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Radiation Contamination after the Chernobyl Nuclear Accident and the Effective Dose Received by the Population of Croatia

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ABSTRACT

Because of the Chernobyl nuclear accident which led to enhanced deposition of all fission products, contamination of the human environment in the Republic of Croatia was much higher than in the previous two decades. The paper deals with the investigation of deposition and contamination by fission product radionuclides (^{137}Cs and ^{90}Sr, in particular), especially within the human food chain. Its aim was to determine differences in contamination levels resulting from the Chernobyl accident and from large-scale atmospheric nuclear weapon tests. For the year following the Chernobyl accident, the radiation doses received from external and internal exposures were estimated for 1-year old infants, children at the age of 10-years and adults. The corresponding annual effective doses were 1·49, 0·93 and 0·83 mSv, respectively. The paper also gives data on the yearly intakes of ^{137}Cs and ^{90}Sr in foods and the corresponding effective doses received by the population of Croatia over many years from the global fallout following nuclear weapons testing and the Chernobyl accident.

INTRODUCTION

On 26 April, 1986, the most serious accident in the history of the nuclear industry occurred at the Chernobyl nuclear power plant in Ukraine, near the present borders of Belarus and Russia. As a result of the accident, the reactor was destroyed and, over the ensuing 10 days, large quantities of radionuclides were ejected into the environment. Today, 11 years later, the details of the accident have been well documented, and the total activity of radioactive material that was released is estimated to be around 12×10^{18} Bq, including some $6–7 \times 10^{18}$ Bq of noble gases (WHO, CEC, IAEA, 1996).

Due to fires in the graphite moderator, radioactive material was released up to the troposphere and subsequently dispersed by atmospheric meteorological processes. Finally, it was deposited on the surface of the earth, where it was measurable over the whole northern hemisphere. Specific features of the accident (duration of the release of radioactive products into the atmosphere, change in physical and chemical conditions) and changes in meteorological conditions caused non-uniform local contamination, both from the point of view of fallout density and radionuclide composition. Doses caused by the accident to the population in various countries of the northern hemisphere were assessed by the United Nations Scientific Committee on the Effects of Atomic Radiation, including average doses to various countries (UNSCEAR, 1988).

In the Republic of Croatia, intensive and systematic measurements of environmental samples and the human food chains started on April 30, 1986 when an increase in radioactivity caused by the nuclear accident at the Chernobyl reactor was first recorded. The primary aim of these measurements was to evaluate whether the concentrations of radionuclides detected in the environment reached levels that might be harmful to the population. The secondary aim was to distinguish differences in contamination levels, especially in the human food chain, resulting from large-scale atmospheric nuclear weapon tests and the Chernobyl nuclear accident.

In 1986, the year of the Chernobyl accident, and in the following years, investigations were focused on the long-lived radionuclides, ^{137}Cs and ^{90}Sr, in particular.

MATERIALS AND METHODS

Continuous measurements of radionuclide activity concentrations have been carried out by the Department of Radiation Protection of the Institute for Medical Research and Occupational Health since 1959. These measurements involved samples of fallout, air, soil, human foodstuffs and animal feed, bones, drinking, sea and surface waters, usually compressing several hundred items per year and more than one thousand in the year of the Chernobyl accident (Popović, 1963–1978; Bauman *et al.*, 1979–1993; Kovač *et al.*, 1994–1997).

Caesium (^{137}Cs and ^{134}Cs), iodine (^{131}I) and other gamma emitters were measured by a gamma ray spectrometry system based on a low-level ORTEC Ge(Li) detector (relative efficiency 15·4% and resolution, FWHM 1·82 keV at 1·33 MeV) coupled to a computerized data acquisition system (4096-channel pulse height analyzer and personal computer). Samples were measured in cylindrical plastic containers and Marinelli beakers of appropriate volume which were placed directly on the detector. Counting time depends on sample activity, but was never less than 60 000 s and was typically 80 000 s. As the minimum detectable activity depends on geometry, sample volume as well as counting time, it varied, for ^{137}Cs and a counting time of 80 000 s, from 0·068 $\pm$ 0·009 mBq l^{-1} for milk and 0·113 $\pm$ 0·015 Bq kg^{-1} for grain to 3·38 $\pm$ 0·67 Bq m^{-3} for water samples.

For strontium determination, radiochemical methods were used (Brayant *et al.*, 1966) and ^{90}Sr radioactivity was determined by beta counting its decay product, ^{90}Y, in a low-background anti-coincidence counter with efficiency of 27%. Average background was 0·023 $\pm$ 0·007 cps. For a counting time of 80 000 s the lower limit of detection was 0·0025 cps.

An efficiency calibration was carried out using sources provided by the International Atomic Energy Agency (IAEA) and the World Health Organization (WHO). Intercalibration was also performed on samples provided by the IAEA and WHO as part of their international intercalibration programmes.

To calculate radiation doses received by the population of Croatia, statistical data from the Federal Bureau of Statistics (FBS, 1970–1989) on dietary habits were used. As the food consumption rates did not change significantly over the period of the measurement programme, the following input data were used in calculations for adult persons: 176 kg of grain, 118 kg of vegetables, 66 kg of fruit, 101 l of milk and 50 kg of meat. For 1 year old infants and 10 year old children, correction factors of 0·3 and 0·7, respectively, were used to calculate the food consumption of grain, vegetables, fruit and meat, with a factor of 2·5 and 1·8, respectively, for milk.

The estimates of external exposure are based on measurements of external dose rates performed at several locations in the Republic of Croatia. An occupancy factor of 0·7 was used in the calculations due to the large proportion of the rural population that spends much time outdoors. The sheltering factor for buildings was 0·1 The physiological parameters and dose conversion factors were taken from the data recommended by the International Atomic Energy Agency, International Commission on Radiological Protection and World Health Organization (IAEA, 1986; ICRP 1989; WHO, 1988).

RESULT AND DISCUSSION

Gamma-spectrometric analyses of fallout were carried out in the Republic of Croatia every year on monthly samples collected in the areas of Zagreb (northwest Croatia), Zadar (south Croatia-Adriatic region) and Osijek (east Croatia). In the period immediately following the Chernobyl nuclear accident, significant activities of the following radionuclides were recorded in fallout: ^{140}Ba, ^{140}La, ^{141}Ce, ^{144}Ce, ^{95}Zr, ^{95}Nb, ^{103}Ru, ^{106}Ru, ^{106}Rh, ^{99m}Tc, ^{132}Te, ^{134}Cs, ^{137}Cs, ^{89}Sr, ^{90}Sr and ^{131}I. In addition to the short lived ^{131}I, particular attention was directed on the biologically significant, long-lived radioisotopes of caesium and strontium, which owing to their characteristics, may enter foodstuffs and thus become sources of significant internal radiation exposure. In 1986, the year of the Chernobyl accident, the results of the fallout analyses for the area of Zagreb showed highest deposition rates of these biologically important radionuclides as follows: ^{131}I, 3·1 kBq m^{-2}; ^{134}Cs, 3·1 kBq m^{-2}; ^{137}Cs, 6·3 kBq m^{-2} and ^{90}Sr 211 Bq m^{-2} (Lokobauer, 1988).

The data on ^{137}Cs deposition in the Zagreb area resulting from nuclear weapon tests and the Chernobyl accident are given in Fig. 1. Thirty years of investigation of ^{137}Cs deposition in the Zagreb area allow us to conclude that, in the year of the Chernobyl accident, the total deposition of ^{137}Cs exceeded the value recorded in the mid-1960s, after the period of the most intensive atmospheric nuclear tests. Deposition of 6·3 kBq m^{-2} ^{137}Cs measured in 1986 was approximately 4130 times higher than that of the

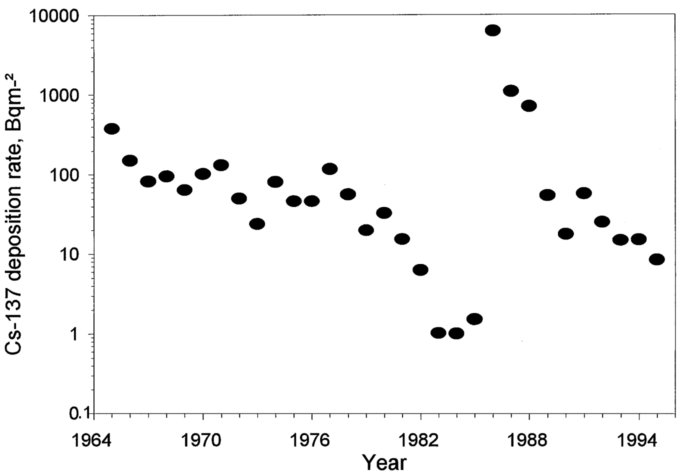

Fig. 1. ^{137}Cs in fallout in the area of Zagreb.

previous year, and approximately 3000 times higher than that measured in 1995.

Due to the low volatility of strontium, the nuclear accident at Chernobyl has not caused a significant increase in ^{90}Sr activity concentrations in the environment of the Republic of Croatia, the exceptions being in May and June 1986. This anomaly was occurred especially in the Adriatic region because of the prevailing meteorological conditions at that time, contaminated air plumes not passing over middle Adriatic regions (UNEP, 1991). In addition, the late spring and early summer of 1986 were very dry in the east Adriatic region (only 5 mm of precipitation was recorded in May 1986, while the 1963–1990 May average was 50 mm) leading to very low radioactive contamination of the Adriatic region (Franić and Bauman, 1993). Figure 2 gives the data on ^{90}Sr deposition for Zagreb and Zadar. However, most of the ^{90}Sr has been deposited immediately after the Chernobyl accident, i.e. in May 1986.

In the area of Zagreb, considering that the last announced major nuclear weapons test in the atmosphere took place in October 1980 (UNSCEAR, 1988), a ^{90}Sr deposition rate of 6·9 Bq m^{-2} in 1985 was 30 times lower than the ^{90}Sr deposition rate in the year of the Chernobyl accident. The total ^{90}Sr deposition rate in the area of Zagreb in 1986 was found to be approximately the same as that recorded in the middle of the 1960s.

Apart from ^{137}Cs and ^{90}Sr, significant amounts of ^{134}Cs and ^{131}I were also deposited in Croatia in May 1986. In the Zagreb area, the measured

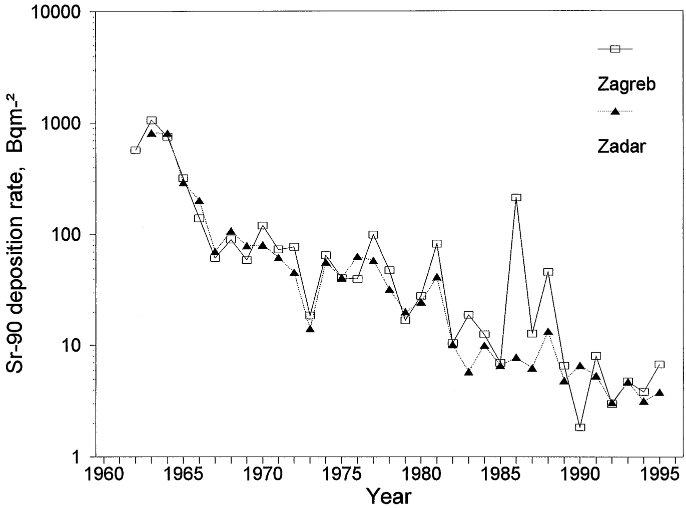

Fig. 2. ^{90}Sr in fallout in the areas of Zagreb and Zadar.

deposition rates were 3·1 kBq m^{-2} for ^{134}Cs and 3·1 kBq m^{-2} for ^{131}I. Before the Chernobyl nuclear accident, these two radionuclides were not present in the Croatian environment, and after the accident, ^{131}I was present for approximately two months, while ^{134}Cs was still found in some samples in the 1990s, although in very small amounts.

Systematic radioactivity measurements in the environment and in the human food chain in the Republic of Croatia were carried out in the year following the Chernobyl accident to estimate the radiation doses received from external exposure, inhalation and ingestion pathways. Special attention was paid to milk activities, as milk is a sensitive indicator for the presence of fission-derived radioactive contamination in the environment (Bauman *et al*., 1979–1993; Kovač *et al*., 1994–1997, Franić et al., 1998). In 1985, ^{90}Sr and ^{137}Cs average annual activity concentrations in milk collected in the Zagreb area were 0·15 and 0·05 Bq l^{-1}, respectively. However, in May 1986, as a consequence of the Chernobyl accident, activity concentrations increased to 1·5 Bq l^{-1} for ^{90}Sr and 46·0 Bq l^{-1} for ^{137}Cs, exponentially decreasing ever since, in 1996 being only 0·1 and 0·2 Bq l^{-1}, respectively.

The assessment of the annual effective dose for 1 year old infants, children at the age of 10 year and for adults (received from all pathways of exposure) are given in Table 1. Table 2 shows the distribution of the doses received (in percentages) in relation to the total annual effective dose, via pathways of exposure depending on different living and dietary habits within the investigated population age groups.

According to the estimates obtained, for all three age groups radiation doses due to inhalation could be neglected compared to external exposure and ingestion. In the year following the Chernobyl nuclear accident, 1 year old infants received the highest radiation dose. In this age group, 70% of the received dose (1·074 mSv) was attributed to the intake of ^{131}I by milk and leafy vegetables contaminated during May 1996. Ten-year old children

TABLE 1

Assessment of the Annual Effective Dose After the Chernobyl Accident for 1 Year Old Infants, Children at the Age of 10 Year and the Adult Population in the Republic of Croatia

	Effective dose (mSv)
Infants (1 year)	1·49
Children (10 year)	0·93
Adults	0·86

TABLE 2

The Assessed Distribution of Doses (in %) Received by Different Pathways of Exposure in Relation to the Total Annual Effective Dose for the Investigated Groups of the Population

Pathway of exposure	*Infants 1 year (%)*	*Children 10 year (%)*	*Adults (%)*
External exposure	17·0	51·3	55·0
Inhalation	0·4	1·5	1·3
Ingestion	82·6	47·2	43·7

TABLE 3

Estimates of the Yearly Intake of ^{137}Cs and ^{90}Sr in Foods by the Adult Population of the Republic of Croatia

Year	*Yearly intake (Bq)*	
	^{137}Cs	^{90}Sr
1964	17 857	3583
1965	1714	406
1970	214	201
1981	86	120
1985	36	71
1986	7786	182
1990	138	99
1991	123	76
1992	141	66
1993	119	71
1994	130	81
1995	88	78

received approximately corresponding doses through external exposure and ingestion, whereas the adult population received a slightly higher dose through external exposure.

Shortly after the Chernobyl nuclear accident, the '*critical radionuclide*' was ^{131}I, the '*critical pathway*' was the direct deposition of ^{131}I on leafy vegetables and pastures and the '*critical population*' was 1 year old infants.

In the years following the Chernobyl accident, particular attention was given to ^{137}Cs and ^{90}Sr transfer into the food chain. On the basis of all the data on the contamination of the most significant food components, estimates of ^{137}Cs and ^{90}Sr yearly intakes in foods are given in Table 3 for more than 30 years, covering both the global fallout following nuclear weapons tests and the Chernobyl accident. The assessment of the annual effective doses due to ^{137}Cs and ^{90}Sr ingestion is given in Figs 3 and 4. The results show that the risk to the population of Croatia from ^{137}Cs and ^{90}Sr ingestion was highest in the 1960s due to the intensive atmospheric nuclear weapons tests and in 1986, the year of the Chernobyl accident. However, it should be noted that in 1986 ^{137}Cs was the dominant contributor to the total dose.

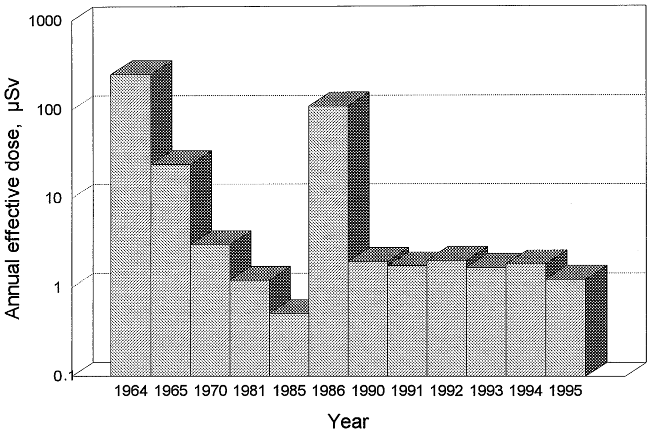

Fig. 3. Annual effective dose received from ^{137}Cs intake via food.

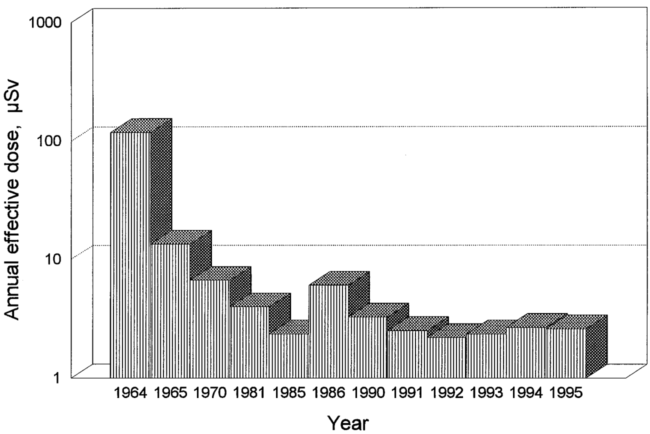

Fig. 4. Annual effective dose received from ^{90}Sr intake via food.

A continuous thirty-five-year investigation of ^{137}Cs and ^{90}Sr levels in foodstuffs in the Republic of Croatia has indicated that, after the Chernobyl accident, contamination by ^{137}Cs was approximately the same as that recorded in the middle of the 1960s immediately after the period of intensive atmospheric nuclear weapons testing whereas contamination by ^{90}Sr was about the same as that recorded in the 1970s.

Although in the year of the Chernobyl accident the ^{137}Cs levels in the environment were much higher than those of ^{90}Sr, since ^{90}Sr transfer from soil to the food chain is considerably more efficient than for ^{137}Cs, in the following years the doses to the population of Croatia from ^{137}Cs and ^{90}Sr ingestion were approximately the same.

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