Chapter

Radionuclide Contamination as a Risk Factor in Terrestrial Ecosystems: Occurrence, Biological Risk, and Strategies for Remediation and Detoxification

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Abstract

Radionuclide contamination poses serious hazards for terrestrial ecosystems. Beyond the readily apparent damage to the biota at high doses, low doses of ionizing radiation produce stochastic effects: mutation, carcinogenesis, and genomic instability. The proposed chapter is a review of the biological and ecological effects of radionuclides. The authors discuss, beyond the Chernobyl accident, other contamination events. The review includes the biological and ecological effects of the three principal technogenic contaminants in terrestrial ecosystems: Cs-137, Sr-90, and I-131. Ecological risks to terrestrial small mammals are assessed in detail. In addition, the chapter provides some of the lesser-known methods of remediation and detoxification, including the use of modified natural zeolites as environmental remedies and bio-sorbents. Presented herein is little-known information on environmental protection against radioactive contamination.

Keywords: radionuclides, radioecology, contamination, remediation, detoxification, zeolites

1. Introduction: the essence of radionuclides—emission types and biological effects

Radionuclides are unstable isotopes of different chemical elements. Usually, this instability is due to excess energy in the atomic nucleus, leading to the release of particles with different energies in a process called **radioactive decay**. Natural radionuclides emit three types of radiation: alpha (α), beta (β-), and gamma (γ). Of these types, α-particles have the strongest biological effects, causing 20 times more biological damage than an equivalent dose of β- or γ radiation [1, 2]. While α- and β-particles tend not to penetrate into matter, γ-radiation, especially at the higher end of the energy spectrum, penetrates deep into living and non-living matter. This means that, when considering the biological and ecological effects of radionuclide contamination, α- and β-emitters are only relevant if incorporated into living organisms. In contrast, γ-emitters are relevant as both internal and external components of the total absorbed dose. In the context of anthropogenic contamination, it needs to be taken into account that some of the man-made radionuclides emit other types of radiation. For example, radioisotopes used in medical PET scans such as ^{18}F, ^{11}C, ^{13}N, ^{15}O are positron (β^+) emitters. Other, more exotic man-made radionuclides such as Californium-252 (^{252}Cf) are capable of spontaneously emitting neutrons. Both positron and neutron emitters require specific equipment for handling and detection of the radiation sources [1]. Some radionuclides emit multiple types of particles. The anthropogenic radionuclide ^{137}Cs emits β^- particles at two energies: 511 and 1173 kiloelectronvolts (keV), and γ-rays at 32 and 661.6 keV [3, 4].

The biological effects of radionuclides are mainly due to the emitted ionizing radiation (IR). IR interacts with biomolecules directly by damaging them or indirectly—by producing reactive oxygen species (ROS), which in turn damages biomolecules. According to the paradigms of classical radiobiology, the principal target of IR on a cellular level is genomic DNA—it can be damaged directly or indirectly, leading to cell cycle arrest and an activation of DNA repair systems, followed by recovery, cell death, or mutagenesis [5, 6]. Sparsely ionizing radiations such as β- particles and γ-rays cause around 70% of DNA damage indirectly through ROS, while densely ionizing radiations, such as α-particles and high-energy cosmic particles, cause only about 30% of the biologically significant damage indirectly [7]. Researchers have elucidated the biological effects of high and medium doses of radiation. Nevertheless, biological effects at low doses remain insufficiently understood and a subject of much debate [1, 8]. Currently, radiation risk is extrapolated linearly to the low doses by using the **Linear Non-Threshold (LNT)** mathematical model [1, 9]. However, other hypotheses include **radiation hormesis**, which is the idea that small doses of radiation are beneficial [10], and **low-dose hypersensitivity**, which is the assumption that low doses of radiation are more mutagenic because they do not activate DNA repair systems [11]. While radiation hormesis has been well researched recently [10], it has still not been taken into account in radiation protection calculations, where every minimal dose of radiation is assumed to carry a small but non-negligible risk [12]. On the other hand, the low-dose hypersensitivity hypothesis is supported by recent studies, raising questions about the validity of current assumptions in radioprotection [13]. Living organisms tend to display different radiation sensitivity. Mammalian species are very sensitive to radiation, while insects tend to be comparatively radioresistant. The champion of radiation resistance is the bacterium *Deinococcus radiodurans*, which can withstand an acute dose of 5000 Gray with almost no loss of viability. Similarly, tardigrades can withstand 5000 Gray with 50% loss in viability (LD_{50} = 5000 Gy). For comparison, the LD_{50} for humans is around 6 Gray, for mice around 6.4 Gray, and for goats only around 2.4 Gray [14].

A significant concern in radionuclide-contaminated areas arises from the process of **bioaccumulation**. Similar to other chemical elements from their respective groups, radioisotopes are incorporated preferentially into different target organs and tissues. Thus, ^{137}Cs, a chemical analogue of potassium, is preferentially accumulated into nerve and muscle tissue. ^{90}Sr, an analogue of calcium, has a very strong affinity for bone and hematopoietic tissue. Some of the properties of the three most environmentally significant anthropogenic radionuclides are presented below (**Table 1**).

As evident from the table, the most significant environmental contaminants of the above are ^{137}Cs and ^{90}Sr due to their long half-lives and persistence in nature. ^{131}I was

Radionuclide	Symbol	Half-life (λ)	Emitted radiation	Target tissues and organs	Biological effects
Cesium-137	^{137}Cs	30.17 years	β-(511, 1173 keV), γ (32, 661.6 keV)	Nerve, muscle	Different cancers
Strontium-90	^{90}Sr	28.8 years	pure β-(546 keV)	Bone	Bone cancer, leukemia
Iodine-131	^{131}I	8.02 days	β-(333.8, 606.3 keV), γ (364.5, 636.9 keV)	Thyroid gland	Thyroid cancer

Table 1.
The most significant anthropogenic radionuclides and their biological effects (data adapted from [3, 4]).

only a very significant contaminant in the first year following the Chernobyl accident, causing ~4000 excess thyroid cancers in the most significantly affected populations of Russia, Belarus, and Ukraine [15].

2. Radionuclide occurrence in nature: natural and anthropogenic sources

Natural radioactivity, including external terrestrial γ radiation, internal α-, β-, and γ-radiation from naturally occurring radionuclides, cosmic radiation, and exposure to radon (^{222}Rn) and thoron (^{220}Rn) and their radioactive progeny molecules, accounts for ~95% of the annual radiation dose for the terrestrial biota [1]. The global annual dose for an average person is 3.6 millisieverts/year (mSv/a), of which 82% is due to natural radiation exposure, around 15% is due to medical exposure, and only about 0.8% is due to anthropogenic contamination of the environment. Natural radioactivity has been a subject of concern for decades. Globally, there are areas with increased natural radiation, often due to thorium (^{232}Th) deposits in the form of monazite rocks. Two such areas are Guarapari, Brazil, and the state of Kerala in southern India. The area of Ramsar, Iran, has enormously increased natural radioactivity due to radioactive hot springs containing ^{222}Rn and its progeny. Although annual doses in these areas reach an average of 35–40 mSv/a, compared to 3.6 mSv/a average in Europe and 2.5 mSv/a in Bulgaria, modern biomedical studies report no excess cancer risk, leading researchers to believe that a 10-fold increase in natural radioactivity is harmless [16].

In contrast, environmental contamination by anthropogenic radionuclides without doubt creates serious risks. The Chernobyl accident is the most prominent example of environmental damage due to technogenic sources, although it is not the only one; Chernobyl caused significant chronic morbidity and mortality in people and enormous damage to the environment and economies in Europe. This is mostly due to ^{131}I, ^{137}Cs, and ^{90}Sr, and their tendencies for **bioaccumulation** and **biomagnification** in terrestrial ecosystems [17]. Although the Chernobyl accident is the best-known example, there are many other significant contamination events in the period 1945–2011 (**Table 2**).

One aspect evident from the table is that, according to atmospheric radioactivity released, the Chernobyl accident exceeds all other INES scale 5–7 accidents combined. At the same time, during this accident, only about 30% of the core radioactivity was released, suggesting that a full-blown reactor explosion can cause even greater damage to the environment. Another noteworthy peculiarity is that most reactor accidents

Accident site, year	Country	INES scale	Date	Accident type	Radioactivity released to the atmosphere, PBq	Iodine-131 released, PBq	Cesium-137 released, PBq
Chernobyl, 1986	USSR	7	26.04.1986	Reactor meltdown	12,000	1760	85
Fukushima, 2011	Japan	7	11.03.2011	Reactor meltdown	630	<380	<37
Mayak (Chelyabinsk-40), 1957	USSR	6	29.09.1957	Nuclear waste explosion	1850	Not known	Not known
Chalk River, 1952	Canada	5	12.12.1952	Reactor meltdown	0.3	Not known	Not known
Windscale, 1957	UK	5	10.10.1957	Reactor fire	1.6	0.7	0.02
Simi Valley, 1959	USA	5	26.07.1959	Partial reactor meltdown	>200	Not known	Not known
Beloyarsk, 1977	USSR	5	1977	Partial reactor meltdown	not known	Not known	Not known
Three Mile Island, 1979	USA	5	28.03.1979	Partial reactor meltdown	1.6	<0.007	Not known

Table 2.
The most significant radioactive release accidents, their IAEA INES severity scale, and radioactivities released to the environment (data from [18]).

so far occurred either with new or experimental power plants (Chernobyl, Chalk River, Simi Valley, Beloyarsk) or military reactors (windscale). Nevertheless, the Fukushima accident in 2011 presents a new precedent—the reactors in the plant were old, nearing the end of their design life. Since this is true for many of the currently operating reactors, this presents a new, threatening perspective. Aging, crumbling nuclear infrastructure may present a new, unmitigated radiation hazard in the future.

3. Radionuclides and nature: significant risks and unknowns

Some of the risks to ecosystems posed by radionuclide contamination are well understood. They include, at high doses >1 Gray acute dose, teratogenesis in developing embryos, stunted plant growth, and visible damage to the flora and fauna. These are **deterministic effects**, and they occur definitely after exposure to strong doses of ionizing radiation and are dose dependent (**Figure 1**).

As shown in **Figure 1**, pine trees are very radiosensitive; they can serve as a bio-indicator of severe radioactive contamination at doses exceeding 3 Sv acute exposure [19]. The other depicted deterministic effect is teratogenesis in pregnant mammalian species. At doses exceeding 1 Sv acute *in utero* exposure, the number of resorbed fetuses decreases, and so does the number of offspring born with malformations [1].

Perhaps more worrying are the **stochastic effects**, which occur with a small probability even at low radiation doses. These include **radiation mutagenesis** and, as a consequence of it, **radiation carcinogenesis** [1, 12]. Based on data from experiments with specially bred laboratory mice and results from the radiobiological monitoring of humans, exposed to γ-rays and neutrons during the bombings of Hiroshima and Nagasaki, it is estimated that the doubling dose of radiation-induced mutagenesis is 1 Gy. This means that an acute exposure to 1 Gy of γ-rays doubles the spontaneously occurring rate of mutation [20, 21]. Nevertheless, this perspective is being challenged. For example, Belarussian researchers observed transmission of chromosomal damage in the progeny of wild rodents from the vicinity of Chernobyl, indicating **genomic instability** [22]. An international team observed a higher mini- and microsatellite mutation rate in the children of Chernobyl liquidators [23]. Both of these findings support the theory that even low doses of radiation can be harmful to the biota, as well as current and future generations of humans. Another, more recent venue of

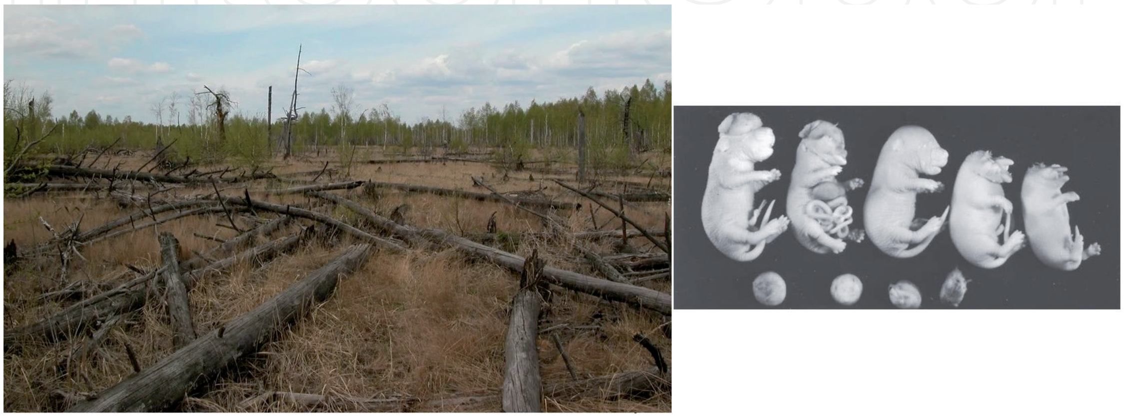

Figure 1.
Deterministic effects of ionizing radiation: Dead pine trees near Chernobyl, Ukraine in 1990 (left, taken from [19]), and experimental radiation teratogenesis in mouse embryos (right, photograph by Dr. Roberts Rugh, taken from [1]).

research with significant progress is the radiation-induced bystander effect (RIBE) phenomenon, in which non-irradiated cells show similar cytotoxicity and genetic damage to their irradiated neighbors [24, 25]). The results from bystander effect studies generally support the theory of low-dose hypersensitivity and highlight possible molecular mechanisms for increased radiation risks in the low-dose range [24, 25]). Radiation risk is still to be taken very seriously, and every effort should be made to keep radioactive contamination of ecosystems to a minimum.

4. Estimation and appraisal of radioactive contamination and its effects on the components of terrestrial ecosystems

Radioecology is a sub-discipline of ecology concerning the presence and effects of radioactivity on Earth's ecosystems. Some of the risks of ionizing radiation were known in the early twentieth century. Nevertheless, the discipline *de facto* started developing in the period following World War II and the bombings of Hiroshima and Nagasaki [26]. The advent of the Atomic Age not only gave the impetus to study radiation effects on ecosystems, but also gave them powerful tools in the form of radioactive isotopes, which could be used as tracers [26, 27]. Initially, studies were carried out by the US Atomic Energy Commission (AEC) at several sites crucial to the Manhattan Project, principally Oak Ridge, Tennessee, and Hanford, Washington; many of these studies dealt with the cycling with biogenic carbon, phosphorus and oxygen through ecosystems and were conducted with radioactive tracers (^{14}C, ^{32}P, and others) [27]. In parallel, studies were conducted in the former USSR in the closed town of Ozyorsk (Chelyabinsk-40). Some studies were conducted in secret; most of them dealt with dispersal and deposition of bomb radionuclides and with the bioaccumulation of radioactivity in crop plants and farm animals [28–30].

Without a doubt, the most significant contamination event in the context of terrestrial ecosystems is Chernobyl. It is estimated that, at the time of the accident, around 10% of the total core radioactivity was released, including 100% of all noble gases and around 30% of volatile atoms including 30% of the core radiocesium (^{134}Cs and ^{137}Cs), 55% of the core ^{131}I, and ~ 45% of the core ^{132}Te. Less volatile radionuclide species such as radiostrontium (^{89}Sr and ^{90}Sr) were also released in smaller amounts (~5% of core inventory), as well as <3.5% of the core transuranic nuclides (neptunium, plutonium, curium) [31, 32]. The core inventories and releases are summarized in **Table 3**.

The most significant release of radioactivity from the damaged reactor was in the form of noble gasses (^{85}Kr and ^{133}Xe). Nevertheless, fast atmospheric dispersal and the lack of chemical reactivity of noble gasses mean that radioactive krypton and xenon resulted only in trace global contamination. In contrast, the volatile iodine-131, released in significant quantities during the reactor fire, was the predominant problem in contaminated areas during 1986. It is estimated that up to 4000 additional thyroid cancers among people can be attributed to this nuclide [4]. In the long term, the most significant contribution of radiation dose to the biota is attributed to radiocesium (^{134}Cs, ^{137}Cs), particularly ^{137}Cs, due to its long half-life (30.17 years), its propensity to accumulate in plant and fungal matter and animal nerve and muscle tissue. The contribution of ^{90}Sr to the background dose is also significant, but much lower and often indistinguishable from pre-Chernobyl global fallout from atmospheric nuclear testing [34].

Radioecological research after 1986 in Europe involved multinational teams working in the Chernobyl exclusion zone (ChEZ) and the most contaminated areas of Belarus and Russia (Gomel and Bryansk regions), as well as many studies on a

Chernobyl core inventories at the time of accident				**Radioactive release**	
Radionuclide	**Symbol**	**Half-life (λ)**	**Core activity, PBq**	**% core activity**	**Released, PBq**
Krypton-85*	^{85}Kr	10.76 years	35	100	35
Xenon-133*	^{133}Xe	5.3 days	6500	100	6500
Iodine-131	^{131}I	8.02 days	3200	55	1760
Cesium-134	^{134}Cs	2.0 years	180	30	54
Cesium-137	^{137}Cs	30.17 years	280	30	85
Tellurium-132	^{132}Te	78 hours	2700	45	1150
Strontium-89	^{89}Sr	52.0 days	2300	5	115
Strontium-90	^{90}Sr	28.8 years	200	5	10
Barium-140	^{140}Ba	12.75 days	4800	5	240
Zirconium-95	^{95}Zr	1.4 hours	5600	3.5	196
Molybdenum-99	^{99}Mo	65.9 hours	4800	3.5	168
Ruthenium-103	^{103}Ru	39.26 days	4800	3.5	168
Ruthenium-106	^{106}Ru	1.0 year	2100	3.5	73
Cerium-141	^{141}Ce	32.5 days	5600	3.5	196
Cerium-144	^{144}Ce	284.9 days	3300	3.5	116
Neptunium-239†	^{239}Np	2.4 days	2700	3.5	95
Plutonium-238†	^{238}Pu	86.0 years	1	3.5	0.035
Plutonium-239†	^{239}Pu	24,110 years	0.85	3.5	0.03
Plutonium-240†	^{240}Pu	6580 years	1.2	3.5	0.042
Plutonium-241†	^{241}Pu	13.2 years	170	3.5	6
Curium-242†	^{242}Cm	163 days	26	3.5	0.9

**noble gases*
†transuranic nuclides

Table 3.
Core inventories and releases of the most important contaminants originating from the Chernobyl accident. Data obtained from [31–33].

national level focusing on areas with known contamination. Among the projects conducted in the ChEZ, several exemplary studies of the bioaccumulation of different radionuclides in wildlife stand out [17, 19, 34, 35]. Researchers have demonstrated that the appropriate sentinel species for radioecological studies comprise small rodents including representatives of family Cricetidae like *Myodes glareolus* Schreber, 1870, *Microtus arvalis* Pallas, 1778, *Microtus oeconomus* Pallas, 1776 as well as European murid species: the yellow-necked wood mouse *Apodemus flavicollis* Melchior, 1834 and the wood mouse *Apodemus sylvaticus* Linnaeus, 1758.

During the 200 s, researchers reported very high internal doses in Cricetidae, particularly the bank vole (*M. glareolus*) due to high dietary intake of ^{137}Cs [17, 34]. This has been confirmed by subsequent monitoring studies in the ChEZ [19, 35, 36], as well as in Alpine ecosystems in Bulgaria [37, 38]. Recent monitoring data suggest that *M. glareolus* is potentially the best rodent zoo monitor for residual contamination in Europe. A selection of results from two groups of monitoring programs, mentioned above is presented in **Table 4**.

Study	Location	Result
Chesser et al., 2001 [17]	Six different biotopes within the Chernobyl Exclusion Zone	Very high internal doses from ^{137}Cs in dry muscle of *M. glareolus* from areas with high and medium contamination; average ^{137}Cs body burden in *M. glareolus* 2902–24,720 Bq/g. High body burden of ^{137}Cs in *Sorex araneus*—2592–5901 Bq/g).
Beresford et al., 2008 [36]	Three different biotopes within the Chernobyl Exclusion Zone	High total-body internal doses from ^{137}Cs in *M. glareolus* 2260 ± 1290 Bq/g; much lower doses from ^{90}Sr in different species of small rodents (for *M. glareolus* 81.3 ± 22.1 Bq/g, for *Microtus* sp. 107 ± 35.0 Bq/g, for *Apodemus* sp. 66.6 ± 28.3 Bq/g).
Beresford et al., 2020a [19]	Reference (low-contamination) biotopes within the Chernobyl Exclusion Zone	Comparatively high doses from ^{137}Cs in *M. glareolus* from low-contamination "reference areas" in the ChEZ, total body burden of ^{137}Cs in *M. glareolus* = 649 Bq/g; comparatively high total body burden of ^{137}Cs in *Microtus* sp. (952 Bq/g); Much lower doses from ^{137}Cs in Soricidae (161 Bq/g for *S. araneus*, 121 Bq/g for *S. minutus*).
Iovtchev et al., 1996 [37]	Two localities (Musala peak and Skakavtsite), Rila Mountain, Bulgaria	Comparatively high whole-body total β-activities in wild rodents from both localities (2.5–3.0 Bq/g for *Apodemus* species, 3.25 Bq/g for *Chionomys nivalis* from Musala Peak, 2.75 Bq/g for *M. glareolus* from Skakavtsite).
Beltcheva et al., 2019 [38]	Two localities (Musala peak and Skakavtsite), Rila Mountain, Bulgaria	Overall 10-fold reduction in whole-body total β-activities in wild rodents from both localities. Highest residual activities observed in *M. glareolus* (0.52 Bq/g). β-activities in other rodents show more significant reduction (0.23–0.37 Bq/g for *Apodemus* sp., 0.38 Bq/g for *Ch. nivalis*).

Table 4.
A summary of the findings of five radioecological studies using small mammals as zoo monitors.

The summarized works show evidence for the high value of *M. glareolus* as a monitoring species for residual radioactivity from the Chernobyl accident due to its propensity to accumulate radiocesium. While accounting for the differences in values obtained by the various research groups, and the different time frames, another aspect of Chernobyl contamination becomes apparent: There are significantly higher depositions and animal body burdens of radiostrontium (^{90}Sr) within the Chernobyl exclusion zone, as opposed to very low amounts of ^{90}Sr present at greater distances from the accident site; this can be explained by the much lower volatility of strontium compared to cesium. This is one of the main reasons why ^{90}Sr is still a significant contaminant within the ChEZ, but in most of Europe the largest part of the Chernobyl-associated dose burden to the biota comes from ^{137}Cs.

During recent monitoring studies, conducted in Bulgaria in the period 1996–2020, small mammals such as rodents and insectivores were selected mainly due to their positions in the food chain like primary consumers, rapid maturity, large population number, and rapid biological reaction to the environmental changes [38]. The possible biological response of the organism was studied at different levels of organization of living matter, and evaluated the population number and structure, food spectrum, total beta-activity in target tissues, and organs of the investigated animals, standard hematological methods—to determine hemoglobin contents, hematocrit, and morphological characterization of erythrocytes, as well as cytogenetic methods. The food spectrum was analyzed as a basis for further investigations on the transfer of beta-emitters through the rodent populations and the whole ecosystem.

Moussala Peak 2925 m a.s.l.		**β-activity /mean ± SD/, Bq/kg**	**Beli Iskar (Skakavtsite area), 1400–1500 m a. s. l.**		**β-activity /mean ± SD/, Bq/kg**
Ap. flavicollis	**n = 12**	**230.3 ± 7.2**	*Ap. flavicollis*	**n = 13**	**366.3 ± 8.1**
Ch. nivalis	n = 12	382.0 ± 8.3	*M. glareolus*	n = 22	424.2 ± 5.3

Table 5.
Whole-body total β-beta activities at two localities (Rila Mountain, Bulgaria), 2019–2020 [38].

The total body burden of β-emitters of a species depends on the trophic chain position, food, life mode, physicochemical composition of the atmospheric precipitation, total suspended dust content in atmospheric air, and other factors. The total β-activities in Bq/kg of some small mammal species were investigated at two different altitudes in Rila Mountain, Bulgaria. The results, obtained in 2019–2020, are presented on **Table 5**.

All values were below 480 Bq/kg and were considered as referent.

Significant differences between mice and voles were obtained only due to the difference in their food specialization. Mice are omnivorous, while voles are mainly herbivorous species. Green vegetable parts accumulate radiocesium more actively than seeds and the quantity of the consumed low-caloric green food by animals is higher.

The comparison of the results obtained with the data 20 years ago makes it obvious that the values of total β-activity decreased by about 10 times in the period 1995–2019. Data obtained in the bodies of different monitor species of small mammals from Rila Mountain during 1995 varied from about 3500 Bq/kg in the yellow-necked wood mouse to 5000 Bq/kg in the snow vole. The total level of beta-activity in bank vole and yellow-necked wood mouse from Beli Iskar region during 1995 was between 2000 and 3000 Bq/kg [37].

High doses of radiation can influence the normal function of the blood and disturb the hematopoiesis. These were possible basophilic granulations that appear in enhanced, but also disturbed erythropoiesis, basophilic DNA fragments observed in a blood smear, frequently as a result of decreased spleen function, anemia, and overloaded bone marrow. However, the given results do not suggest such changes, and they have not been established.

A correlation between total beta-activity loading and chromosome aberration frequency in bone marrow cells was established. The percentage of chromosome aberrations in mice was about 1.6% and breaks were 0.2% and in herbivorous voles respectively 7.0 and 2.5%. The percentage of aberrant bone marrow cells of mice from the investigated regions is visibly lower than in vole species. This fact correlates with the recorded total body burden of β-emitters in herbivorous species in comparison with the omnivorous murids.

5. Principal remediation strategies for radioactive contamination

The issue of remediating radioactively contaminated terrestrial ecosystems dates back to the early years of the Atomic Age (1945–1965) when protection measures were a secondary consideration to weapons production. Tests were conducted in contaminated areas such as near Hanford, Washington, and Ozyorsk (Chelyabinsk-40) [27, 29].

After 1986, to protect the environmental health and resolve the liabilities due to eventual radioactive contamination, severely contaminated countries and the responsible institutions have undertaken certain remediation and protection measures:

1. mechanical/physical methods—creation of barriers, burial/demobilization of radioactive sources; deep tilling of agricultural fields for facilitating downward migration of radioisotopes;

2. forestry management—clearing and burial of severely contaminated coniferous forests presenting a fire hazard, natural succession/ecosystem restoration, and manual afforestation of contaminated agricultural areas with deciduous trees;

3. selective use of soil additives—addition of lime to increase soil pH and limit the uptake of ^{90}Sr by plants, use of fertilizers containing phosphorus, and potassium in order to reduce ^{137}Cs bioaccumulation in plant matter, the addition of complexing agents such as powdered zeolites, and other aluminosilicate minerals in order to demobilize ^{137}Cs; addition of hydroxyapatite (HAP) to prevent ^{90}Sr cycling in ecosystems;

4. crop selection in agricultural areas—production of non-food/feed crops such as cotton, flax, timber, and biofuels; use of land with low levels of contamination for sugar and oil production, whereby most residual radioactivity is removed during the refining processes;

5. careful livestock farming—feeding farm animals clean fodder, administration of powdered zeolites as bio-sorbents, the addition of salt licks containing Prussian Blue to reduce ^{137}Cs uptake by grazing animals.

Most of these strategies are discussed in detail elsewhere [39–44]. All of the methods were applied to some degree within the ChEZ and the highly contaminated areas of the former USSR [42]. By far, the most widespread method used was the deep tilling of agricultural fields. Nevertheless, one of the strategies for remediation, the use of zeolites for demobilization and biodetoxification of ^{137}Cs has only been tested on a small scale in the ChEZ, while, at the same time, being the most promising approach for countering the toxicity of radiocesium [39, 45].

6. Zeolites as bio-detoxifiers of radionuclides

Natural zeolites are one of the most interesting groups among minerals, some of which (clinoptilolite, mordenite, chabazite) have enormous potential in science and technology due to their high sorption capacity and the presence of deposits with huge reserves in many countries, including Bulgaria. In the early years of zeolite research, Ames (1960) found that clinoptilolite from the Hector deposit, California, is highly selective for Sr^{2+} and Cs^{+} [46]. Other heavy metals, especially monovalent ones, were also well adsorbed—respectively ion-exchanged by this natural zeolite. The author introduced an order of selectivity of clinoptilolite, which is:

$$Cs^{+} > Rb^{+} > K+ > NH_4^{+} > Ba^{+} > Sr^{2+} > Na^{+} > Ca^{2+} > Fe^{2+} > Al^{3+} > Mg^{2+} > Li^{+}$$

The ion exchange properties of clinoptilolite and its selective sorption are especially valuable in the control of radioactive waste from nuclear energy production. The mineral has been successfully used as a sorbent of radionuclides from water and contaminated soils, as well as a food additive to limit ^{137}Cs absorption in livestock [39, 41, 45].

Very significant research on zeolites has been conducted in Bulgaria for the past five decades, with two deposits of clinoptilolite in the Eastern Rhodope Mountains—Beli Plast and Beliya Bair-Zhelezni Vrata—being particularly suitable for bio-sorbents of heavy metals and radionuclides in the form of additives to food and livestock feed [47]. Recently, it was demonstrated that modified natural clinoptilolite from the Golobradovo deposit in the Eastern Rhodopes was practically non-toxic to laboratory mice and facilitated significantly the excretion of Pb^{2+}ions from the gastro-intestinal tract of the experimental animals, thus protecting them against lead toxicity [48, 49]. In parallel, other Bulgarian researchers validated the use of zeolites from the Eastern Rhodopes in decontamination procedures and as soil fertilizer and even developed a clinoptilolite-based artificial soil ("Balkanin") that was used for growing vegetables in space onboard the Mir station [50]. In the early 1990s, researchers from the Bulgarian Academy of Sciences developed a specially modified natural clinoptilolite (CLS-5) as a bio-sorbent for radiocesium (^{134}Cs and ^{137}Cs) and radiostrontium (^{89}Sr and ^{90}Sr) [51]. In a modified form and labeled KS-3, CLS-5 was used in the production of over 55,000 personal radiation protection emergency kits, most of which were distributed among the personnel of the Kozloduy Nuclear Power Plant and the people from the surrounding areas (**Figure 2**).

Two plastic vials containing CLS-5 with a quantity of 7 grams each have been integrated in the radiation emergency kit. The other components of the radiation protection kit are a painkiller syrette, a syrette with an antiemetic, a broad-spectrum antibiotic, potassium iodide (KI) tablets, and CBT (a radioprotector for abating acute exposure to radiation), bandages, and ethanol for disinfection [51].

As evident from the material presented, research into zeolites as bio-sorbents of radionuclides and heavy metals is fairly advanced in Bulgaria. The past achievements in developing modified clinoptilolite derivatives as ^{90}Sr and ^{137}Cs sorbents, and current and ongoing basic research in clinoptilolites as a countermeasure to Pb^{2+} and Cd^{2+} intoxication in mammalian species promise to yield the interesting results.

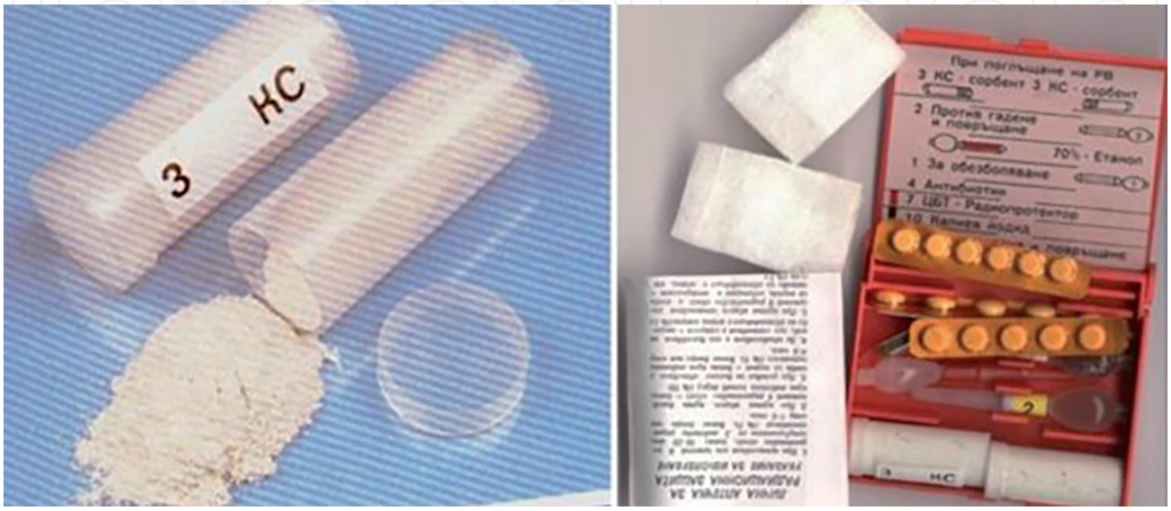

Figure 2.
Modified natural zeolites as part of a radiation protection emergency kit: Plastic vials containing CLS-5 (left), and the entire emergency kit (right) [51].

7. Conclusion

Ionizing radiation is one of the best understood cytotoxic and genotoxic agents. Nevertheless, much remains to be understood about the behavior of radionuclides in nature and the biological responses they induce. The radiobiology of low-dose, protracted irradiation is still an open area of research.

At the same time, bioaccumulation of certain radioisotopes along food chains poses serious ecosystem risks, or as the doyen of modern ecology Eugene Odum stated: "we could give nature an apparently innocuous amount of radioactivity and have her give it back to us in a lethal package!" [26].

The mitigation of environmental risks from radionuclides involves responsible management of the nuclear fuel cycle, as well as careful monitoring and safeguarding of nuclear installations. Among the strategies discussed in the chapter, all have been applied to a varying degree in severely contaminated agroecosystems and forest ecosystems. Perhaps the most promising venue of detoxication research is the application of zeolites as immobilizers and bio-detoxifiers for radiocesium and radiostrontium. Nevertheless, no method can fully remediate a contaminated ecosystem, meaning that prevention of radioactive contamination remains the first and best defense against anthropogenic radioactive pollution.

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