg, dry weight)

Sampling	Radezky, 704 km	channel KNPP, 686 km	Batatovetz, 682 km	Oriahovo, 678 km
Jun 1995	<0.08	0.50 ± 0.09	<0.23	n.t.
Sep 1995	0.29 ± 0.09	0.50 ± 0.08	<0.37	n.t.
Jun 1996	0.66 ± 0.07	1.11 ± 0.07	0.43 ± 0.06	n.t.
Sep 1996	0.54 ± 0.05	<0.25	0.77 ± 0.06	n.t.
Jun 1997	0.36 ± 0.07	0.43 ± 0.09	0.45 ± 0.06	0.73 ± 0.07
Sep 1997	0.35 ± 0.06	2.08 ± 0.14	0.97 ± 0.08	1.03 ± 0.08
Jun 1998	0.32 ± 0.09	0.67 ± 0.07	0.32 ± 0.09	0.44 ± 0.06
Sep 1998	<0.15	0.24 ± 0.04	0.72 ± 0.10	<0.19
Jun 1999	<0.21	<0.22	0.56 ± 0.07	0.94 ± 0.09
Sep 1999	<0.21	0.34 ± 0.08	0.24 ± 0.06	0.22 ± 0.06
Jun 2000	0.75 ± 0.08	0.57 ± 0.08	0.34 ± 0.08	0.44 ± 0.06
Sep 2000	0.41 ± 0.07	<0.23	0.32 ± 0.06	0.66 ± 0.06
Jun 2001	0.19 ± 0.05	<0.21	0.58 ± 0.07	0.16 ± 0.05
Sep 2001	0.23 ± 0.07	0.67 ± 0.08	0.30 ± 0.05	0.31 ± 0.08
Jun 2002	0.68 ± 0.06	0.60 ± 0.06	<0.15	0.76 ± 0.06
Sep 2002	0.79 ± 0.06	<0.21	0.55 ± 0.06	1.33 ± 0.09
Jun 2003	1.09 ± 0.09	0.54 ± 0.07	1.93 ± 0.09	0.32 ± 0.07
Sep 2003	0.36 ± 0.08	0.68 ± 0.10	0.74 ± 0.08	0.53 ± 0.08
Sep 2004	1.31 ± 0.08	1.47 ± 0.10	0.30 ± 0.05	0.33 ± 0.06
Jul 2005	0.61 ± 0.08	1.41 ± 0.07	1.41 ± 0.08	3.32 ± 0.11
Sep 2005	2.34 ± 0.11	1.51 ± 0.09	1.51 ± 0.09	0.64 ± 0.07
Average:	0.57 ± 0.51 (±1 SD; *n*=21)	0.69 ± 0.52 (±1 SD; *n*=21)	0.69 ± 0.52 (±1 SD; *n*=21)	0.67 ± 0.71 (±1 SD; *n*=17)

Conclusions

Improvement of the procedure for the separation of strontium from calcium was achieved by performing hydroxide precipitations at elevated temperatures (80–90 °C) in order to obtain better purification and minimization of the loss of strontium. The number of washings with hot 0.2M NaOH is reduced and depends only of the amount of the formed precipitates which additionally saves laboratory time and effort.

The improved method was applied successfully for routine analysis of radiostrontium in environmental samples around Kozloduy Nuclear Power Plant. There is good comparison in the results obtained by the classical method with fumic nitric acid and the new method with sodium hydroxide.

The presence of ^{90}Sr in environmental samples around Kozloduy Nuclear Power Plant is determined only by the global fallout after nuclear atmospheric tests and in smaller extent by the Chernobyl accident. Annual gaseous and liquid discharges from the power plant are insignificant, in average below 0.0018 Bq/(m^{2}·year), and their contribution in the environment can not be measured.

The obtained background activities of ^{90}Sr in different environmental objects are typical for the geographical region and the northern hemisphere as a whole. Similar results were measured and by other researchers throughout the world.

For a number of environmental objects (milk, soil, vegetation, etc.) a decrease in the levels of ^{90}Sr had occurred for the period of 1972–2005. For some samples like milk, the decrease is about 5 times. That is why the determination of actual and future fallout concentrations of ^{90}Sr will require conducting the analyses with more sensitive measurement equipment combined with the use of radiochemical analytical methods capable of dealing with increased amount of the samples.

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References

1. Measurement of Radionuclide in Food and Environment, IAEA Technical Report Series, No. 295, IAEA, Vienna, 1989.
2. L. POPOV, X. HOU, S. P. NIELSEN, Y. YU, R. DJINGOVA, I. KULEFF, J. Radioanal. Nucl. Chem., 269 (2006) 161.
3. Q. CHEN, X. HOU, Y. YU, H. DAHLGAARD, S. P. NIELSEN, Anal. Chim. Acta, 466 (2002) 109.
4. F. PATTI, A. HERNANDEZ, Anal. Chim. Acta, 55 (1971) 325.
5. V. MARINOV et al., Influence of Kozloduy Nuclear Power Plant on the Environment and Health Condition of the Population and Workers, Expert Report, KNSB, Sofia, 1993 (in Bulgarian).
6. R. TZIBRANSKI, V. AVRAMOV, L. POPOV, I. NAIDENOV, Results from Radiation Monitoring of the Environment around Kozloduy Nuclear Power Plant in 2005, Annual Report, Kozloduy, 2006 (in Bulgarian).
7. UNSCEAR 2000 Report, Vol. I, Sources and Effects of Ionizing Radiation, United Nations, New York, 2000.
8. C. CVETKOV, S. BACHVAROVA, M. JOREVA, A. ZLATEV, M. POJNAROVA, C. BINEVA, D. STANEVA, I. YORDANOVA, L. MISHEVA, Y. HRISTOZOVA, S. STOEV, V. MARINOV, Twenty Years after Chernobyl Accident, National Center for Agricultural Sciences, Sofia, 2006 (in Bulgarian).
9. Z. HOLGYE, E. SCHLESINGEROVA, J. TECL, R. FILGAS, J. Environ. Radioact., 71 (2004) 115.
10. I. AL HAMARNEH, A. WREIKAT, K. TOUKAN, J. Environ. Radioact., 67 (2003) 53.
11. A. AARKROG, L. BOTTER-JENSEN, Q. CHEN, J. CLAUSEN, H. DAHLGAARD, H. HANSEN, E. HOLM, B. LAURIDSEN, S. P. NIELSEN, M. STRANDBERG, J. SOGAARD-HANSEN, Environmental Radioactivity in Denmark 1992 and 1993, Riso National Laboratory, Roskilde, Denmark, Riso-R-756(EN), February 1995.
12. S. EHKEN, G. KIRCHNER, J. Environ. Radioact., 33 (1996) 147.
13. S. KROUGLOV, A. FILIPAS, R. ALEXAKHIN, N. ARKHIPOV, J. Environ. Radioact., 34 (1997) 267.
14. M. GASTBERGER, F. STEINHAUSLER, M. GERZABEK, A. HUBMER, J. Environ. Radioact., 54 (2001) 267.
15. S. FABRI, G. RIVA, R. SOGNI, G. FUSKONI, E. LUSARDI, G. BORASI, Health Phys., 66 (1994) 375.
16. Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Temperate Environments, IAEA, Technical Report Series, No. 364, IAEA, Vienna, 1994.
17. PENTSCHEV et al., Radioaktivitätsuntersuchungen an Donauwasser und Fischen im bulgarishen Donauabschnitt in der Zeit von 1964 bis 1974, Proc. Annual Meeting of the IAD, Vol. 1, Intern. Arbeitsgem. Donauforsch., 1975, p. 31.
18. E. J. MARINGER, Proc. Intern. Radiat. Protect. Assoc., IRPA 10, P-4b-259, May 2000.
19. G. I. E. PODEANU, Der Gehalt an Sr-90, Cs-137, H-3 und I-131 in Oberflachenwasser und Ablagerungen der Donau, Olt und Arges sowie in Milchproben für das Jahr 1972, Proc. Annual Meeting of the IAD, Vol. 1, Intern. Arbeitsgem. Donauforsch., 1975, p. 57.
20. N. STOKOZOV, K. BUESSELER, J. Environ. Radioact., 43 (1999) 173.
21. L. GEDEONOV, Z. GRITCHENKO, L. IVANOVA, T. ORLOVA, V. TISHKOV, V. TOPORKOV, V. PROKOPENKO, Atomnaya Energiya, 74 (1993) 58.
22. V. EGOROV, P. POVINEC, G. POLIKARPOV, N. STOKOZOV, S. GULIN, L. KULEBAKINA, I. OSVATH, J. Environ. Radioact., 43 (1999) 137.