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Cesium-137 activity concentrations in soil and brick samples of Mirpur, Azad Kashmir; Pakistan

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ABSTRACT

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Background: The aim of this study was to measure 137 Cs levels in soil and brick samples to set the baseline data and an indicator of fallout radioactivity for this part of the world. This study will help to probe, if there is any health threat posed by ¹³⁷Cs exposure to the residents of the area. *Materials and* Methods: Twenty five samples of soil and 6 samples of bricks were collected from different locations of Mirpur of Azad Kashmir. A P-type HPGe detector was used to measure gamma spectra of samples. Results: Results showed that for soil samples activity concentration range from 0.076±0.071 to 2.94±0.17 Bq.Kg⁻¹ with average value of 1.39±0.17 Bq.Kg⁻¹. Similarly activity concentration of ¹³⁷Cs in Brick samples range from 0.22±0.09 BqKg⁻¹ to 2.14±0.13 BqKg⁻¹ with average value of 0.73±0.10 Bq.Kg⁻¹. For soil samples the average values of outdoor, indoor and annual effective dose were found to be 5.12×10^{-5} , 20.47×10^{-5} and 25.58×10^{-5} mSv.y $^{-1}$ respectively. For brick samples specific activity of the ¹³⁷Cs range from 0.22±0.09 to 2.14±0.13 Bq Kg⁻¹. The average values of outdoor, indoor and annual effective dose for brick samples were 2.71×10^{-5} , 10.52×10^{-5} and 13.23×10^{-5} mSv.y⁻¹ respectively. Conclusion: The results indicate that the measured values were extremely small when compared with the annual dose rate recommended by ICRP (1.0 mSv.y⁻¹), as well as the annual external gamma radiation dose (0.48 mSv.y⁻¹) received per head from the natural sources of radiation.

Keywords: Background radiations, fallout radionuclide, annual effective dose, gamma radiation dose, Mirpur, Azad Kashmir.

INTRODUCTION

Ionizing radiations are the part of human environment from the time of earth formation. These radiations can be divided in to three groups according to their origin; (1) radiations coming from primordial radionuclides with long half-lives along with their progenies; (2) radiations coming from other sources than by the decay of primordial radionuclides; and (3) radiations coming from manmade sources(2). Once the *detrimental* effects associated with radiations have been identified, since than this subject has got considerable importance. Several studies have been conducted throughout the world for monitoring and identification of

health effects linked with radiation exposure.

An extensive data related with monitoring of radioactivity originated from primordial and man-made radionuclides (3-17) is available in the literature. Α significant proportion environmental radioactive contamination has been resulted from past nuclear power plant accidents, nuclear weapons testing, nuclear waste disposal and embezzlement of radioactive sources (18). After the Chernobyl nuclear power plant accident (on 26th of April 1986) radioactive contaminations has got the public attention. This accident has not only affected Ukraine, Belarus and Russia but the whole world and changed the public and government attitudes to nuclear safety on an international scale (19).

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The most important anthropogenic radionuclides widely distributed in global fallout due to nuclear weapon testing and nuclear power plants accidents are ⁹⁰Sr and ¹³⁷Cs. ⁹⁰Sr with half-life of 28.78 years dowries a significant health threat since it replaces for calcium in bone ⁽²⁰⁾. On the other hand ¹³⁷Cs with half-life of about 30.17 years reacts with water producing cesium hydroxide ⁽²¹⁾.

137Cs exposure is of much concern for human health. Energy released by radioactive ¹³⁷Cs can cause a substantial damage to living cells. Since ¹³⁷Cs emits beta and gamma particles which may damage to tissue and cellular functions due to ionization of molecules with in cells of body. Most prominent health threat route due to exposure of ¹³⁷Cs is the ingestion of radioactive cesium-contaminated food (US DHHS 2001). In case of fallout from nuclear weapons tests, or from reactor accidents, milk is an important source of ¹³⁷Cs. Another route of radioactivity ¹³⁷Cs exposure for human is through the entrance in plants by direct deposition or uptake from the soil.

¹³⁷Cs uptake from soil has been studied in the 1960s ⁽²²⁾ and to observe long-term effects of the Chernobyl accident there has been renewed interest in the subject. ¹³⁷Cs uptake is very much reliant on the type of soil. Within plants ¹³⁷Cs is more movable as compared to ⁹⁰Sr. It moves readily from leaves to seeds within plants ⁽²³⁾. Besides these routes of exposures Inhalation and dermal exposure routes may also present a significant health threat.

This study aims at the measurement of ¹³⁷Cs levels in soil and brick samples, in order to set the baseline data for this part of world. ¹³⁷Cs presence in soil and brick samples will be the indicator of fallout radioactivity in the area under investigation. Results will be compared with the standards set by international organizations in order to assess the level of threat posed by ¹³⁷Cs exposure to local population.

Area under study

Current study has been carried out for the soil and brick samples collected from the district Mirpur of Azad Kashmir. Mirpur is the largest city of Azad Kashmir, and it is located at the extreme south of Azad Kashmir at an elevation of 459 meters (1509ft). coordinates are 33°9'4" N 73°44'10" E / 33.1425; 73.75611. District Mirpur is the industrial hub of Azad Kashmir, having industries of Foam, Polypropylene, Synthetic yarn, Motorbikes, Textile, vegetable ghee, logging and sawmills, soap, cosmetics, marble, ready-made garments, matches, rosin etc. Along with all these there is a huge investment in the field of construction building materials. brick factories. industries include Marble factories and crush/concrete factories etc.

MATERIALS AND METHODS

Sampling procedures

Thirty one samples of soil and bricks were collected from different places of the district Mirpur in the month of June, 2010. These included 25 samples of soil and 6 samples of bricks. The samples were initially packed in polythene bags and properly catalogued in accordance with the location of sampling site, brought to the Physics laboratory of University of Azad Jammu & Kashmir for processing before analysis. The collected samples were crushed. sieved and dried at 110 ±1 °C for 4 to 6 hours. These samples were then placed in Marinelli beakers and hermitically sealed. In order to attain equilibrium between 226Ra and 222Rn, beakers were placed in an undisturbed position for a period of 60 days.

Gamma Spectrometry and Analysis

A P-type HPGe detector was used to measure gamma spectra of soil and brick samples. The resolution and relative efficiency of the detector for 1332 keV (60Co) was 2.5 keV and 90 %, respectively. In order to perform the data acquisition, the packed soil and brick samples were placed inside the lead castle in which detector is present. The Marinelli beaker were placed on the detector so that detector occupies

the hollow region of the beaker and detector was covered from the material sample by all its dimensions. The samples are in this way exposed to the detector for sixteen hours (57600 seconds) individually.

Initially the detector was calibrated so that the energy of each peak should be in accordance with corresponding channel number. Here in this study the reference material used for the calibration was mix Radionuclide Gamma Reference Source, supplied by AEA technology QSA GmH Germany. The calibration was made for the naturally occurring radio nuclides of uranium series, thorium series, cesium and potassium respectively.

¹³⁷Cs concentration was measured directly through its gamma-ray energy peak of 661.66 keV. The software Personnel Computer Analyzer (PCA-II) was used for the collection of the spectra. The lowest limit of detection (LLD) for ¹³⁷Cs was found as 0.12 Bq.Kg⁻¹. LLD was calculated using formula.

$$LLD = \frac{4.66 \times \sqrt{BackGround\ Counts}}{Live\ Time \times Yield \times Efficiency}$$
(1)

For the anthropogenic nuclide ¹³⁷Cs, the formula of dose rate calculation was used as below:

$$\dot{D}_{cs} = 0.03 \times A_{cs} \tag{2}$$

Here " A_{Cs} " is the specific activity of the ¹³⁷Cs and 0.030 nGyh⁻¹ per BqKg⁻¹ is the dose conversion factor for the ¹³⁷Cs and it should be noted that dose rate is expressed in the units of nGyh⁻¹.

Effective dose rates

The effective dose of gamma radiation received annually by an individual depends upon the time for which the person is exposed to the gamma radiations. According to a report prepared by UNSCEAR, 2000 it is assumed that the populations of urban locations spend eighty percent of their time of the solar day in indoor environment whereas twenty percent of the time is spent outdoors. Thus for the indoor exposure of the public the occupancy factor is

0.8 and for the outdoor radiation exposure the outdoor occupancy factor is 0.2 as indicated in the UNSCEAR, 2000 $^{(1)}$. Therefore to estimate the annual effective dose for an individual a conversion factor (Q = 0.7 SvGy-1) is used to cover the absorbed dose in air to the annual effective dose $^{(24)}$.

Thus the annual effective dose has been calculated by following relation:

$$E = Q \times T \times O \times \dot{D} \times 10^{-6} \tag{3}$$

Where the term on left side of the above equation represents annual effective dose equivalent in mSvy-1, "T" is the time in hours for one year, i.e. 8760 hours, "D" is the dose rate in nGyh-1 and "O" is the occupancy factor for outdoor and indoor circumstances. For indoor exposure occupancy factor is taken as 0.8 and for outdoor 0.2 (UNSCEAR 2000) (1).

RESULTS ANS DISCUSSION

The data obtained for 137Cs in soil and brick samples collected from Mirpur, indicates significant variability. For soil samples, ¹³⁷Cs activity levels vary from 0.076±0.071 to 2.94±0.17 Bq. Kg⁻¹ (as may be seen from table 1) with average value of 1.39±0.17 Bq. Kg⁻¹. Similarly dose rates measured for corresponding values of specific activities ranged from (0.2 to 8.8) \times 10⁻² nGy h⁻¹ and with average value of 4.2 nGy h-1 (see table 1). It can be seen from the table 1 that specific activity and dose rate due to ¹³⁷Cs are lowest for the soil sample #20, collected from the bank of Mangla dam Mirpur. The maximum value of specific activity and dose rate are associated with the soil sample # 12, collected from Wespa factory Mirpur. Table 2 shows the outdoor, indoor and annual effective dose equivalent which is received by an individual from the anthropogenic radionuclide ¹³⁷Cs. Results shows that the outdoor, indoor and annual effective dose received from the soil of Mangla dam Mirpur is minimum with the values of 0.25×10^{-5} , 0.98×10^{-5} and 1.23×10^{-5} mSv.y-1 respectively, whilst the maximum values

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of outdoor, indoor and annual effective dose (10.79, 43.17 and 53.96) \times 10⁻⁵ mSv. y⁻¹ were recorded for samples collected from Wespa Factory Mirpur. The average values of outdoor, indoor and annual effective dose are (5.12, 20.47 and 25.58) \times 10⁻⁵ mSv.y⁻¹ respectively.

Spectral analysis of bricks samples used in Mirpur Azad Kashmir also showed the presence of the ¹³⁷Cs in the bricks. The specific activity and dose rate are described in the table 3. The table shows that the specific activity of the ¹³⁷Cs has a minimum value of 0.22±0.09 BqKg-1 for the bricks of trademark A.A.P Mirpur which delivers a minimum dose rate of 0.01nGyh-1.

The maximum value of specific activity of ¹³⁷Cs and dose rate was found in bricks with trademark M.A. Mirpur whose values are 2.14±0.13 BqKg⁻¹ and 0.06 nGyh⁻¹ respectively.

Results (see table 4) shows that the outdoor, indoor and annual effective dose received from the bricks. Minimum values for outdoor, indoor and annual effective dose were found as with the values of 0.81 c, 3.24×10^{-5} and 4.05×10^{-5} mSv.y $^{-1}$ respectively, whilst the maximum values of outdoor, indoor and annual effective dose (7.87, 30 and 30.87) \times 10^{-5} mSv. y^{-1} , were recorded for Brick samples with trademark name M. A Mirpur. The average values of outdoor, indoor

Table 1. Specific activity and dose rate due to ¹³⁷Cs in soil of Mirpur Azad Kashmir.

Sample Code	Location	Activity of ¹³⁷ Cs	Dose rate
		A _{Cs} (BqKg ⁻¹)	$\overset{ullet}{D}$ (× 10 $^{ ext{-2}}$) (nGyh $^{ ext{-1}}$)
Soil-1	Industrial area Mirpur	Below L.L.D ^a	N.D ^b
Soil-2	Bohar colony Mirpur	0.55±0.12	1.7
Soil-3	MUST Mirpur	2.93±0.21	8.8
Soil-4	Naangi Mirpur	0.33±0.08	1
Soil-5	Bailyan Mirpur	0.96±0.15	2.9
Soil-6	NAB office Fazalabad	1.45±0.17	4.4
Soil-7	Chaichyan-1	0.69±0.10	2.1
Soil-8	Khari Shareef-1	1.20±0.16	3.6
Soil-9	Nagyal Mirpur	0.38±0.09	1.1
Soil-10	Jatlaan Mirpur	2.08±0.20	6.2
Soil-11	Jordiyan Mirpur	2.13±0.20	6.4
Soil-12	Wespa Factory -1	2.94±0.21	8.8
Soil-13	Kotli Road Mirpur	1.74±0.17	5.2
Soil-14	MUST Mirpur	2.56±0.21	7.7
Soil-15	Khalaqabad Mirpur	Below L.L.D	N.D
Soil-16	Tarpa stop Mirpur	1.42±0.17	4.3
Soil-17	New city Mirpur	Below L.L.D	N.D
Soil-18	Kotli Road Mirpur	0.87±0.14	2.6
Soil-19	Industrial area Mirpur	1.53±0.16	4.6
Soil-20	Bank of Mangla Dam Mirpur	0.076±0.071	0.2
Soil-21	Dahola	2.2±0.18	6.6
Soil-22	Mangla Fort & Museum	2.4±0.19	7.2
Soil-23	Wespa Factory -2	1.94±0.17	5.8
Soil-24	Chaichyan-2	2.46±0.19	7.4
Soil-25	Khari Shareef-2	1.81±0.18	5.7
Over all mean	value (BqKg ⁻¹)	1.39±0.17	4.2

^aBelow LLD=Below lowest limit of detection

^bN.D= Not detected

and annual effective dose are (2.71, 10.52 and 13.23) $\times\,10^{\text{-}5}$ mSv. $y^{\text{-}1}$ respectively.

At some sampling points the activity levels of ¹³⁷Cs in soils samples was below the detection limit and mean level of activity for other sampling positions was found as average value of 1.39±0.17 Bq. Kg⁻¹. This activity might be due to anthropogenic radionuclides fallout of previous worldwide nuclear explosion and nuclear reactor accidents. Due to the consequences of nuclear activities, the radionuclide ¹³⁷Cs enters into the atmosphere and most of the fallout radiation accumulates in the soil.

Table 3. Specific activity of ¹³⁷Cs and dose rate associated with bricks used in Mirpur Azad Kashmir.

Sample code	Location	Activity of 137 Cs A _{Cs} (BqKg ⁻¹)	Dose rate D ⁰ (nGyh ⁻¹)
B-1	M.S Mirpur	0.72±0.12	0.02
B-2	R.Y Mirpur	0.66±0.12	0.02
B-3	M.A Mirpur	2.14±0.13	0.06
B-4	A.A.P Mirpur	0.22±0.09	0.01
B-5	A.G Mirpur	0.40±0.09	0.01
B-6	W.Z Mirpur	0.29±0.07	0.01
Over all m	iean value	0.73±0.10	0.02

Table 2. Annual effective dose equivalent, outdoor and indoor average annual effective dose equivalent in soil samples used of Mirpur Azad Kashmir for ¹³⁷Cs.

Sample code	Sample location	Annual effective dose equivalent E (mSvy ⁻¹) × 10 ⁻⁵	Outdoor average annual effective dose E_{out} (mSvy $^{-1}$) × 10^{-5}	Indoor average annual effective dose E_{in} (mSvy $^{\text{-1}}$) × $10^{\text{-5}}$
Soil-1	Industrial area	N.D	N.D	N.D
Soil-2	Bohar colony	10.42	2.08	8.34
Soil-3	MUST-1	53.96	10.79	43.17
Soil-4	Naangi	6.14	1.23	4.91
Soil-5	Bailyan	17.79	3.56	14.23
Soil-6	NAB office Fazalabad	26.98	5.4	21.58
Soil-7	Chaichyan	12.88	2.58	10.3
Soil-8	Khari Shareef	22.08	4.42	17.66
Soil-9	Nagyal	6.75	1.35	5.4
Soil-10	Jatlaan	38.01	7.6	30.41
Soil-11	Jordiyan	39.25	7.85	31.4
Soil-12	Wespa Factory	53.96	10.79	43.17
Soil-13	Kotli Road	31.89	6.38	25.51
Soil-14	MUST-2	47.21	9.44	37.77
Soil-15	Khalaqabad	N.D	N.D	N.D
Soil-16	Tarpa stop	26.36	5.27	21.09
Soil-17	New city	N.D	N.D	N.D
Soil-18	Kotli Road	15.94	3.19	12.75
Soil-19	Industrial area	28.21	5.64	22.57
Soil-20	Bank of Mangla Dam	1.23	0.25	0.98
Soil-21	Dahola	40.47	8.09	32.38
Soil-22	Mangla Fort & Museum	44.15	8.83	35.32
Soil-23	Wespa Factory -2	35.56	7.11	28.45
Soil-24	Chaichyan-2	45.38	9.08	36.3
Soil-25	Khari Shareef-2	34.95	6.99	27.96
Over all N	Mean value	25.58	5.12	20.47

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Table 4. Annual effective dose equivalent, outdoor and indoor average annual effective dose equivalent due to ¹³⁷Cs in bricks samples used in Mirpur.

Sample code	Location	Annual effective dose equivalent	Outdoor average annual effective dose	Indoor average annual effective dose
		E (mSvy ⁻¹) × 10 ⁻⁵	E_{out} (mSvy ⁻¹) × 10 ⁻⁵	E_{in} (mSvy ⁻¹) × 10 ⁻⁵
B-1	M.S Mirpur	12.65	2.65	10
B-2	R.Y Mirpur	12.14	2.43	9.71
B-3	M.A Mirpur	37.87	7.87	30
B-4	A.A.P Mirpur	4.05	0.81	3.24
B-5	A.G Mirpur	7.36	1.47	5.89
B-6	W.Z Mirpur	5.34	1.07	4.27
Over all mean value		13.23	2.71	10.52

Above scatter graph shows a correlation the data obtained between for concentration with the year in which study is undertaken. A linear relationship is evident from the regression plot. From the graph a decrease in the values of ¹³⁷Cs concentration can be seen. Maximum value has been recorded by Hassan et al. (3) carrying his study for Charsaddah district of Pakistan in 1994. The regression equation, y = -0.2922x + 589.48, represnts least square line drawn through the scatter points. The coefficient R2 also reffered as co-efficient of determinations measures fit of regression line to the data. Value obtain for R²= 0.8115 shows strong correlation between year measurement with concentration of ¹³⁷Cs.

Here we cannot predict exactly about the source of ¹³⁷Cs in Mirpur, Azad Kashmir other than past fallout deposition from nuclear testing or reactor accident. As may be seen from figure

1 a decreasing activity concentration of ¹³⁷Cs trend can be due to no more significant reactor accident or atmospheric weapon testing have been done since major nuclear reactor accident of Chernobyl in 1986. ¹³⁷Cs activity concentration found in soil and brick samples may be attributed due to global fallout (Chernobyl fallout).

In this study, the observed $^{137}\mathrm{Cs}$ concentrations are consistent with the world average.

¹³⁷Cs activities (for soil samples, mean value 1.39±0.17 Bq. Kg⁻¹) found for current study are consistent with values reported for other parts of Pakistan. For example the average value of activity concentration of ¹³⁷Cs in barren and cultivated soils of Pakka Anna was found as 3.6 (3.57-3.63) Bq kg⁻¹ and 4 (1.98-5.42) Bq kg⁻¹, respectively ⁽¹³⁾. For Southern part of Punjab, Pakistan, mean value of ¹³⁷Cs for soil samples

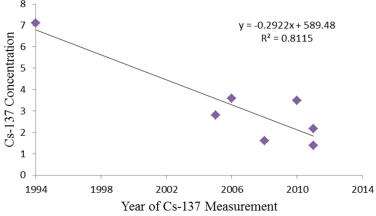


Figure 1. Correlation between the data obtained for ¹³⁷Cs concentration with the year of measurement.

was reported as 1.6 Bq. Kg^{-1} (14) and for Mid-Rechna, Pakistan the mean values is found as 3.5 Bq. Kg^{-1} (15). For some districts of Punjab Province Pakistan, mean value of ¹³⁷Cs was found as 2.18 Bq. Kg^{-1} (16) and for Charsaddah mean value of ¹³⁷Cs activity is reported as 7.1±2.0 Bq. Kg^{-1} (3).

Comparison of current study results with the data available in literature for other countries has also been made in table 5. As may be seen the table the range of activity concentration of $^{137}\mathrm{Cs}$ is much smaller than the values reported for Venezuela, 5 Bq. Kg-1 (25), Bangladesh 7 Bq. Kg-1 (26). Majorca (Spain) (10-60 Bq. Kg⁻¹) (4), Inshass, Cairo (Egypt) (1.6–19.1 Bq. Kg⁻¹) (7), Algeria (15–35 Bq. Kg⁻¹) (5), Kocaeli basin (Turkey) (2-25 Bq. Kg-1) (11), (USA) (5-58)Kg-1) Louisiana Bq. Montenegrin coast (Yugoslavia) (1.5-28.4 Bq. Kg^{-1}) (8), Sudan (0–18.5 Bq. Kg^{-1}) (27), North-western Libya (0.9-1.7 Bq. Kg-1) (9), Riyadh (Saudi Arabia) (0-2 Bq. Kg-1) (10), Northern Taiwan (1.48-27 Bq. Kg-1) (6), Ordu, Turkey (67.4 to 275.3 Bq kg⁻¹) (28).

The activity concentration and resulting doses due to $^{137}\mathrm{Cs}$ exposure are too small to add

to the total radiation exposure. The values of activity concentration and the estimated dose rates are towards the lower side when compared with that of other countries and standards set by different world organizations like ICRP and in UNSCEAR 2000 report (1).

CONCLUSIONS

Based on the obtained data, it is concluded that

- (i) Activity concentration of ¹³⁷Cs in soil samples is found to be in the range from 0.076±0.071 to 2.94±0.17 Bq. Kg⁻¹ with average value of 1.39±0.17 Bq. Kg⁻¹.
- (ii) Activity concentration of ¹³⁷Cs in Brick samples is found to be in the range from 0.22±0.09 BqKg⁻¹ to 2.14±0.13 BqKg⁻¹ with average value of 1.39±0.17 Bq. Kg⁻¹.
- (iii) Outdoor, indoor and annual effective dose received from the soil samples of Mangla dam Mirpur is minimum with the values of (0.25-10.79, 0.98-43.17 and 1.23-53.96) × 10⁻⁵ mSv.y⁻¹ respectively. The average values of outdoor, indoor and annual effective dose are

 Table 5. Comparison of current results with data available in literature.

Country	Concentration (Bq kg ⁻¹)	References
Ordu, Turkey	67.4 - 275.3	(28)
Venezuela	5	(25)
Bangladesh	7 (3-10)	(26)
Majorca (Spain)	10–60	(4)
Inshass, Cairo (Egypt)	1.6-19.1	(7)
Algeria	15–35	(5)
Louisiana (USA)	5–58	(11)
Montenegrin coast (Yugoslavia)	1.5-28.4	(8)
Sudan	0-18.5	(27)
North-western Libya	0.9-1.7	(9)
Riyadh (Saudi Arabia)	0–2	(10)
Northern Taiwan	1.48–27	(6)
Punjab province (Pakistan)	2.8±1 (1.1-5.3)	(12)
Pakka Anna, Pakistan	3.6(3.57-3.63)	(13)
Southern part of Punjab, Pakistan	1.6	(14)
Mid-Rechna, Pakistan	3.5	(15)
Punjab Province, Pakistan	2.18	(16)
Charsaddah, Pakistan	7.1±2.0	(3)
Mirpur Azad Kashmir	1.39±0.17	Present study

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- found as $(5.12, 20.47 \text{ and } 25.58) \times 10^{-5} \text{ mSv.}$ y^{-1} respectively. These measured values are relatively small when compared with the recommended annual dose rate limit of 1.0 mSv set by ICRP and action level for annual external gamma radiation dose $(0.48 \text{ mSv.} y^{-1})$ received per head from the natural sources of radiation recommended in UNSCEAR (2000) report (1).
- (iv) To conclude, it is expected that ¹³⁷Cs exposure to the local population will not pose any significant health threat.

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