

## **SESSION 3**

# **RADIOLOGICAL ASSESSMENT OF AREAS CONTAMINATED WITH RADIONUCLIDES**



# FRACTIONATION, MOBILITY AND BIOLOGICAL ACCESSIBILITY OF RADIONUCLIDES IN THE ENVIRONMENT

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## ***Abstract***

*Fractionation of radionuclides is stipulated by varied distribution of certain isotopes on aerosol particles from nuclear explosions and accidents. The character of isotope distribution on the aerosol particles governs peculiarities of radionuclide distribution in the environments - the air, soil, surface and ground water and sea environment. Mobility of radionuclides after their fallout on the Earth's surface is determined by possibility of their contact with the aquatic environment and chemical properties of elements (or their compounds) in the form of which the radionuclides do exist. Biological accessibility is determined by mobility of radionuclides and their penetration into living organisms.*

*The presentation discusses the degree of transition of radioactive isotopes of certain elements from the particles to the water and muriatic acid (and gastric juice); the dependence is highlighted between biological accessibility of radionuclides, the distance from an epicentre of an explosion or accident and characteristic features of particles - carriers of radioactivity.*

*The processes of wash-off of radionuclides from soil to the water, vertical migration into the soil and their redistribution by surface and ground water are described. The use of wash-off coefficients, obtained by observation after nuclear explosions, provided a possibility to forecast already in May 1986 water contamination in rivers and storages in summer, 1986 and spring flood of 1987 in the zone of the Chernobyl accident.*

*The paper demonstrates that the reliable data on fractionation and biological accessibility are needed to solve effectively the problems of rehabilitation of the zones contaminated after nuclear explosions and accidents.*

The most important characteristics stipulating the behaviour and transformation of radionuclides in the natural environment are their fractionation, mobility, susceptibility to dissolving, and further biological accessibility responsible for their penetration into living organisms including the human body.

A knowledge of the listed characteristics together with physical properties of radionuclides (the decay rate, type of emitted radiation, etc.) provides the possibility to estimate contamination hazard and take the most effective measures to prevent or reduce the hazard and rehabilitate contaminated natural objects to a degree suitable for life and economic activities.

## 1. FRACTIONATION

After a release of radioactive products from certain sources they begin to propagate, deposit on the terrain together with soil particles, bomb construction or destroyed reactor fragments, etc., and migrate into different environments.

The radionuclide ratios vary continuously in the released mixture and in the transfer processes, their fractionation occurs, namely, changes in ratios of the amounts of radionuclides. Fractionation effects were described in studies [1, 2], etc., the data were

summarised in the study [3]. The main causes of fractionation are different rates of radionuclide penetration (sorption) into melted (glasses) elements of soil and constructions formed at an explosion or reactor accident. The propagating of these particles results in contamination of the environment [4].

E.g., after a nuclear explosion rather dangerous radionuclide  $^{89}\text{Sr}$  which has short-life predecessors is deposited on the surface of particles (the smaller ones have the greatest surface), and runs to great distances. While refractory  $^{95}\text{Zr}$  which penetrates into the particles (including the large ones) is deposited mainly in the close-in zone (“the local pattern”).

After breakdown of a reactor the essential amount of  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  was stipulated by their accumulation before the accident, and these radionuclides behaved as the refractory ones depositing in the close-in zone. Caesium isotopes ( $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ) being those of a volatile chemical element behaved as volatile products and run to very great distances after nuclear explosions and reactor accidents. On the other hand, transuranium elements are refractory ones.

The processes taking place during the first seconds (minutes) after nuclear explosions and having a dominant role in fractionation of radionuclides have been discussed in our study [4]. The results of calculation of fractionation factors for certain most important radionuclides agree well with the experimental data - Figure 1.

Analogous study [7] devoted to the Chernobyl accident was published. It pointed out the similarity to and difference in behaviour of radioactive products from the nuclear explosions and Chernobyl accident.

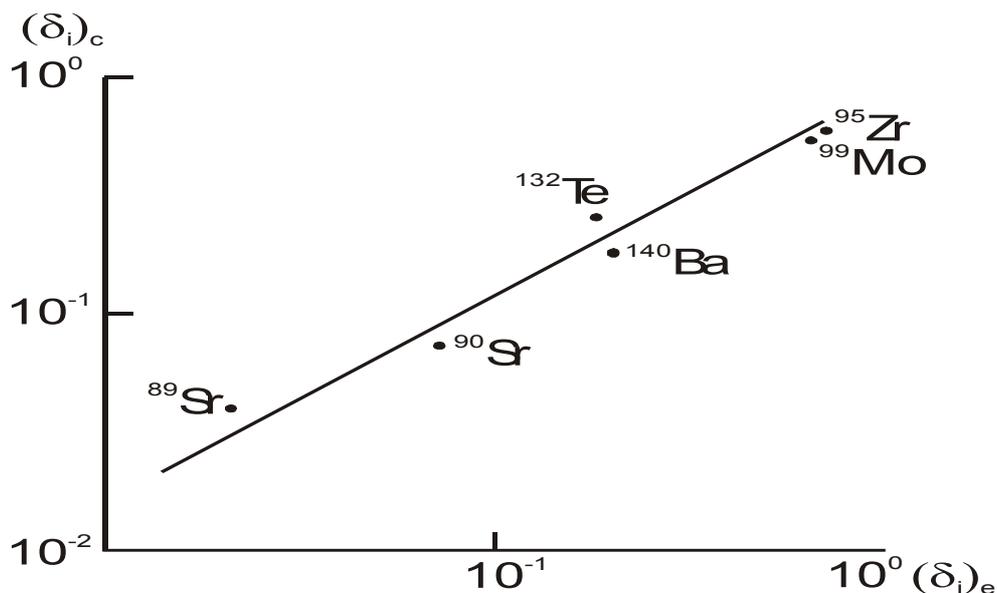


FIG. 1. Correlation between calculated  $(\delta_i)_c$  and experimental  $(\delta_i)_e$  values of the fractions of radionuclides deposited on the close-in pattern of a ground nuclear explosion.

TABLE 1. THE MAIN CHARACTERISTICS OF RADIONUCLIDES RELEASED AT THE CHERNOBYL ACCIDENT

Radio-nuclide	PC <sub>air</sub> <sup>*</sup> , 10 <sup>-17</sup> Ci/l	PC <sub>wat</sub> <sup>**</sup> , 10 <sup>-9</sup> Ci/l	Reactor inventory, Bq	Release, % of the inventory	Fractionation, relative to <sup>95</sup> Zr						Transfer Coefficient from soil in vegetation, relative to Cs <sup>137</sup>	Distribution coefficient, K <sub>d</sub>	Water mobility coefficient	Wind mobility
					Near pattern		Far pattern		Far pattern					
					North	South	North	South	North	South				
<sup>131</sup> I	1,5.10 <sup>4</sup>	1	2,4-3,2.10 <sup>18</sup>	50-60	4-9	0,9	190-290		1,0					
<sup>137</sup> Cs	4,9.10 <sup>4</sup>	15	2,2-3,3.10 <sup>17</sup>	23-33	3-5	0,6	170-370	55	1,0	10 <sup>2</sup> -10 <sup>4</sup>	<1%			
<sup>90</sup> Sr	4.10 <sup>3</sup>	0,4	1,7-2,5.10 <sup>17</sup>	4-5	0,5	0,5-4,0	5,0-7,5	2,5	1,0-10	10 <sup>1</sup> -2.10 <sup>3</sup>			Insigni-	
<sup>144</sup> Ce	2,2.10 <sup>4</sup>	12	3,2-4,9.10 <sup>18</sup>	3,5	0,9-1,0	1,0	1,4-2,0	1,4	1/20	10 <sup>4</sup> -2.10 <sup>5</sup>	<<1%			
<sup>106</sup> Ru	1,9.10 <sup>4</sup>	12	7,9.10 <sup>17</sup> - 2,1.10 <sup>18</sup>	3,5-6,0	0,6-1,5	0,5-0,75	20-30	7,7	1,0	10 <sup>2</sup> -10 <sup>3</sup>	<1%		ficant	
<sup>140</sup> Ba	1,5.10 <sup>5</sup>	25	4,8-5,7.10 <sup>18</sup>	3,5	1-1,6	0,8-1,0	10-15			10 <sup>2</sup> -10 <sup>4</sup>				
<sup>239</sup> Pu	3,0	2,2	8,0-9,6.10 <sup>14</sup>	2-3	0,9-1,0	1,0	1,4-2,0	1,4	Insigni-	10 <sup>2</sup> -10 <sup>4</sup>				
<sup>241</sup> Pu	160	110	1,7-1,9.10 <sup>17</sup>	2-3	0,9-1,0	1,0	1,4-2,0	1,4	ficant	10 <sup>2</sup> -10 <sup>4</sup>				

\* The permissible concentration in the air.

\*\* The permissible concentration in drinking water.

## 2. MOBILITY OF RADIONUCLIDES AFTER EXPLOSIONS AND ACCIDENTS

Fractionation of radionuclides and characteristics of their distribution on different particles are largely stipulated by their mobility and accessibility in the broadest sense (see Table 1). This conclusion was made in proper time after the Chernobyl accident.

Fractionation of radionuclides considerably varies over patterns of different directions resulted from certain portions of the radioactivity release from the destroyed reactor. Only the first portion was fractionated slightly at the initial moment of the release. It was of the approximate composition of products accumulated at the reactor ( $f^{137}_{144} \sim 1.0$ ). As to other directions, fractionation of volatile products was very high relative to the refractory ones even in the close-in zone ( $f^{137}_{144}$  reached 25-35) excluding the southern directions where  $f^{137}_{144}$  was again approximately equal to unit (the patterns of such a direction were formed from 30 April to 1 May, 1986 [7]). The analogous  $^{137}\text{Cs}$  fractionation factors were also observed at very great distances in many European countries [8].

“Caesium spots” were formed at great distances northward and north-eastward (in the Gomel, Mogilev, Tula oblasts of Belarus and Russia). They were characterised by huge fractionation factors of volatile radionuclides relatively to refractory  $^{95}\text{Zr}$ , with  $f_{i,95}$  values of up to 200-290 for  $^{131}\text{I}$ , 100-190 for  $^{110\text{m}}\text{Ag}$ , 150-250 for  $^{125}\text{Sb}$ , and 170-370 for  $^{137}\text{Cs}$  [7].

## 3. SOLUBILITY AND BIOLOGICAL ACCESSIBILITY

As already noted, migration and mobility of radionuclides depend on their solubility quite considerably. If the substance comprising the basis of particles (e.g.,  $\text{SiO}_2$ ) is practically insoluble in water and weak acids, so the fraction of isotope subjected to leaching can be calculated on the base of surface contamination of the particles  $N_i(t,r)$ . This fraction, strongly dependent on a particle size, practically coincides with the value of biological accessibility which may be calculated as a quotient from division of relative accumulation of this isotope in the given biological system resulted from radioactive particles of explosion, by relative accumulation of the isotope from solution (see [3,4]) - Figure 2.

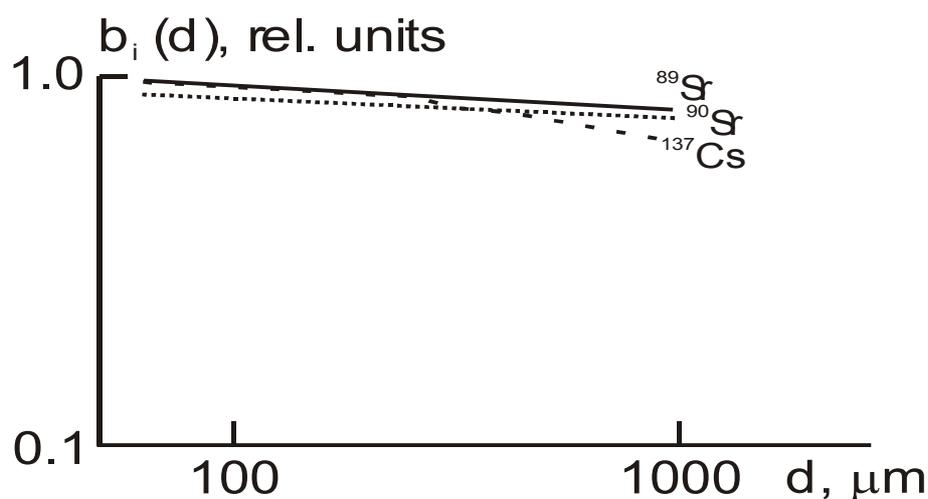


FIG. 2a. Dependence of the coefficient of biological accessibility  $b_i$  of  $^{89}\text{Sr}$ ,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  on the particle size after ground nuclear explosions.

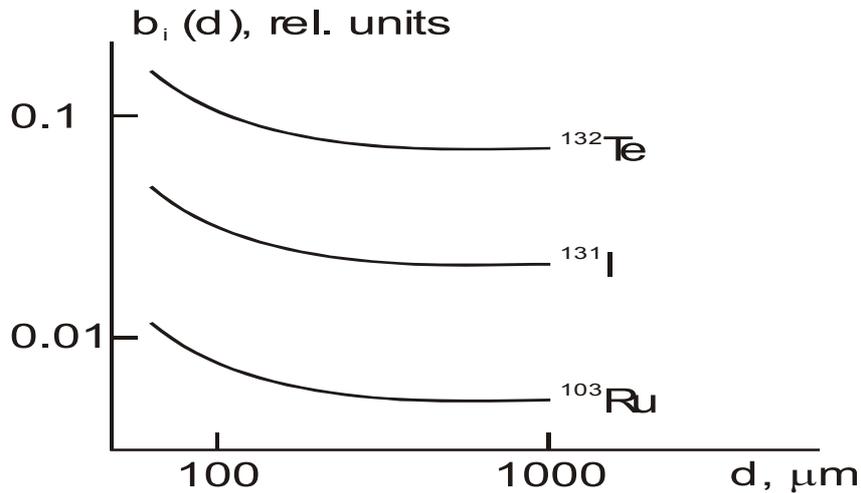


FIG. 2b. Dependence of the coefficient of biological accessibility  $b_i$  of  $^{103}\text{Ru}$ ,  $^{131}\text{I}$  and  $^{132}\text{Te}$  on the particle size after ground nuclear explosions.

The examined isotopes from underground nuclear explosions with cratering may be arranged in the following series according to their solubility, where  $^{90}\text{Sr}$  solubility is assumed to be the unit [9]:

$^{89}\text{Sr} >$	$^{90}\text{Sr} >$	$^{125}\text{Sb} >$	$^{140}\text{Ba} >$	$^{103}\text{Ru} >$	$^{106}\text{Ru} >$	$^{137}\text{Cs} >$	$^{134}\text{Cs} >$	$^{141}\text{Ce} >$
1.25	1.0	0.3	0.24	0.1	0.04	0.02	0.02	
$> (^{54}\text{Mn}, ^{60}\text{Co}, ^{144}\text{Ce}, ^{91}\text{I}, ^{95}\text{Zr})$								
0.01-0.001								

Using the data on the radionuclide solubility and ion-exchange reactions, one can estimate their mobility with ground and surface water.

#### 4. MIGRATION OF RADIONUCLIDES WITH GROUND AND SURFACE WATER

Studies [9, 10] provide the assessments of the possibility of radioactive products to migrate with ground water. Equations of ion-exchange include constant  $K_d$  called a distribution coefficient. It reflects the ratio between concentration of ions in bonded state (solid phase) and their concentration in the water solution.  $K_d$  values vary approximately within the range of  $10^1$ - $10^5$  for individual minerals and isotopes. Among the nuclides the least values of coefficient  $K_d$  are inherent to ruthenium, than strontium, caesium, cerium and plutonium. The mean values of  $K_d$  are: for ruthenium (on nuclear explosion patterns)  $10^2$ - $10^3$ , strontium  $10$ - $2 \cdot 10^3$ , caesium  $10^2$ - $10^4$  (in some cases  $10^5$ ), cerium  $10^4$ - $2 \cdot 10^5$ .

On the base of the study [5],  $K_d$  for strontium radionuclides (on the Chernobyl patterns) was  $2 \cdot 10^3$ , for caesium  $10$ - $5 \cdot 10^4$ , cerium  $5.8$ - $6 \cdot 10^3$ , zirconium  $50$ - $6 \cdot 10^3$  (the lowest values of  $K_d$  were observed in sandy soils).

After the Chernobyl NPP accident dozens of control wells were drilled around the NPP at a depth of the aquifer. Observations of  $^{90}\text{Sr}$  concentrations in water of these wells conducted for a year and a half after the accident showed that these concentrations did not exceed the background (pre-Chernobyl) levels.

Wash-off of radionuclides  $^{134,137}\text{Cs}$ ,  $^{106}\text{Ru}$ ,  $^{125}\text{Sb}$  and  $^{144}\text{Ce}$  by flood was investigated over five catchments after the Chernobyl accident in March-April, 1987 [1]. All the “wash-off” coefficients were less than 1 per cent.

## 5. BIOLOGICAL ACCESSIBILITY. ACCUMULATION OF RADIONUCLIDES BY PLANTS

“Fresh” radionuclides are the most accessible for plants after direct deposition on the plant surface and in the initial period of their being in soil. For instance, reduction of caesium radionuclides’ transfer to plants was detected with time, i.e. when the radionuclide became “older”.

It was mentioned that coefficient of radionuclide accumulation in plants  $K_a$  (the ratio between radionuclide concentrations in plant and soil, correspondingly) or  $K_s$  (the ratio between radionuclide concentration in plant  $C_p$  and soil deposition  $S$  per unit surface in sq. m) were closely correlated with distribution coefficients  $K_d$  in soil. This demonstrates the essential influence of both physical-chemical properties of radionuclides, and soil characteristics.

Accumulation of  $^{89}\text{Sr}$  and  $^{90}\text{Sr}$  varies within a wide range (30-400 fold) in accordance with soil properties and biological characteristics of plants. So,  $K_s$  is equal to 0.6-2.8 for grain and cereals, and it is 0.7-3.2 and 7.8-48 for potato tubers and tops (Bq  $^{90}\text{Sr}$ /kg of plant dry mass per kBq  $^{90}\text{Sr}$ /sq.m).

In the average,  $^{137}\text{Cs}$  accumulation by plants varies 20-30 fold according to the soil properties, and ten-fold to biological features of plants.  $K_a$  of agricultural plants varies from  $n \cdot 10^{-3}$  to  $n \cdot 10^{-1}$  for  $^{137}\text{Cs}$  which is in the average, 5-10 times less than this on  $^{90}\text{Sr}$ .

Fission products  $^{91}\text{Y}$ ,  $^{95}\text{Zr}$ ,  $^{95}\text{Nb}$ ,  $^{103}\text{Ru}$ ,  $^{106}\text{Ru}$ ,  $^{141}\text{Ce}$ ,  $^{144}\text{Ce}$  are not accumulated well in agricultural plants. It is stipulated by their considerable sorption in soil.  $^{106}\text{Ru}$  and especially  $^{144}\text{Ce}$  are the least accessible to plants, absorption of these radionuclides by plants being significantly higher from soddy-podzolic sandy-loamy soil ( $K_a \sim 0.01$ ) than from chernozem ( $K_a \sim 6 \cdot 10^{-4} - 2 \cdot 10^{-3}$ ).

Among neutron activation radionuclides,  $^{65}\text{Zr}$  is the most mobile, and the least one is  $^{60}\text{Co}$ . Concentration of transuranium elements is  $10^2 - 10^3$  times less in fruits and grains than in vegetative organs of plants. Accumulation of transuranium radionuclides by plants decreases in the following order:  $\text{Np} > \text{Am} > \text{Cm} > \text{Pu}$ .

To summarise, one can make a conclusion that among the set of radionuclides released in the environment after nuclear explosions and accidents, taking into account their fractionation, mobility and biological accessibility, their relative hazard varies significantly – Fig. 3. Only a bare handful of the radionuclides can be considered as extremely dangerous to all living.

Thus, from the point of view of human internal irradiation,  $^{131}\text{I}$ , plutonium isotopes (especially  $^{241}\text{Pu}$ ),  $^{144}\text{Ce}$ ,  $^{106}\text{Ru}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{140}\text{Ba}$ ,  $^{132}\text{Te}$ ,  $^{239}\text{Np}$  etc. are in the list of the most dangerous radionuclides released into the atmosphere during the Chernobyl accident. But taking into account all their transformations beyond the limits of the close-in pattern (restriction zone) this list should be restricted mainly by  $^{131}\text{I}$  and  $^{137}\text{Cs}$ . In the case of nuclear explosions the most dangerous radionuclides are  $^{131}\text{I}$ ,  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ; as to the Southern Urals accident (Kyshtym pattern) it is  $^{90}\text{Sr}$ .

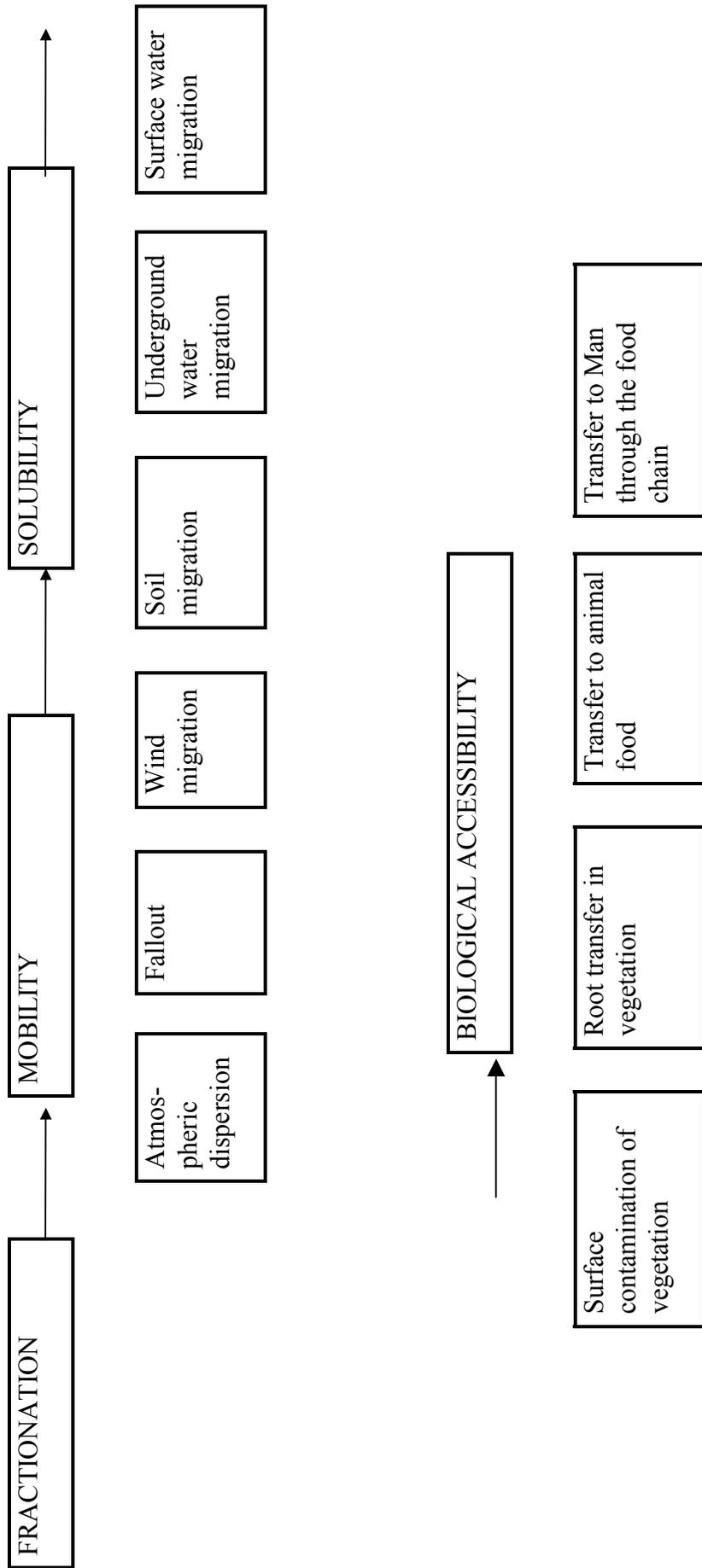


FIG. 3. Scheme of mobility and biological accessibility of radionuclides.

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# COMPARATIVE ESTIMATION OF RADIOECOLOGICAL SIGNIFICANCE OF NATURAL AND TECHNOGENIC OBJECTS OF EXCLUSION ZONE

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## **Abstract**

*Stocks of radionuclides in natural and technogenic objects of a Zone (forests, meadows, former agricultural lands, water objects etc., object "Shelter ", Sites of Radioactive Waste Disposal (SRAW) and Sites of Temporary Localization of Radioactive Waste (STLRAW)) are estimated. The estimations of fluxes of radionuclides both inside a Zone and out of its borders in normal conditions as well as in case of different natural and technogenic cataclysms (floods, fires, emergencies etc.) with account of probabilities of such events are discussed. The significance of the main fluxes of radionuclide migration beyond the Zone borders is estimated: a surface water flux (river Pripjat), air (wind) transfer, biogenic flux, technogenic migration. The significance of the non-radiological factors influenced on the ecological situation in a Zone (phyto-and zoosanitary, sanitary-epidemiological conditions) is discussed. The problems of irradiation of vegetation and animal organisms in terrestrial and water ecosystems, of personnel and people living on a Zone territory without permission (so called "self-settlers") are considered. The priority directions of activities for control over the radiological and ecological situation on the territory and in objects of a Zone are formulated.*

## 1. INTRODUCTION

According to the law of the Ukraine "On a legal status of the territory subjected to the radioactive contamination due to the Chernobyl accident, the Exclusion Zone (EZ) is defined as the territory, wherefrom the population have been evacuated in 1986. The zone is the territory with lands removed from an economic use, with the special form of management carried out by the Administration of the Zone. The area of the Zone is 2044 square kilometers. There are two towns (Pripjat and Chornobyl) and 74 villages within the territory of the Zone [1-5].

The radioactive contamination of the Zone is potentially dangerous for the Ukraine because of possible outflux of radionuclides beyond its borders. It is especially important because of the Zone's geographical position in the top part of the Dnieper water collecting basin, that is the main waterway of the Ukraine.

## 2. STOCKS OF RADIONUCLIDES IN OBJECTS OF THE EXCLUSION ZONE

The radiation factor till now is the main one in potential danger of the Zone objects for the population, living on the territories adjoining the Zone and for population of the Ukraine as a whole. A significant part of radionuclides released during the Chernobyl accident is concentrated here. The estimation of the radionuclides stocks distribution in various objects of the Zone according to the generalized data [6-81] is presented in Table 1.

TABLE 1. RADIONUCLIDES STOCKS IN VARIOUS OBJECTS AND COMPONENTS OF THE EXCLUSION ZONE (AS OF 1998)

Object	Activity, PBq			
	<sup>137</sup> Cs	<sup>90</sup> Sr	TUE	Total
Territory of EZ	5.5	2.5	0.13	8.13
Cooling pond	0.16	0.10	0.005	0.27
SRAWDs	3.4	2.8	0.15	6.35
STLRAWs	1.10	0.70	0.04	1.84
Total activity	10.2	6.1	0.33	16.6
SHELTER Object	480	260	10	740

The main activity of radionuclides is concentrated in the Shelter object. By the maximal estimations, there is about 180 tons of nuclear fuel in it, that contains the radioactive substances with about 740 PBq of total activity. Besides fuel, a plenty of radioactive materials, which consist of the residues of the destroyed unit active zone, reactor graphite, contaminated metal and building constructions of the power - unit are concentrated in the Shelter object [6-8].

The significant part of radioactive substances (about 8.2 PBq) released from the destroyed power Unit 4 is located in Sites of Radioactive Waste Disposal (SRAW) and Sites of Temporary Localization of Radioactive Waste (STLRAW). Approximately the same amount of radionuclides (8.1 PBq) is located in various natural objects of the Zone. The greatest radionuclides stock is in forests, that at the moment of accident have concentrated radioactive dust and aerosols. The territory with density of caesium-137 contamination over 555 kBq·m<sup>-2</sup>, strontium-90 contamination over 111 kBq·m<sup>-2</sup> and plutonium contamination over 3,7 kBq·m<sup>-2</sup> is about 1800 km<sup>2</sup>.

### 3. THE RADIONUCLIDE FLUXES INSIDE AND BEYOND THE ZONE BORDERS

The presence of radionuclides in various Zone objects predetermines the various intensity of radionuclides inclusion in transfer chains and, respectively, distinctions in the radionuclide flows inside and beyond the borders of the Zone. The main sources of the radionuclide inclusion in migration flows are the territory of the Zone (components of terrestrial and water biocoenoses) and technogenic objects of the Zone: SRAW and STLRAW, Shelter object, ChNPP, etc. The intensity of the radionuclide inclusion into migration chains and intensity of their migration flows inside and out the borders of the Zone may essentially grow, from the components of biocoenoses of the Zone in case of various natural and technogenic catastrophes (floods, fires etc.), and from the Shelter object and ChNPP in emergency cases. It is noteworthy that the influence of a number of the factors on the migration flows intensity is of constant character, the action of a number of other factors can be shown with rather low (but final) probability.

Generalization of results of a complex research in the Zone has allowed to estimate the importance of the basic ways of radionuclides migration beyond the Zone borders:

- *surface water drainage in the river Pripyat* - up to 0.09% of Cs and up to 0.6% of <sup>90</sup>Sr from the total activity of these radionuclides on the zone territory or 80-97% from a general flux of these radionuclides out of the boundaries of a zone by all paths of migration;

- *air (wind) transfer* - up to 0.004-0,02% of radionuclides from the total activity on the zone territory or 5 - 15% (in case of forest fires - up to 20%) from a general flux of radionuclides out of the boundaries of a zone by all paths of migration;
- *biogenic outflux* - up to 0.001% of  $^{137}\text{Cs}$  and up to 0.0006% of  $^{90}\text{Sr}$  from the total activity of these radionuclides on the zone territory or 0.1-3% from a general flux of these radionuclides out of the boundaries of a zone by all paths of migration;
- *technogenic migration* - up to 0.0002-0,8% from a general flux of radionuclides out of the boundaries of a zone by all paths of migration.

For the Shelter object and ChNPP, the basic ways of radionuclides transfer to the environment are as follows: the "organized" emissions, spontaneous outflux owing to leakage from the Shelter object, the emissions as a result of emergencies.

Let's consider the quantitative characteristics of the basic radionuclide flows within the territory of the Exclusion Zone (Table 2).

TABLE 2. RADIONUCLIDE FLOWS FROM VARIOUS SOURCES WITHIN THE TERRITORY OF THE EXCLUSION ZONE

Flow	Activity flux, TBq·year <sup>-1</sup>			Probability of realization per year
	Total	$^{137}\text{Cs}$	$^{90}\text{Sr}$	
Territory of EZ (flows and flux out of the borders of EZ)				
Superficial drain (limits of variation during 1990-1997)	4.4 -17.6	1.2-4.6	2.7-14.4	Constantly
Outflux by river Pripyat out of EZ borders in 1999 (high water of 13-15 % provision )	13.3	3.1	10.2	0.13-0.15
Outflux by river Pripyat out of EZ borders in 2000 (during 11 months)	4.5	1.5	3.0	Constantly
Drain with filtration waters from a reservoir - cooler	~ 0.4, Return - (0.02-0.04)	0.07	0.30	Constantly
Wind transfer	0.7	0.5	0.2	Seldom
Biogenic outflux	0.07	0.055	0.015	Constantly
Technogenic migration	0.016	0.012	0.004	Constantly
Shelter object (emissions of long-living RN in the environment)				
The "organized" emission	0.011			Constantly
Spontaneous emission through cracks	0.0006	0.0005		Constantly
ChNPP (emissions and dumps of long-living RN in the environment )				
Gaseous-aerosol emissions	0.048	0.047	0.001	Constantly
Dump in the pond-cooler with waste water	0.035	0.026	0.009	Constantly

### 3.1 Water transfer

The basic route of radionuclide migration out of the Zone borders is the river drainage formed at the expense of the surface drain and radionuclides outflow with underground waters. During 1990-1996 113 TBq of  $^{137}\text{Cs}$  and 130 TBq of  $^{90}\text{Sr}$  were flown out of the Zone. [7,8]. Nowadays, the contamination of water happens basically at the expense of washoff of radionuclides from the contaminated territories. The outflux of radionuclides by a waterway out of boundaries of a Zone can increase significantly in a case of extremely large flooding.

For example, in case of a maximal during post-accidental period high water with 13-15% provision, which was held in 1999, radionuclides flux out of the zone boundaries was about 13.3 TBq as a result of radionuclides wash-off from the territory of floodland.

Cooling pond of the ChNPP is the reservoir of the 22.9 km<sup>2</sup> area, the stock of water in it is 160 mln m<sup>3</sup>. The total activity of <sup>137</sup>Cs in bottom sediments reached 165 TBq, <sup>90</sup>Sr - up to 100 TBq, <sup>238,239</sup>Pu - up to 5 TBq [7,8]. The contribution of the cooling pond in the total radionuclides outflux is approximately 6%.

The radionuclides migration into the underground waters has not resulted in steady area contamination of them on the significant part of territory of the Zone. The integral outflux of <sup>90</sup>Sr with a underground drain in surface pools of a near-field ChNPP region for 300 years is evaluated as 15 TBq, or 3% from a general initial stock on water drain territory. The hydro-geological migration of <sup>90</sup>Sr from STLRAW can not result in a disastrous outflux of <sup>90</sup>Sr to river Pripyat.

### 3.2 Air (wind) transfer

The air (wind) transfer gives the essentially smaller contribution into radionuclide outflow beyond the Zone borders, in comparison with the water drain its contribution to the total flow does not exceed 10-15%. The significantly higher intensity of wind radionuclides transfer (up to 20% of the total flow) is observed in a case of forest fires. It is necessary to note, however, that the distance to which the radionuclides are transferred by air is rather small. In case of the top fire, the wind transfer may be up to 1.1-1.5 TBq within the territory of 100 ha. The calculations which have been made with the use of the obtained data show that for forest fire-fighters, the radionuclides content in the body reaches 0.1-0.2 of the permissible level for the persons of category A (personnel). Essential flux of radioactive substances out of borders of the Exclusion Zone may occur in case of extreme weather conditions, in particular, in case of hurricanes, tornados, etc.

### 3.3 Biogenic transfer

Outcome of the research of academician L. Frantsevich with co-authors have permitted to make a number of the extremely relevant estimations of biogenic migration of radionuclides out of the boundaries of a zone. From all the diverse organisms living in the zone, only separate groups are capable to create significant flows of radionuclides both in a Zone, and beyond its limits (Table 3).

TABLE 3. RADIONUCLIDES FLOWS WITH LIVING ORGANISMS OUT OF THE EXCLUSION ZONE

Groups of living organisms	Flow, GBq.year <sup>-1</sup>	
	<sup>90</sup> Sr	<sup>137</sup> Cs
Migrating birds	14.8	55
Small mammals	0.37	0.82
Large mammals	0.29	0.44
Total	15.5	56.8

### 3.4 Groups of living organisms transferring radionuclides within the Zone

Soil invertebrata with a total biomass stock of about 25 thousand tons which with participation of animals throw round about 20 - 30% of the phytomass and the biggest part of leaf-fall for the season. Estimations on radionuclide movement are absent. *Forest plantings* with phytomass stock of 75 up to 250 tons of dry substance per 1 ha and specific activity of  $^{137}\text{Cs}$  of 26-1700  $\text{Bq}\cdot\text{kg}^{-1}$  form the radionuclide flow of 1.85-370  $\text{MBq}\cdot\text{ha}^{-1}$ .

The mushrooms with a stock of 0.001 kg of dry weight per 1  $\text{m}^2$  and specific activity of  $^{137}\text{Cs}$  3.7-13  $\text{kBq}\cdot\text{kg}^{-1}$  form flow of 3.7-130  $\text{Bq}\cdot\text{m}^{-2}$ .

### 3.5 Groups of living organisms transferring radionuclides out of the Exclusion Zone.

*Large animals* with a total mass of several hundred tons and specific activity of  $^{137}\text{Cs}$  in soft tissues up to 3.7  $\text{kBq}\cdot\text{kg}^{-1}$ . Annual flux of radiocaesium out of the Exclusion Zone may be 740 MBq.

*The birds* are the important group of migrating animals existing in the Exclusion Zone. The total mass of settled and nesting birds is estimated as about 50 tons. Biomass of migrating birds exceeds the biomass of settled and nesting ones by ten to hundred times and reaches 4-5 thousand tons per year during migration in both directions. Estimations of maximum annual out-flow by birds may constitute the following: 55 G1Bq of  $^{137}\text{Cs}$  and 14.8 GBq of  $^{90}\text{Sr}$ . The total stock of  $^{90}\text{Sr}$  in zoomass as a whole (bird and animals) for the first years after the accident was 0.74 GBq and 2.2 GBq of  $^{137}\text{Cs}$  [ 10].

It is noteworthy that the contamination of water birds, nesting on the cooling pond, can result in accumulation of activity in food chains (bottom sediments - fish - bird) and separate individuals up to 37  $\text{kBq}\cdot\text{kg}^{-1}$ . The total activity of animals living in the Exclusion Zone is approximately 93 GBq.

From above mentioned estimations it is evident that the mushrooms and game water birds can contribute essentially to formation of human internal dose.

### 3.6 Technogenic transfer

Data on the role of technogenic transfer of radioactive substances out the exclusion Zone borders (use of the contaminated transport, etc.) are given in Tables 2 and 4. It is clear from the given estimations, the contribution of radionuclides technogenic transfer to the total flow out of the Exclusion Zone is insignificant.

TABLE 4. TECHNOGENIC RADIONUCLIDES FLOWS OUT OF THE TERRITORY OF EXCLUSION ZONE

Objects	Flow, $\text{GBq}\cdot\text{year}^{-1}$
Vehicle	16.0
Personnel	0.15
Cargoes in whole	13.3
Including forest production	0.037

### 3.7 Flux of radionuclides out of the Shelter object

In spite of the fact that the total radionuclide stock in the Shelter object is by the order of magnitude higher than one in the environment, it contributes insignificantly to formation of radionuclides flows in the Zone (Table 2). Under the conditions of the normal managing of the Shelter object the radionuclides flows from it are three orders of magnitude less in comparison with the water flow of radioactive substances out of the territory of Exclusion Zone.

However, in case of emergency (destruction of the roof) about 155 TBq of long-living radionuclides can be released in the environment (Table 5). In accordance with the forecast of RSC "Kurchatov institute", Institute of Nuclear Safety of RAS, and also GRS (Germany) the soil deposition of  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  on the border of the Exclusion Zone will be about  $150 \text{ kBq}\cdot\text{m}^{-2}$ . At the same time, total activity of the basic radiologically important radionuclides, that would be released into the environment in case of the considered emergency, would be less, than their amount taken out during 10 years with the river Pripyat drain. The main part of activity would be accumulated just nearby the reactor (in 5-10 km zone), however small part of them can be distributed to a significant distance (up to 50 km) [8].

### 3.8 Flow-out of radioactive substances from ChNPP

Under the conditions of normal operation of ChNPP the total out-flow of radioactive substances into the environment with gaseous-aerosol emissions and dumps in the cooling pond with waste water is about  $7.4 \text{ T bq}\cdot\text{year}^{-1}$ , that is much lower than radionuclides fluxes with water drain and wind transfer (Table 2).

TABLE 5. CALCULATED RADIONUCLIDE FLOWS WITHIN THE TERRITORY OF EZ AND OUT OF THE SHELTER OBJECT IN CASE OF EMERGENCY

Flow	Total activity, TBq.year <sup>-1</sup>	Probability, year <sup>-1</sup>
<b>Territory of EZ (fluxes and flow-out of EZ)</b>		
Flow by river Pripyat waters out of EZ due to wash-out flood-lands at high water of 25% provision (without protective structures)	18.5	0.25
Flow by river Pripyat out of EZ due to washout flood-lands at high water of 25% provision (with protective structures)	11	0.25
Wind transfer at fires	1.1 - 1.5	0.2
Wind transfer at the tornado passage above the Exclusion Zone	0.11 - 6.3	0.05
<b>Shelter object (emissions of long - living radionuclides in the environment)</b>		
Emission of long - living radionuclides at the roof destruction	155	$10^{-2} - 10^{-4}$

## 4. NON-RADIOLOGICAL FACTORS

The specific ecological situation that has been formed in Zone both as the result of the radioactive contamination of its territory and termination of the full-value care of forest and former agricultural caenoses, demands the careful study and estimation of the number of non-radiological factors importance, first of all, on forest territories. Nowadays, forests occupy almost a half of the territory, and former agricultural lands occupy the third part of the whole area of the Zone.

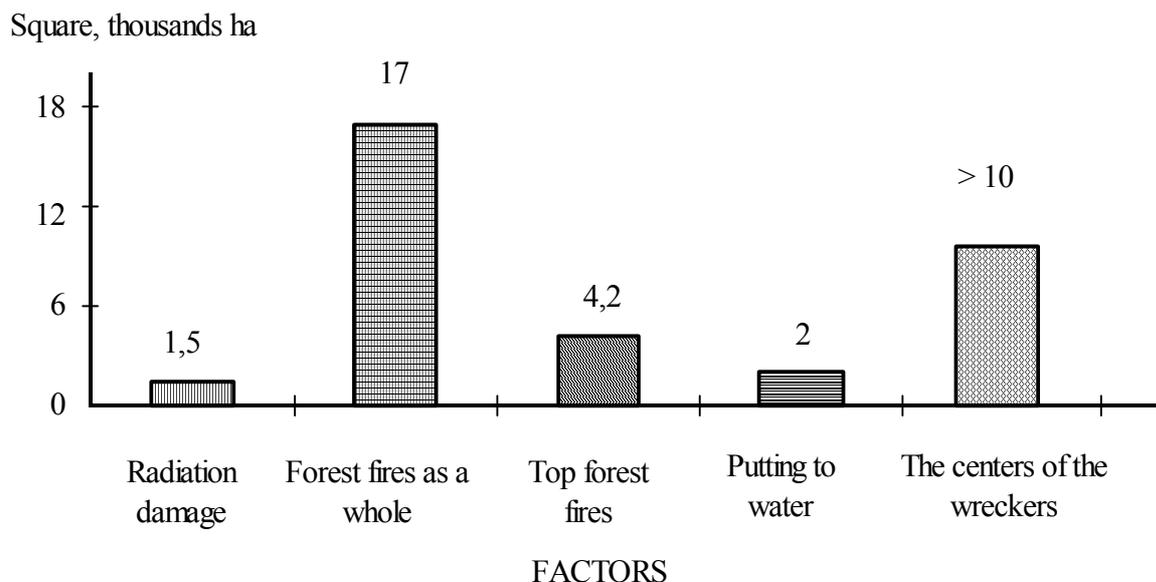


Fig. 1. Causes of damage and death of exclusion zone forests.

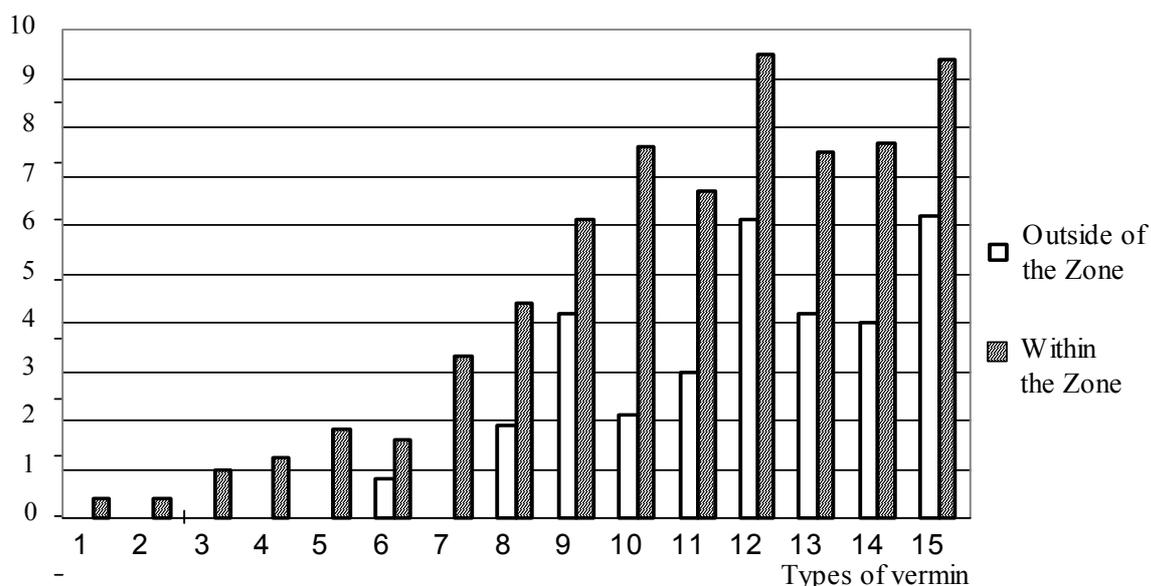


Fig.2 Ratio of phytofags number in gardens of Exclusion Zone and out of its borders  
 Main species of tortricid – garden pests in the given region: 1 – leaden-striped, 2- rose ugly-nest, 3 – motley-golden, 4 – fruit-tree ugly-nest, 5 – willow wry-feeler, 6 – hawthorn, 7 – netted, 8 – omnivorous, 9 – buddy, 10 – fruit-tree ugly-nest changeable, 11 – subcortical, 12 – smoky, 13 – white-spotted, 14 – frosty, 15 – green oak

The unfavourable natural and antropogeneous factors result in deterioration of the total ecological situation. So, for example, the most dangerous negative factor for forests is the forest fires (Fig. 1). Because of the absence of full-valuable system for forest care, the negative influence of the vermin and illnesses on general state of forests grows up.

Any infringement of the natural caenoses integrity results in significant changes of intensity and orientation of migration processes. It can be observed on pine plantings, which have been exposed to radiation and forest fires. The sharp termination of antropogeneous influence on

ecosystems (both agricultural and forest) stimulated natural processes, which not always had the favourable character. During the first years after the Chernobyl accident, the significant growth of rodent number was noted. In the Exclusion Zone the centers of illnesses and vermins are constantly registered, total number of which is almost twice as much, than in cultural crops areas outside the Zone (Fig.2 [10]).

The analysis of data on the disease quantity which are revealed on cultural plants and weeds shows, that on the latter, the number of revealed parasites is twice as much, than on agricultural cultures. In total, about 40 activators of different kinds of diseases are revealed. During the first years after the accident the creation of new forms had intensified considerably. In this period 5 forms, not registered in the European register, were identified, and three of them were highly virulenced and potentially dangerous (see Fig.2).

## 5. IRRADIATION DOSES

### 5.1 Plant exposure

During the first years after the accident it resulted in a large scale forest damage. Only in close proximity to the Chernobyl NPP about 600 ha of pine plantings were lost. As a whole, in the result of irradiation almost 1.5 thousand hectares of forest has dried. The damage of different degree was marked on the area of about 25 thousand hectares. At present, some radbiological effects are noted in mass on the area of about 1 thousand hectares, sporadically on the area of about 5 thousand hectares.

### 5.2 Human exposure [12-14]

First of all, the elevated radiation in the Exclusion Zone can affect the personnel participating in works on elimination of the accidental consequences. The analysis of distribution of doses among the personnel of a Zone displays, that the mean individual dose is at a level  $1.5-2 \text{ mSv}\cdot\text{year}^{-1}$ . The highest individual doses have been received by the personnel of a Industrial Association "ChNPP" and object "Shelter". The collective dose for total personnel of the Zone makes approximately 55 man.Sv. For the personnel of most radiologically dangerous enterprises (Shelter object, Special Enterprise "Complex") the collective dose is less than 10 man.Sv.

The special category of the people within territory of the Exclusion Zone is so-called 91 "self-settlers". Because of their consumption of food products that are produced partially on the territory of the Exclusion Zone, the internal irradiation contributes essentially to their total dose. The growing contribution of radiostrontium in formation of the dose is observed, that is the result of fuel particles transformations and increase of strontium-90 migration in biological chains. It should be noted, that in the case of practically complete life cycle with the use of products of the Exclusion Zone (for example, "self-settlers" in area of Usovo village), the average annual individual dose can reach  $25 \text{ mSv}\cdot\text{year}^{-1}$ .

### 5.3 Comparison of radiation danger of the Exclusion Zone objects

Comparative estimations of radiation danger of the Exclusion Zone objects should be considered (Table 6).

Owing to water migration, the individual dose of critical group of the population (fishermen, the population of the territory of the Dnieper cascade), can reach  $2 \text{ mSv}\cdot\text{year}^{-1}$ . In 1996-97 the

collective dose of the population of the Dnieper cascade territory (about 9 mln people owing to consumption of drinking water and about 26 mln people owing to the consumption of food products, including products of irrigated agriculture and fish) due to this source was estimated to be 20-30 man.Sv.

In case of *emergency wash out* of radionuclides from flood-lands located on the territory of the Exclusion Zone (25% high water) the collective dose could reach 250 man.Sv (without protective structures) and 40-50 man.Sv with the involved protective structures.

TABLE 6. COMPARATIVE ESTIMATION OF RADIATION DANGER OF THE EXCLUSION ZONE OBJECTS

Source of irradiation	Individual dose, mSv.year <sup>-1</sup> (critical groups)	Annual collective dose, man.Sv	Index of radiation danger (IRD)*
Superficial drain	2	up to 250 (high waters)	up to 60
Wind transfer	0.2	up to 3 (fire, tornado)	up to 0.6
Flow-out with vehicle	1.4	1.2	1.2
Non-authorized consumption of food products, produced and/or collected at EZ territory	25	up to 100	up to 100
Accident on Shelter object	up to 2000	up to 200	up to 2

\* IRD = Value of collective dose \* Probability of the event

The dose owing to *air transfer* is approximately one order of magnitude less. For critical groups of population, even in case of fires and tornado the individual doses are about 0.2 mSv, and the collective one is up to 3 man.Sv.

*Biogenic transfer.* The non-authorized consumption of migration birds meat, forest game, mushrooms and berries leads to formation of an individual internal dose up to 25 mSv. year<sup>-1</sup>. The collective dose in this case is estimated at the level of 100 man.Sv.

Doses caused by the *technogenic flow* of radionuclides out of the Exclusion Zone with the use of contaminated vehicles for critical group of the population (drivers, the repairmen) are about 1.4 mSv in the case of the most conservative assessments. The collective dose is estimated at the level of 1.2 man.Sv. In case of hypothetical emergency emission from the Shelter object the increase of vehicle contamination will be observed, however it will not result in essential increase of technogenic out-flow, and the collective dose, respectively, because of the application of countermeasures to prevent distribution of radionuclides (radiation monitoring, sanitary treatment etc.).

The accident on Shelter object (destruction of building) could result in the irradiation of separate persons from the personnel in dose up to 2000 mSv. Thus, the collective dose of the personnel participating in liquidation of this failure (the civil defence troops, the specially trained personnel) can reach 200 man.Sv. Naturally, radionuclides released in the environment will bring additional contribution in the migration flows and doses of irradiation due to contamination of water, air and due to biogenic transfer.

Due to the fact that various considered above processes of radionuclide transfer as well as processes of dose formation to personnel and population are not equally probable, neither individual, nor collective dose themselves can not serve as the complete characteristic of radiation danger of objects of the Exclusion Zone. To take into account the importance of each source of radiation danger, it is possible to enter concept of "an index of radiation danger", which is equal to the product of collective dose and probability of occurrence of an event (process). The event (process) probabilities are presented above in Table 5. The data presented in the right column of Table 6 account for these parameters.

As can be seen from Table 6, the greatest radiation danger represent water outflux and non-authorized consumption of the radioactively contaminated food products. Rather low radiation danger of the Shelter object, despite of high levels of individual and collective doses, is caused by low probability of occurrence of emergency.

## CONCLUSIONS

- (1) Under the conditions of normal operation, the radionuclides flows from technogenic and natural sources of the Zone exceed those from the Shelter object by one to two orders of magnitude.
- (2) In case of emergency on the Chernobyl NPP or Shelter object, the radionuclides flows and human dose from natural and technogenic sources remain compared with those from emergency sources. Besides, in case of accident on the Shelter object the main share of radionuclides will be distributed on adjacent territory and included in migration chains.
- (3) The adverse influence on natural objects both inside the Zone and on the adjacent territory is rendered also by the non - radiological factors, in particular, by phyto-sanitary conditions which has been usual at the territory of the Exclusion Zone.
- (4) For the management of radiation situation on the territory and on the objects of the Exclusion Zone the following is necessary:
  - to focus the basic works on the Shelter object on the reduction of probability of emergencies;
  - priorities of works on natural and technogenic objects of the Zone, where it is difficult to reduce the probability of emergencies, should be reduction of their radiation influence (prevention of wash - outs during flooding and of forest fires, strengthening of the control of the non - authorized use of objects of the Exclusion Zone).
- (5) It is necessary to define priorities in planning and realization of measures on liquidation of consequences of failure in the Exclusion Zone in view of the analysis of the data and those conclusions formulated in the present report.

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## DISCUSSION AFTER THE PRESENTATION OF V.I. KHOLOSHA

**I.J. Vasilenko (Russian Federation):** What fraction of the radioactivity in the exclusion zone is removed each year?

**V.I. Kholosha (Ukraine):** Leaving aside the “Shelter”, the total radioactivity in the exclusion zone is 650-700 000 Ci. Less than 1000 Ci-in other words, less than 1% of the total radioactivity-is removed from the exclusion zone each year.

**I.J. Vasilenko (Russian Federation):** What is the present condition of the “Shelter”?

**V.I. Kholosha (Ukraine):** Recent examinations of the steel structure indicate that the probability of collapse is fairly low- $10^{-2}$ -even in those parts which are considered relatively unstable. The work done in 1996 to strengthen the ventilation duct and two of the girders has reduced the probability of collapse. In our view, the “Shelter” is at present stable.

**K. Mück (Austria):** Are there any plans to resettle parts of the exclusion zone?

**V.I. Kholosha (Ukraine):** In the south-western part of the exclusion zone there are areas where people would receive doses of less than 1mSv/year. However, we must also take economic factors into account since the infrastructure within the exclusion zone has been largely destroyed. Moreover, you cannot expect people to resettle in the exclusion zone without their children, and people with children are going to have grave misgivings about the continuing existence of the “Shelter” and of various restrictions. On the other hand, thought is being given to the idea of using parts of the exclusion zone for the storage of spent nuclear fuel.

# EVALUATION OF THE RADIOACTIVE POLLUTION OF THE TERRITORIES AROUND KRASNOYARSK-26

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## ***Abstract***

*The objective of this study, funded by DGXI (now DGENv) of the European Commission, was to evaluate the impact of radiological contamination in the environment, which has resulted from the operations at the nuclear establishments at Krasnoyarsk-26, and to determine whether any remedial actions are required. Environmental data has been obtained for a wide range of environmental media and for a number of years covering the period before and after the closure of the single pass reactors at Krasnoyarsk-26. These media include air (aerosols and deposition), water, sediments, soil, floodplain deposits, foodstuffs and vegetation. Information was also obtained on demography, meteorology and the general geological, geographical and hydrological characteristics of the regions. A GIS system was established to contain all of this data and to display a number of themes. The sampling and analytical methodologies were evaluated and the data was examined to determine any trends with time or distance from the contamination sources. The data was compared with other global data available in the open literature. A methodology for the radiological assessment of the contamination was established and the calculations performed. The results of the radiological assessments showed that the most significant exposures resulted from the occupancy and use of the contaminated floodplains and from the potential exposure to hot particles. Doses of several millisieverts a year could result from some of the most contaminated sites and of tens to hundreds of millisieverts from hot particles. Criteria for remedial action and a number of remediation options were evaluated.*

## 1. INTRODUCTION

Much of the emphasis of recent and current projects in the former Soviet Union has been targeted at addressing the problems resulting from the accident at Chernobyl, spent fuel management in North West Russia and environmental contamination at Chelyabinsk-65 (Mayak, e.g. Karachai Lake). Relatively little is known about the problems of the nuclear facilities in Siberia and, particularly, about their impact on the environment.

Concerns about these Siberian combines have been raised recently [1, 2]. These reports suggested that significant levels of environmental contamination are present around these sites. For example, contamination of the riverbanks of the Yenisei River, downstream from Krasnoyarsk-26, is reported with measurements of  $^{137}\text{Cs}$  up to 220,000 Bq/kg. About 2 PBq has been reported to be discharged to the Yenisei River and 17 EBq are reported to have been discharged to deep wells near the site. The European Commission, DGXI, commissioned this project in 1997 to evaluate the consequences of the radiological contamination that may exist around Krasnoyarsk-26.

Krasnoyarsk-26, now known as Zheleznogorsk, is the site of a nuclear reactor plant and reprocessing facility in Siberia in the Russian Federation. These facilities are known as the

Mining Chemical Combine (MCC). Operations began there in late 1958. It is located approximately 50 km north from the city of Krasnoyarsk in Siberia. The reactor plant consists of three industrial graphite reactors for the production of weapon-grade plutonium from irradiated natural uranium. The radiochemical plant at the site produces weapons grade plutonium from processing the reactor fuel.

Two of the reactors were designed to abstract cooling water directly from the Yenisei River and then to discharge it back to the river without any purification. The water discharged into the river contained both activation and fission products. These radionuclides have been adsorbed onto sediments that, in turn were distributed by flood action onto the riverbanks and islands. As a result there are areas of contamination for hundreds of kilometres downstream. The pattern of radioactive contamination of river sediments and the floodplains are complex. The maximum contamination density observed was  $200 \text{ Ci/km}^2$  ( $7.42 \text{ MBq/m}^2$ ) of  $^{137}\text{Cs}$  on the island of Atamanovo where the exposure rate was  $150 \text{ } \mu\text{R/hr}$  ( $13 \text{ mSv/year}$ ). The two single pass reactors were shut down in 1992.

Environmental contamination has also arisen as a result of the aerial dispersion of radionuclides from surface ponds on the industrial site of the MCC old liquid radioactive waste reservoirs.

The project was restricted to evaluating radioactive contamination outside of the sanitary protection zone (SPZ). Information concerning the level of discharges from the sites to the environment were also out with the scope of the project, with the exception of that already in the public domain.

The project was managed by AEA Technology and included contributions from the VG Khlopin Radium Institute, based in St Petersburg and the Mining Chemical Combine at Krasnoyarsk-26. VNIPIPT, Moscow, also provided comments on the reports generated during the project.

A series of technical reports have been prepared by AEA Technology and the VG Khlopin Radium Institute which cover the technical work programmes described earlier in detail [3]. It is the intention of this paper to summarise the findings of each of these reports, to bring out their salient points and outstanding issues and to describe the main findings and conclusions from the project.

## 2. METHODOLOGY

The methodology for achieving the objectives of the project was developed during an initial series of discussions within project team. These discussions established the scope of the project, what information could be made available and the responsibilities for obtaining the information and for the subsequent evaluation and assessments of its implications.

It was agreed that information concerning radionuclide concentrations in environmental media outside of the sanitary protection zone (SPZ) for Krasnoyarsk-26 would be made available for a number of years of measurements, including the periods before and after the single pass reactors ceased operation, where possible. However, it was not possible for information concerning the SPZ, and/or discharges from the sites, to be made available.

In order to specify the exact requirements for the radionuclide information a questionnaire was prepared to guide the dissemination of this information and to allow for it to be recorded

electronically and facilitate the production of a GIS (Geographical Information System). The purpose of the GIS was to provide a structured system for the recording of the environmental radioactivity data, and other supporting data, and to allow this information to be represented spatially on maps of the area. The GIS also facilitated interpolation between data points and correlation's between different sets of data.

When the datasets were complete for each site they were evaluated against the following criteria:

- sampling methodology
- analytical methodology
- comparison with global data for
- sites near nuclear establishments
- non-industrial locations
- comparison with other reported data for the area

With the validity and representativeness of the data established, the potential radiological impact was assessed. The radiological assessment considered the intake of radionuclides from inhalation of air (aerosols and particulate material), ingestion of foodstuffs, water and soils/sediments (inadvertent), and external exposures from  $\gamma$ -emitting radionuclides. The preliminary calculations considered a pessimistic scenario whereby the highest observed levels for each environmental medium were used together with very conservative generic assumptions for dietary and occupancy habits. This simple approach allowed the screening out of unimportant pathways for exposure.

Finally, potential remediation options to reduce or remove the exposure pathways were evaluated and recommendations for further work to improve the understanding of potential future exposures from each site were made.

### 3. ENVIRONMENTAL RADIOACTIVITY NEAR KRASNOYARSK-26

In this section the most relevant environmental data obtained for Krasnoyarsk-26 is summarised. All of the original datasets, and their detailed evaluation, are presented in [3].

Information in the following areas was obtained for Krasnoyarsk-26:

- Atmospheric radionuclide concentrations
- Background concentrations in all media
- Demography
- Deposition of radionuclides (bulk)
- Deposition of radionuclides to soil
- Diet

- External dose
- Radionuclide content in fish
- Radionuclide content in food stuffs
- Frequency and radionuclide content in hot particles
- Lifestyle
- Meteorology
- River chemistry & flow
- Radionuclide content in sediment
- Radionuclide content in soil
- Soil type
- Radionuclide content in vegetation
- Radionuclide content in water
- Plutonium data

This data was provided by the MCC and the VG Khlopin Radium Institute and collated by the VG Khlopin Radium Institute. The radionuclide concentrations in environmental media are reported from the late 1980's up to 1997 in most cases. Most of the data is also reported as monthly and/or annual averages.

### **3.1 Atmospheric radionuclide concentrations**

Measurements of radionuclide concentrations in air are made at three sites, two of which are downwind and one is upwind of the MCC. A range of radionuclides was measured, including  $^7\text{Be}$ ,  $^{60}\text{Co}$ ,  $^{90}\text{Sr}$ ,  $^{106}\text{Ru}$ ,  $^{137}\text{Cs}$  and levels of gross alpha and beta activity. There is little evidence for a significant influence of plant plume on air concentrations at the measurement sites since concentrations at the three measurement sites all quite similar (upwind and down wind).

### **3.2 Deposition of radionuclides (bulk)**

Measurements are of bulk deposition (wet and dry) at several sites around the MCC. Two of these sites are approximately 70 km to the west of the MCC, and represent sites that receive background levels of deposition predominantly from global fallout (plus contributions from locally resuspended soil). Annual mean deposition fluxes of  $^{137}\text{Cs}$  are small, and up to 1995, appear to have been declining.

### **3.3 External dose**

Annually averaged external doses were measured at a number of locations within 20 km of the MCC in 1992 and 1994. There are also data for gamma dose rates in air at the locations where soil and sediment samples were taken. The annually averaged external doses, other than those

from the contaminated floodplains and islands, range from between 5 and 10  $\mu\text{R/h}$ , which is well within the normal range expected for natural background radiation. The dose rates measured at the locations where contaminated floodplain soils were sampled were considerably higher than this average value. Up to 150  $\mu\text{R/h}$  has been reported for some locations.

### **3.4 Radionuclide content in fish**

Concentrations of  $\gamma$ -emitting radionuclides are reported for a number of sites upstream of the release point from the MCC and up to 37 km downstream. Extensive data are available in terms of sample frequency, sample locations and species type.

The short lived isotopes  $^{32}\text{P}$ ,  $^{24}\text{Na}$  and  $^{35}\text{S}$  were responsible for the majority of the dose to the local population from eating fish during the period when the single-pass reactors were operating. Concentrations of  $^{137}\text{Cs}$  in fish downstream of the release point do not exceed about 20  $\text{Bq kg}^{-1}$ .

### **3.5 Radionuclide content in foodstuffs**

Information on radionuclide levels was reported for a number of foodstuffs sampled between 1990 to 1997. The foodstuffs included milk, beef, potato, cabbage and carrot. The locations ranged from within a few kilometres of the site to up to 250 kms downstream. Average milk concentrations for the region as a whole are reported as being 0.79  $\text{Bq/kg}$  for  $^{137}\text{Cs}$ .

### **3.6 Frequency and radionuclide content in hot particles**

A number of hot particles have been detected in several areas downstream of the MCC at Krasnoyarsk-26. The areas where particles have been found have been described together with the analysis of the radionuclide activities of the particles and of the floodplain deposits where they were located. The source of the hot particles is likely to be from spent fuel fragments from the single pass reactors at Krasnoyarsk-26. There has however been little analysis of the particles structures, one measurement suggested a graphite-like matrix. The lack of information on plutonium and uranium isotope ratios make it difficult to be certain about the particles origins. There is evidence that the particles found are of different ages implying that the release of the hot particles has taken place over a number of years. There is a wide range in the specific activities of the particles (from  $10^4$  to  $10^7$   $\text{Bq }^{137}\text{Cs}$ ) implying that the particles may come from different sources and originated at different times.

### **3.7 Radionuclide content in sediment**

A substantial dataset was provided about radionuclide levels in the sediments of the Yenisei River. Most measurements were made within 300 km of the discharge point. Upstream background concentrations of  $^{137}\text{Cs}$  are three orders of magnitude lower than the peak concentrations in the sediments down stream of the reactor discharge; however, a few down stream concentrations are comparable to background levels. Concentrations of up to 1600  $\text{Bq/kg}$  of  $^{137}\text{Cs}$  have been observed up to 650 kms from the discharge point.

### **3.8 Radionuclide content in soil**

Essentially, there are two sets of measurement sites: ones which are associated with the floodplains and islands of the Yenisei river, ‘down river’ from the MCC, and others not

adjacent to the river, 'non-river' sites located ten's of kilometres from the MCC. Data is reported from 1992 to 1997. The measurements made in floodland deposits are referred to here as 'soils' but these are in fact hybrid solids, which are neither true soils or sediments and have quite distinctive properties.

The 'non-river' sites have background concentrations of  $^{137}\text{Cs}$  are about  $1 \text{ kBq m}^{-2}$  which are smaller than those at many European sites, even making allowances for the small amounts of radioactive decay between the sampling dates.

The data for 'down-river' sites show that there are elevated concentrations of radionuclides at several locations downstream of the MCC. The concentrations do not fall predictably with downriver distance from the discharge point; for example the concentrations of  $^{137}\text{Cs}$  in soil are not highest close to the discharge point. The data show that concentrations of  $^{137}\text{Cs}$  in soil are elevated at up to 1000 km down river of the discharge point. The vertical profiles of  $^{137}\text{Cs}$  concentrations indicate that the highest concentrations are not always within the rooting zone and the profiles of concentration with depth are not consistent between different sites.

There is considerable spatial variability in the reported measurements. At one site for  $^{137}\text{Cs}$  (176 km downstream of the discharge point), variability of about 2 orders of magnitude is apparent for samples collected at the same time within a few 10's of metres. Such large variability over a small scale could be explained by inhomogeneous contamination from flooding events or perhaps by the presence of 'hot' particles.

The radionuclides present in floodplain soils, and river sediments, are a potential source of future contamination of other areas in the Yenisei/Kara Sea system through re-mobilisation and dispersion of the radionuclides themselves or re-suspension and deposition of the contaminated soils/sediments.

### **3.9 Plutonium data**

During the summer of 1997, soil was sampled at locations very close to the perimeter of MCC and at two locations downwind which might have received deposition, and at a further two locations which are subject to episodic flooding from the Yenisei. Levels of Pu-239+240 were determined in the soil samples, together with some other radionuclides ( $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{60}\text{Co}$ ). The concentrations of  $^{137}\text{Cs}$  and Pu at two locations near the complex (storage accumulator of liquid wastes and storage area for non-technological wastes) were elevated over background levels. Measurements a few hundred metres downwind show concentrations in soil close to background. This suggests the transfer mechanism from the liquid waste storage areas is aerosol transfer of material from the liquid surface. Concentrations of Pu do not seem particularly high at the downriver sites, although  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$  and  $^{60}\text{Co}$  are elevated over background levels. The Pu concentrations very close to the perimeter of the SPZ of the MCC are approximately an order of magnitude greater than concentrations in rural UK and similar to those at a village near the Sellafield site.

## **RADIOLOGICAL ASSESSMENT OF ENVIRONMENTAL CONTAMINATION**

The radiological assessment of the contaminated territories around Krasnoyarsk-26 has been based on the following pathways:

- the ingestion of radionuclides via contaminated foodstuffs and drinking water
- the ingestion of radionuclides via contaminated soils and sediments

- the inhalation of aerosols
- the inhalation of particulate material
- occupancy of contaminated land (via external exposure)

The radiological parameters used in the dose assessment, that is doses per unit intake (Sv per Bq) are based on internationally accepted criteria [4]. The rates for inhalation and ingestion (mass/volume per unit time) are arbitrarily based on standard reference parameters. External exposure models are based on simple models of exposure to infinite plane sources. Occupancy and habit data were based on locally observed patterns [3]. Calculations were carried out for the highest values observed for each sample type. This allowed the most important pathways to be identified.

TABLE 1. SUMMARY OF DOSES FROM ENVIRONMENTAL RADIOACTIVITY AT KRASNOYARSK-26

Pathway	Maximum Annual Individual dose, Sv/y		
	Adult	Child	Infant
Inhalation of air	1.66E-08	1.59E-08	1.34E-08
Inhalation of soil	5.62E-05	1.98E-05	5.96E-06
Inhalation of soil (2)	9.15E-07	7.28E-07	4.69E-07
Inhalation of sediment (1996)	1.17E-06	1.04E-06	6.83E-07
Ingestion of foodstuffs	5.03E-05	5.49E-05	1.20E-04
Ingestion of fish	5.73E-05	1.14E-05	2.85E-06
Ingestion of water	3.12E-06	2.41E-06	3.67E-06
Ingestion of soil (1)	1.30E-05	5.88E-06	3.44E-05
Ingestion of soil (2)	4.03E-06	1.03E-06	4.8E-06
Ingestion of hot particles, mean (3)	2.32E-03	1.80E-03	2.20E-03
Ingestion of hot particles, max (4)	3.84E-02	2.96E-02	3.55E-02
Ingestion of hot particles, max (4)	3.77E-01	2.90E-01	3.48E-01
Ingestion of sediments (1996)	4.17E-06	1.27E-06	6.81E-06
External irradiation	5.34E-03	5.34E-03	5.34E-03

- (1) from 'down-river' soils
- (2) from 'non-river' soils
- (3) mean values from 13 particles
- (4) max values are derived from the highest activity observed in a particle
- (5) detailed assessments of the potential exposures to hot particles are given in [3]

The floodplains and hot particles therefore represent the most significant exposure pathways for the radiation exposure of the population in the region near, or downstream from, Krasnoyarsk-26. A further potential source of exposures, particularly to infants, could arise from the ingestion of contaminated milk. There is evidence at one site, on the contaminated floodplain that levels of <sup>137</sup>Cs contamination in milk is relatively high. Other sites on the contaminated floodplain did not show elevated levels in locally produced milk. The highest values were used in the above calculations and these are therefore conservative. The population also have a wide variety of food sources and are not dependent solely on locally produced food.

A key feature of the contaminated floodplain is the complex nature of the distribution of the radioactivity. There is no simple correlation with distance downstream and samples taken at apparently adjacent sites, at the same time, can have very different activities.

The maximum doses assessed here, associated with the use and occupancy of the contaminated floodplains and islands of the Yenisei, are higher than the Russian standard for the annual dose limit for member of public (1 mSv/y) by a factor of 2 or 3. The exposures associated with the contaminated floodplain would therefore be classed as requiring further study, and possibly intervention, under the Russian regulatory system.

## 5. REMEDIATION OPTIONS

In 1999, Russia adopted new standards of radiation safety, NRB-99, corresponding to international basic safety standards. These standards recommend that the criterion for intervention following radioactive pollution incidents should be where individual doses to the population are more than 0.3mSv per year, based on a collective dose from 70 years occupancy. Based on these criteria the following areas would require remediation.

### 5.1 Areas of contaminated floodplain and riverbank

There is insufficient data to confirm the extent of contamination along the riverbanks. Samples in which contamination has been found have been taken from between 5m and 50m from the edge of the river.

The most recent data from Krasnoyarsk-26, based on a communication received in June 1998, is that for assessment purposes, the following should be assumed:

- only one bank of the river should be assumed to be contaminated;
- the width of contamination should be assumed to be 20m;
- the length of this contaminated strip should be assumed to be 1000km;
- this gives a total area of contamination of 20 km<sup>2</sup>;
- the gamma dose rate should be assumed to be 30-40 µR/hour (2.6-3.5 mSv/year);
- this is equivalent to 50-200 kBq/m<sup>2</sup> for <sup>137</sup>Cs.
- If the average contamination depth is assumed to be 0.3m, then the volume of contaminated material would amount to 6 million (6 E+06) m<sup>3</sup>.

### 5.2 Gorodskoy Island

The island is located in the Yenisei River, approximately 410 km downstream from Krasnoyarsk City at Eniseisk. Average levels of pollution at 1m depth are 1050kBq/m<sup>2</sup> for <sup>137</sup>Cs, 3.3kBq/m<sup>2</sup> for <sup>60</sup>Co and 12.5Bq/m<sup>2</sup> for <sup>152</sup>Eu. The gamma background dose is 20-60 µ R/hour (1.8-5.3 mSv/year). The total area of pollution on the island has been estimated by the Russian partners to be 10000m<sup>2</sup>. A vertical profile of contamination down through soil on the island showed contamination present to at least 0.75m depth. This gives an estimated volume of contaminated material to be at least 7500 m<sup>3</sup>.

### 5.3 Atamanov Island

Atamanov Island is located about 5km downstream of the MCC nuclear plant. If the nature and depth of contamination are assumed to be similar to that on Gorodskoy Island, and the size of Atamanov Island is estimated to be approximately one third of Gorodskoy Island, then the estimated volume of contaminated material is at least 2500 m<sup>3</sup>.

The overall options for management of the contamination are relatively simple, and include:

- Option 1: excavation of all contaminated material and its removal to approved waste treatment, storage and eventual disposal facilities;
- Option 2: in-situ confinement of the contamination in its present location, through emplacement of engineered barriers, in order to prevent further spread of contamination and to minimise doses to critical groups;
- Option 3: minimisation of dose/risk to the population through “agricultural countermeasures”, similar to those employed in areas contaminated from the Chernobyl accident;
- Option 4: removal of the population until risks to them are within acceptable limits;
- Option 5: “do nothing”.

The conclusions of the project team were that a combination of Options 1 and 5 was likely to be the most pragmatic solution.

## 6. RECOMMENDATIONS

The results of the radiological assessments show that the only significant exposures result from the occupancy and use of the contaminated floodplains, particularly if hot particles are present. Doses of several millisieverts a year could result from some of the most contaminated sites. The levels of floodplain contamination are very variable; samples taken at the same approximate location can be very substantially different. There is also no simple correlation with contamination levels and distance from the discharge.

A number of key recommendations have been made in order to understand the implications of the contaminated floodplains and islands of the Yenisei River more precisely. The recommendations include the following:

(1) Define spatial extent of floodplain contamination in a robust and systematic way

- Aerial gamma survey (<sup>137</sup>Cs)
- Ground gamma survey
- Examination of hydrological features of the river
- Evaluation of flooding scenarios and their impact
- Evaluation of alpha-contamination

(2) Identify usage of floodplain site in detail

- Land use (pasture, crop production, recreation etc)
- River use (fishing, swimming, etc.)
- Occupancy times
- Demographic data

(3) Evaluate contamination of food produced on or near contaminated floodplain sites

- Sampling and analysis programme for foods produced on, or near, contaminated floodplains
- Modelling of food-chain transfer for representative scenarios

(4) Evaluate potential impact from hot particles

- Small programme currently supported by DGXI
- Distribution of hot particles
- Analysis of hot particles
- Sources of hot particles
- Critical groups
- Risk assessment

(5) Dose measurements for critical groups

- Whole body and TLD measurement programmes
- Dose assessments for representative scenarios, including the impact of potential future flood events

(6) Evaluation of key source term data

- Understanding radwaste disposed of and stored at Krasnoyarsk-26, future operations and the final site closure strategies
- Understanding (or development) of a strategy for each site for future radwaste management, in order to minimise future releases to the biosphere

(7) Detailed evaluation of remediation strategies

- Cost per averted man-sievert
- Regulatory issues
- Pragmatic solutions which can be achieved easily, mainly with existing equipment, and at little cost

(8) Evaluate potential for re-contamination of remediated sites

- Evaluate the potential for re-contamination to occur should areas be remediated
- Identify patterns of sediment movement and deposition

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- [4] ICRP PUBLICATION 72, *Age-dependent Doses to Members of the Public from Intake of Radionuclides: Part 5-Compilation of Ingestion and Inhalation Dose Coefficients*, Annals of the ICRP, **26** (1), 1996.

#### DISCUSSION AFTER THE PRESENTATION OF J.L. SMITH-BRIGGS

**E.D. STUKIN (Russian Federation):** All available RADLEG data show that the contamination maxima in the Yenisey valley occur at depths of 30 cm. Consequently, sampling at a depth of 10 cm or less gives incorrect results.

**J.L. SMITH-BRIGGS (United Kingdom):** Contamination is present from the surface down to 50-60 cm, in places even down to 1 m. The contamination profile can be very variable; sometimes the contamination is greater at the surface and sometimes it is greater at deep levels.

**E.D. STUKIN (Russian Federation):** You spoke about aerial photography. Were you aware that the Institute of Global Climate and Ecology had carried out an aerial photographic survey of the Yenisey valley over a distance of 2000 km and across a front of 3 km?

**J.L. SMITH-BRIGGS (United Kingdom):** I was not aware of that, and I would be interested in seeing the results.

**M. GOLDMAN (USA):** What data were there for the time before 1987, when the releases were greatest?

**J.L. SMITH-BRIGGS (United Kingdom):** Measurements carried out when the reactors were operating indicated very light levels of phosphorus-32 and some other short-lived nuclides in the fish and also fairly high doses to people living in the settlements who consumed fish.

**M. GOLDMAN (USA):** Are the hot particles mentioned by you plutonium particles?

**J.L. SMITH-BRIGGS (United Kingdom):** They are mainly caesium particles, but there are also americium, plutonium and uranium particles. Most of the dose comes from <sup>137</sup>Cs.

**S.M. VAKULOVSKY (Russian Federation):** Did you estimate the doses received by people living along the river before the closure of the single-pass reactors?

**J.L. SMITH-BRIGGS (United Kingdom):** No, our study did not cover doses to earlier population groups-only current and possible future doses.

**Yu.A. IZRAEL (Russian Federation-Session Chair):** In this connection, I would draw attention to the proceedings of the International Conference on Radioactivity after Nuclear Explosions and Accidents held in Moscow from 24 to 26 April 2000, which contain reports by three institutes (one Siberian and two Moscow-based) on radioactive contamination around Krasnoyarsk-26.

# RADIOACTIVE CONTAMINATION IN THE ARCTIC — PRESENT SITUATION AND FUTURE CHALLENGES

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## *Abstract*

*There is currently a focus on radioactivity and the Arctic region. The reason for this is the high number of nuclear sources in parts of the Arctic and the vulnerability of Arctic systems to radioactive contamination. The Arctic environment is also perceived as a wilderness and the need for the protection of this wilderness against contamination is great. In 1991, the International Arctic Environmental Protection Strategy (IAEPS) was launched and the Arctic Monitoring and Assessment Programme (AMAP) established. AMAP is undertaking an assessment of the radioactive contamination of the Arctic and its radiological consequences. This paper summarises some of current knowledge about sources of radioactive contamination, vulnerability, exposure of man, and potential sources for radioactive contamination within Arctic and some views on the future needs for work concerning radioactivity in Arctic.*

## 1. INTRODUCTION

There is currently a focus on radioactivity and the Arctic region. The reason for this is probably the high number of nuclear sources in parts of the Arctic and the vulnerability of Arctic systems to radioactive contamination. The Arctic environment is also perceived as a wilderness and the need for the protection of this wilderness against contamination is great. In the last decade information has also been released concerning the nuclear situation which has caused concern in many countries. Due to such concerns, the International Arctic Environmental Protection Strategy (IAEPS) was launched in 1991 and the Arctic Monitoring and Assessment Programme (AMAP) was established. AMAP is undertaking an assessment of the radioactive contamination of the Arctic and its radiological consequences. In 1996 IAEPS became part of the Arctic Council. AMAP presented one main report in 1997 [AMAP 1997] and another in 1998 [AMAP 1998]. There are also several other national, bilateral and international programmes in existence which deal with this issue. This paper summarises some of current knowledge about sources of radioactive contamination, vulnerability, exposure of man, and potential sources for radioactive contamination within Arctic and some views on the future needs for work concerning radioactivity in Arctic.

## 2. PAST AND PRESENT RADIOACTIVE CONTAMINATION

There are three main sources of radioactive contamination in the Arctic today: global fallout from nuclear tests, releases from nuclear reprocessing plants in Western Europe and Chernobyl fallout. A total of 518 atmospheric nuclear explosions took place up to 1980. The spatial distribution of  $^{137}\text{Cs}$  of this fallout onto land is shown in Figure 1. [Strand et al. 1998].

The major source of radiocaesium to the Arctic from West European nuclear reprocessing plants is Sellafield in the UK. Releases started in 1952 and continue up to the present day with major releases having occurred between 1974 and 1982. The releases of  $^{99}\text{Tc}$  and  $^{137}\text{Cs}$  from Sellafield and the subsequent contamination levels in the Barents Sea are shown in Figure 2. Releases of  $^{99}\text{Tc}$  increased in 1994 from the reprocessing plant at Sellafield and have recently

led to increased concentrations of  $^{99}\text{Tc}$  in northern marine environments and further into the Arctic Seas.

Following the Chernobyl accident in 1986, a major release of radioactive material spread over large areas in Europe, with the radioactive cloud and fallout also reaching Arctic areas, mainly the North West Russia and Northern Fenno-Scandia, with deposition levels in the order of 1-5 kBq/m $^2$   $^{137}\text{Cs}$ . Deposition levels in the order of 10-200 kBq/m $^2$  were, however, observed just south of the Polar Circle. Radioactive contamination of the North and Baltic Seas provides continuously indirect contamination to the Arctic marine environment via transport pathways along the Norwegian coast.

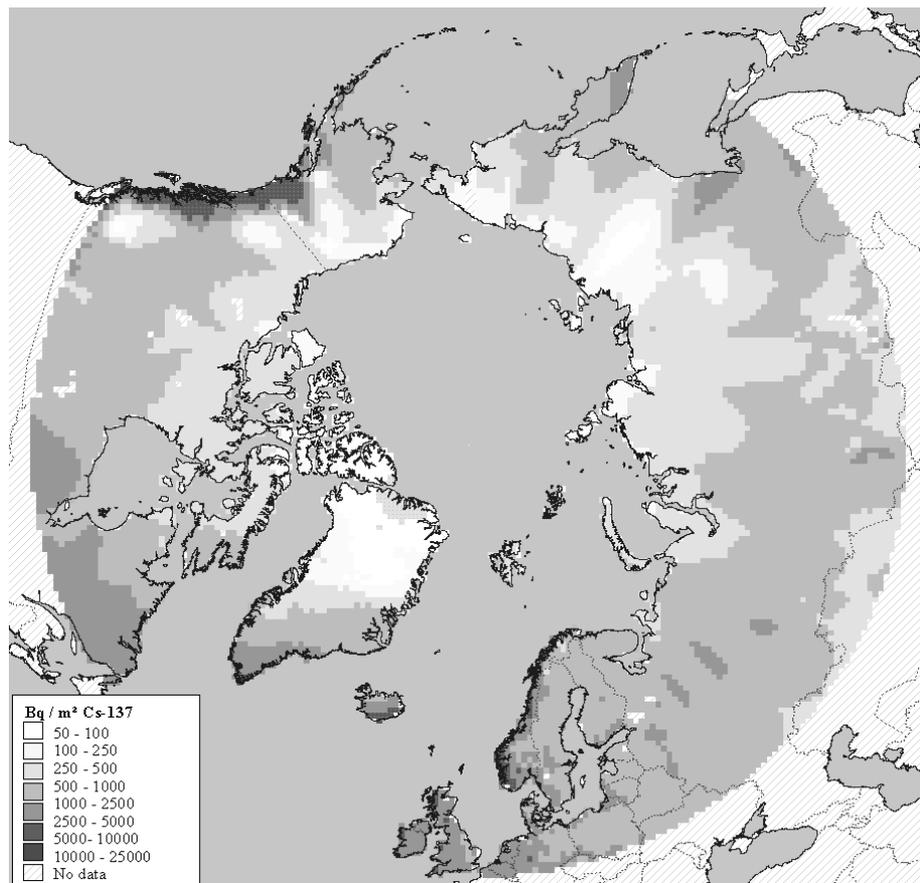


FIG. 1. Estimated spatial distribution of  $^{137}\text{Cs}$  fallout from nuclear weapons testing.

There do also exist several other sources which have given small or only localized contamination such as the Thule accident in Greenland, where an American plane carrying nuclear bombs was involved, from the accident with the Komsomolets submarine in the Norwegian Sea, waste storage sites and from dumping of radioactive waste in the sea. Former releases from the nuclear reprocessing plant at Mayak in the Urals, even if it has not contaminated Arctic areas to a substantial degree with earlier discharges, is today a considerable source contained in the environment and, through the potential for transport of radionuclides via the Ob River, is a source to be considered as a threat to the Arctic environment. The releases of radioactive caesium and strontium from Mayak were nearly 100 times higher than the releases of these radionuclides from Chernobyl. Today the radionuclide inventory is stored mainly in lakes and reservoirs, with potential risk for leakage to the rivers and into the Northern Seas.

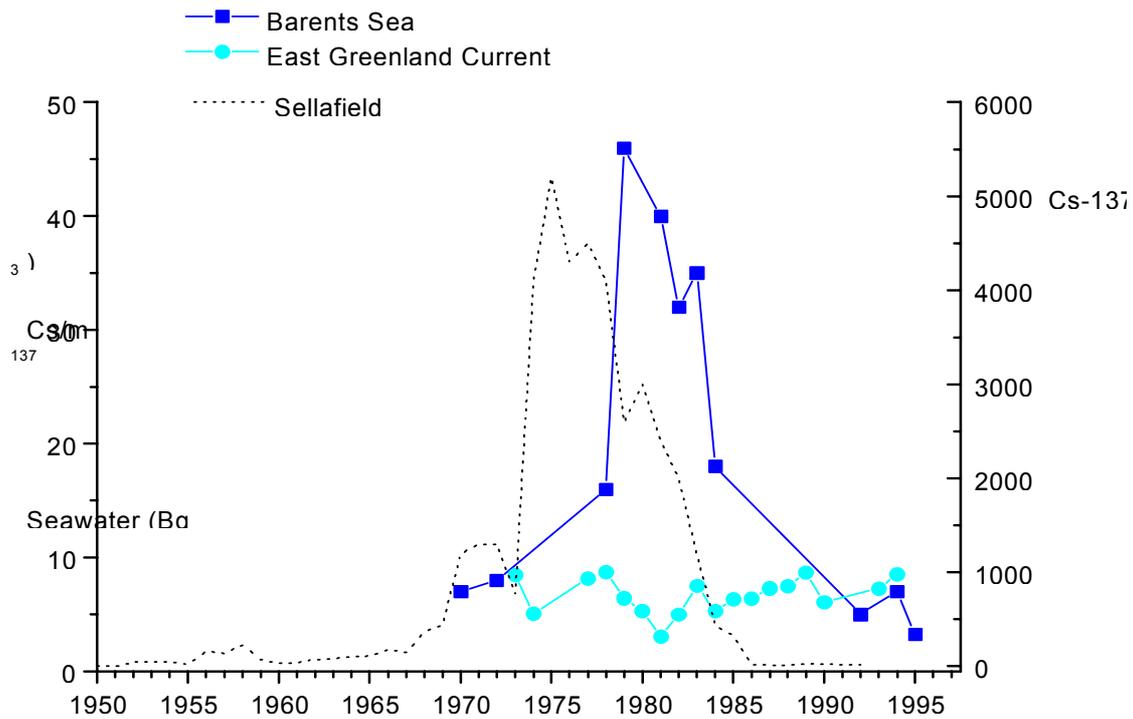


FIG. 2. The releases of  $^{137}\text{Cs}$  from Sellafield and the subsequent contamination levels in the Barents Sea and East Greenland Current.

### 3. EXPOSURE OF MAN FROM PAST RELEASE

The largest contribution to radiation doses from radioactive contamination to the population in northern areas has come from fallout from nuclear tests in the 1950's and '60's. In some areas also the Chernobyl fallout has contributed significantly to the total dose. The population in northern areas is exposed to a dose, from radioactive contamination, which is about 5 times higher than the dose for people in more temperate areas with the same deposition. This illustrates the higher transfer and vulnerability of the Arctic areas from  $^{137}\text{Cs}$  contamination.

Indigenous people in the Arctic, mainly living off traditional food products from mountain, forest and lakes, e.g. reindeer-herding Saamis, receive the highest doses from radioactive contamination.

Radiation doses to both the average population and the special groups depend mainly on intake of locally-produced terrestrial products. In comparison, Arctic population groups with mainly marine products in their diet, receive relatively low doses. The most exposed population groups in the Arctic can on average receive up to 50 times higher individual doses than members of the average population.

The intake by the general Arctic population of radiocaesium from different foodstuffs during the period 1990-1994 in the 8 Arctic countries are shown in Figure 3 [Strand et al. 1998]. For the average Arctic populations, a range of different food products contribute to the total  $^{137}\text{Cs}$  intake. Canada is an exception, with the domination of reindeer meat consumption in the intake of radiocaesium.

The regional effect on dietary preferences is also clearly shown in Figure 3. For example, goat cheese is only an important source for radiocesium intake in Norway. Mushrooms are important in Sweden, Finland and Russia. However, there is a considerable lack in knowledge concerning the importance of natural food products in the transfer of radionuclides to man. In contrast, people who consume only marine products, such as marine fish and mammals, receive doses that are at least an order of magnitude lower than people consuming terrestrial products such as reindeer/caribou meat, freshwater fish and mushrooms. The intake for the selected groups were considerably higher as shown in Figure 4. The major contributor to the intake for these groups was reindeer meat.

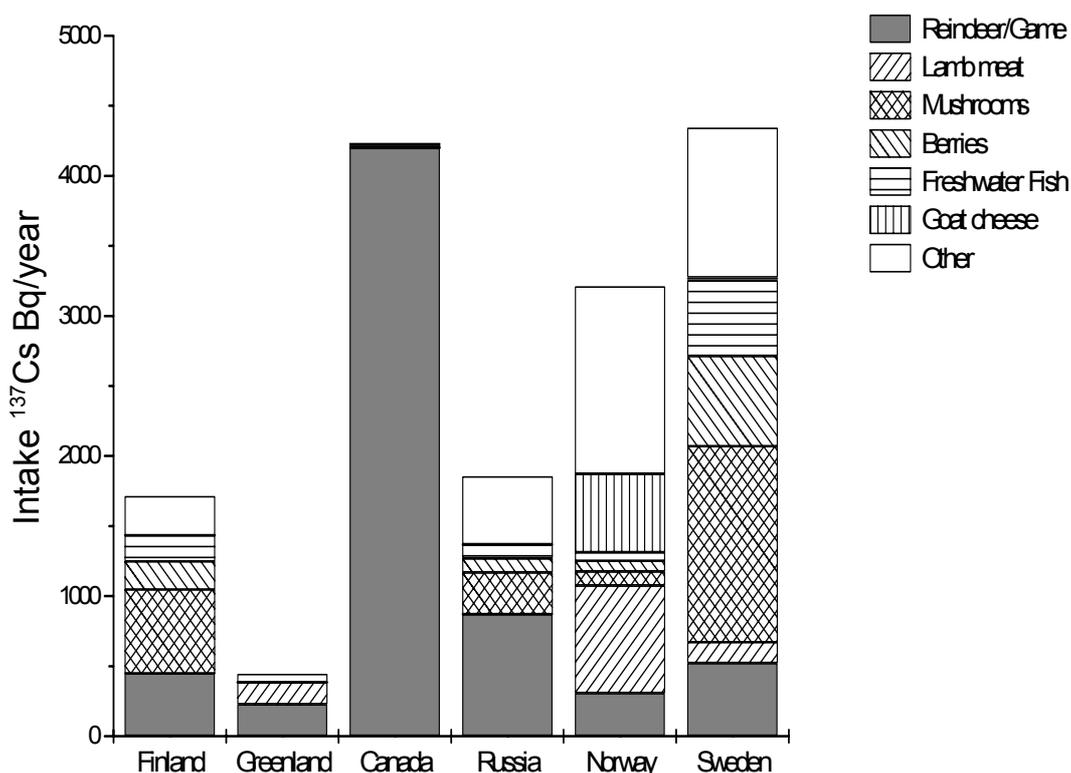


FIG. 3. Yearly intakes by the average populations during 1990-1994.

The major contribution ( $\gg 15\ 000\ \text{manSv}$ ) to the collective dose to Arctic populations results from fallout from nuclear weapons testing, with a range of individual dose commitments between 1 and 150 mSv [Strand et al. 1998]. The second most important contribution ( $\gg 500\ \text{manSv}$ ) to collective dose within the Arctic derives from the Chernobyl accident, with individual dose commitments normally in the range 1 to 50 mSv. Individual annual doses to the most exposed residents of the Arctic from Chernobyl releases, however, could be approximately 10 to 20 mSv/y in the most affected areas. Countermeasures introduced by some countries following the Chernobyl accident and, in some cases, maintained to the present day, have resulted in reduced individual doses and dose commitments. Releases from the Sellafield fuel reprocessing plant provide the third most important collective dose contribution ( $\gg 50\ \text{manSv}$ ) with a relatively small contribution to individual dose (*i.e.* in the range 0 to 0.05 mSv) [CEC 1990].

The total exposure of man from the dumping of nuclear waste is very small even taking into consideration the potential future dose. Smaller-scale releases from accidents in military operations, such as those in northern Russia, the plutonium spill at Thule and the loss of the

*Komsomolets* submarine in the Norwegian Sea, have resulted in no significant increases in radiation exposures to the Arctic populations. For other releases to the environment, such as from the Mayak reprocessing plant, it has been difficult to assess the collective or individual doses to Arctic populations from the releases especially, before 1960.

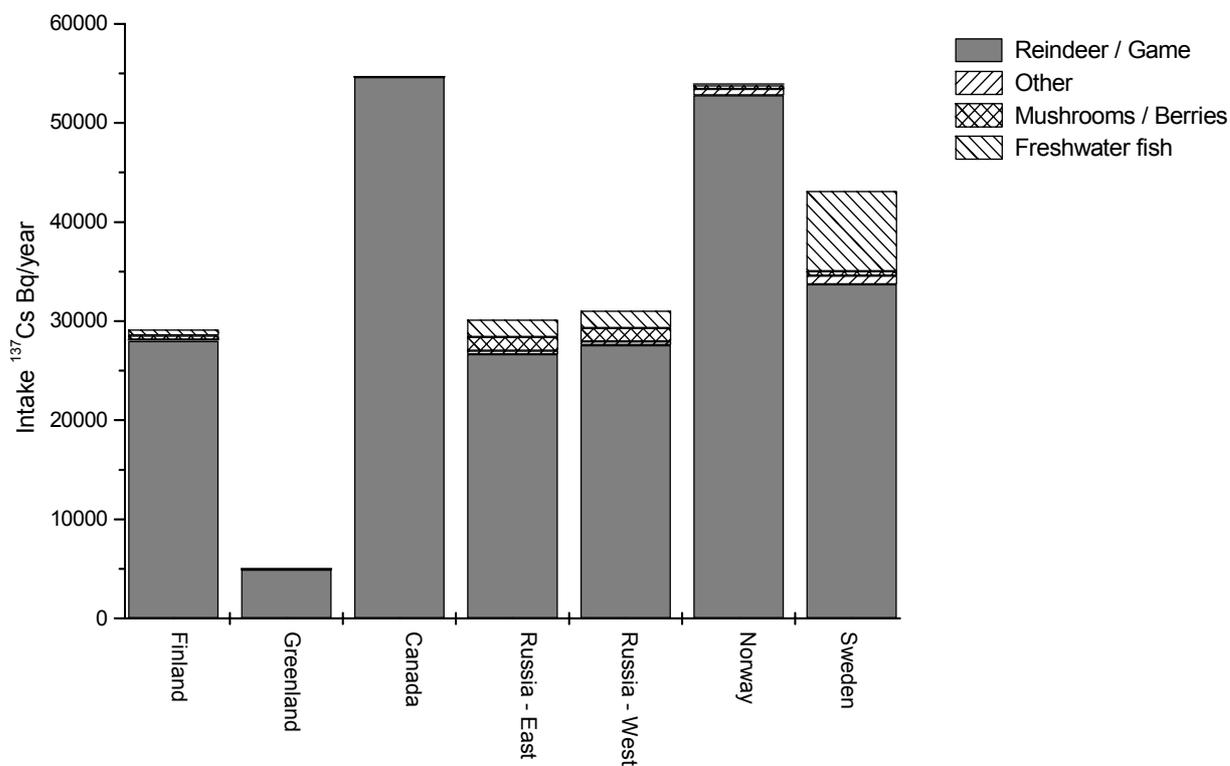


FIG. 4. Yearly intakes by the selected indigenous groups, during 1990-1994.

#### 4. POTENTIAL THREATS TO ARCTIC AND FUTURE NEED FOR RISK AND IMPACT ASSESSMENT

The concentration and number of nuclear installations and the potential for releases cause concern, especially since the vulnerability of Arctic populations is much greater than for populations in temperate areas due to the importance of terrestrial semi-natural exposure pathways. The largest threat to the environment and the population in the Arctic today is connected to potential accidents in nuclear power plants, during handling and storage of nuclear weapons, decommissioning and refuelling of nuclear powered vessels and during storage of radioactive waste.

The future work should concentrate on an assessment of the possible consequences of potential major radiation accidents or releases in the Arctic terrestrial or aquatic environments and their vicinity. Knowledge gaps relating to important pathways of human exposure and environmental contamination in the Arctic, such as long-term migration of radionuclides, changes with time in characteristics of the diet of different Arctic population groups, reasons for the high variability in levels determined in different foodstuffs need to be addressed. To fulfil some of these gaps, additional experimental, surveys and modelling work will be needed. High priority should be given to studying the site-specific vulnerability of particular Arctic regions and communities as an essential basis for the prediction of consequences of

potential radioactive contamination. Radiation monitoring in the Arctic environment should be continued, in order to acquire both spatial and temporal information as input data for assessment. Finally, the need for developing a system for assessing the consequence of radiation exposure for Arctic flora and fauna has a high priority. There has historically been a focus on the consequences on peoples' health, not the effect on the environment itself, in assessing the impact of radioactive contamination. This work needs collaboration at an international level and, with this in mind, AMAP and IUR will work together on this topic.

***Risk Management-need for closer links between Risk Assessment and Action Programs.*** It is imperative that action is based on risk and impact assessments and furthermore that the results of such actions are reassessed. Currently, communication and interaction is poor between the existing Risk and Impact assessment Programs devised to assess and monitor contamination in the Arctic and the Action Programs tasked to devise strategies and respond to existing radioactive contamination sources by implementing short-or long-term solutions. It is vital to bridge this gap and foster an interdependence between the Risk Assessment and Practical Programs to improve monitoring, response strategies and the implementation of action plans.

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## DISCUSSION AFTER THE PRESENTATION OF P. STRAND

**E.D. STUKIN (Russian Federation):** You said that two of your colleagues are working on the platform above the “Kursk” submarine. What can you say about the radioactivity levels due to the submarine’s reactor?

**P. STRAND (Norway):** They have not detected any radioactivity that could be ascribed to the reactor. The data obtained so far do not suggest that the reactor is leaking. It looks as if the reactor safety system worked well and that the reactor was shut down properly.

**YU. A. IZRAEL (Russian Federation-Session Chair):** That conclusion is supported by measurements which were carried out by specialists from Russia’s Institute of Global Climate and Ecology after the “Kursk” accident and the results of which were included in the proceedings of the International Conference on Radioactivity after Nuclear Explosion and Accidents held in Moscow last April.

**V. NOVIKOV (International Institute of Applied Systems Analysis):** Could you give any examples of cases where the countermeasures which were taken resulted in an increase-rather than a decrease-of risk?

**P. STRAND (Norway):** No, I cannot. It is simply a principle that countermeasures should not be taken if they will result in a risk increase. For that reason, there are cases where radioactive waste dumped at sea has been left where it is.

# A SUMMARY OF ESTIMATED DOSES TO MEMBERS OF THE PUBLIC FROM ATMOSPHERIC NUCLEAR TESTS AT THE NEVADA TEST SITE

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## ***Abstract***

*This paper discusses estimates of radiation dose to representative members of the public of the United States (U.S.) from atmospheric nuclear tests conducted from 1951 through 1962 at the Nevada Test Site. The estimates provided here summarize five studies conducted over the past two decades. From those studies, an estimate of the average deposition of  $^{137}\text{Cs}$  within each of the more than 3,000 counties across the country has been derived as well as doses to representative persons in each county and to specific subpopulations. The years of the largest contributions to the collective external dose were 1952, 1953, and 1957. Those years accounted for about 70% of the 84,000 person-Gy received by the U.S. public. Irradiation of the thyroid gland of members of the U.S. public was also a consequence of dispersion of radioiodine in the fallout. Thyroid doses varied by location and by birth year. The population weighted thyroid dose for a child born in 1951 and for an adult in 1951 were 30 and 5 mGy, respectively. Maps are provided to show the geographic distribution of  $^{137}\text{Cs}$  as well as the average thyroid dose received in each county from the Nevada tests.*

## 1. INTRODUCTION

The United States began conducting nuclear tests above ground at the Nevada Test Site (NTS) on January 27, 1951. Atmospheric testing continued intermittently until August 5, 1963. During that period, 119 tests were conducted, most of them aboveground. The total nuclear yield of these explosions was approximately one megaton of TNT-equivalent explosive energy.

Beginning in the early 1980s and continuing until 1997, four dose reconstruction studies related to exposure from the NTS were completed. Presently, a fifth study is in progress. The mandate for these studies has come about largely as a result of interest by the public and the U.S. Congress. These studies vary considerably in scope, design, and purpose. All five of these studies will be briefly discussed and a summary of the estimated doses received by members of the public will be presented.

The individual studies are known as: 1) the ORERP study of the U.S. Department of Energy; 2) the Utah Leukaemia Case-Control study; 3) the Utah Thyroid Cohort Study; 4) the National Cancer Institute  $^{131}\text{I}$  fallout study; and 5) the Feasibility Study on fallout exposures from NTS and global fallout being jointly conducted by the Centres for Disease Control and Prevention (CDC) and the National Cancer Institute (NCI). The first three studies were concerned with

doses received by “local” populations (less than 800 km from the NTS), while the fourth and the fifth studies deal with the estimation of doses received by populations across the continental United States. The fourth study is limited to the estimation of thyroid doses from  $^{131}\text{I}$  while the fifth study, now in progress, includes all other important radionuclides present in fallout and all other organs and tissues. In the first four of these studies, uncertainty estimates were attached to the calculated doses.

## 2. SUMMARY OF STUDIES AND ESTIMATED DOSES

Testing of nuclear weapons in Nevada resulted in the release of fission and activation products to the atmosphere that were dispersed across the U.S. in the form of gases and fallout particles. About 6 PBq of  $^{137}\text{Cs}$  was released from NTS tests; about one-third or 2 PBq was deposited in the U.S. The years of 1952, 1953, 1955, and 1957 were years of particularly large releases and depositions. The time-dependence of the  $^{137}\text{Cs}$  deposition in the U.S. from the NTS is shown in Figure 1 and the geographic pattern of deposition is shown in Figure 2.

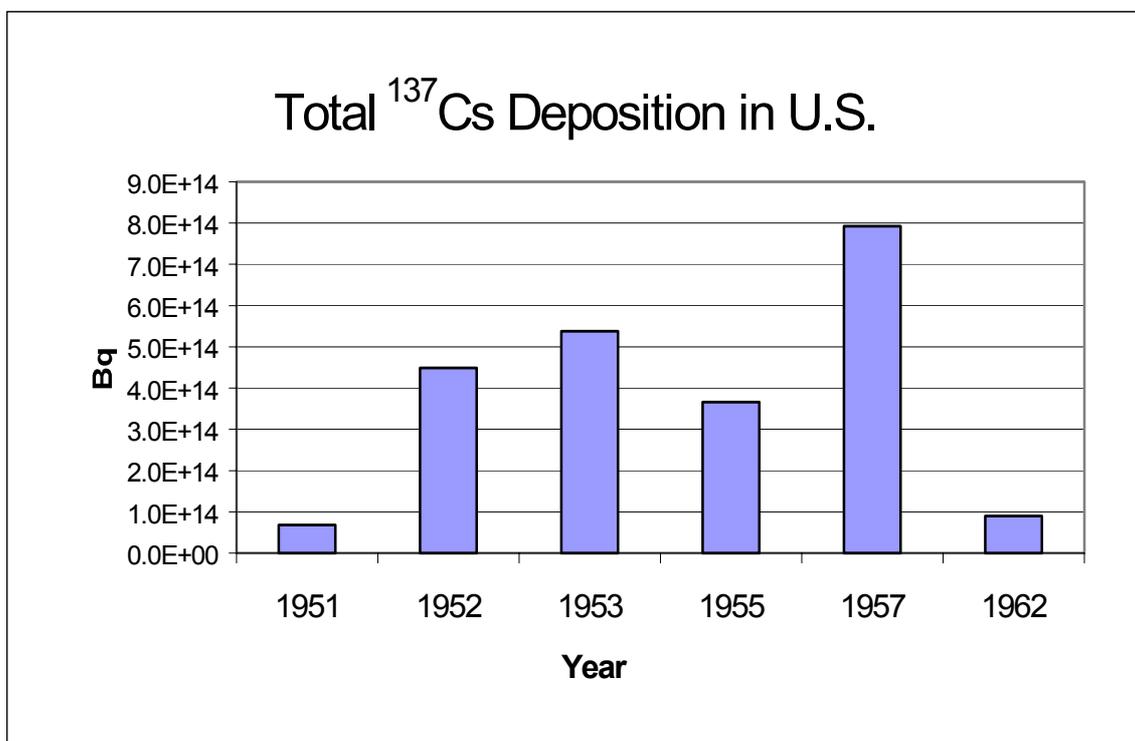


FIG 1. Total  $^{137}\text{Cs}$  (Bq) deposited in the U.S. from NTS tests as a function of year of testing.

Information released by the U.S. Government about the release of  $^{137}\text{Cs}$ ,  $^{131}\text{I}$  and other radionuclides has engendered public and Congressional interest in the effects of fallout on the health of Americans. The various efforts at dose reconstruction in the U.S. related to Nevada weapons testing have taken place, in part, as a result of such interest, though the studies described here fulfilled several different objectives. The five studies varied primarily according to the degree of methodological development and dose estimation for representative persons versus attention to estimating doses to real individuals in the population.

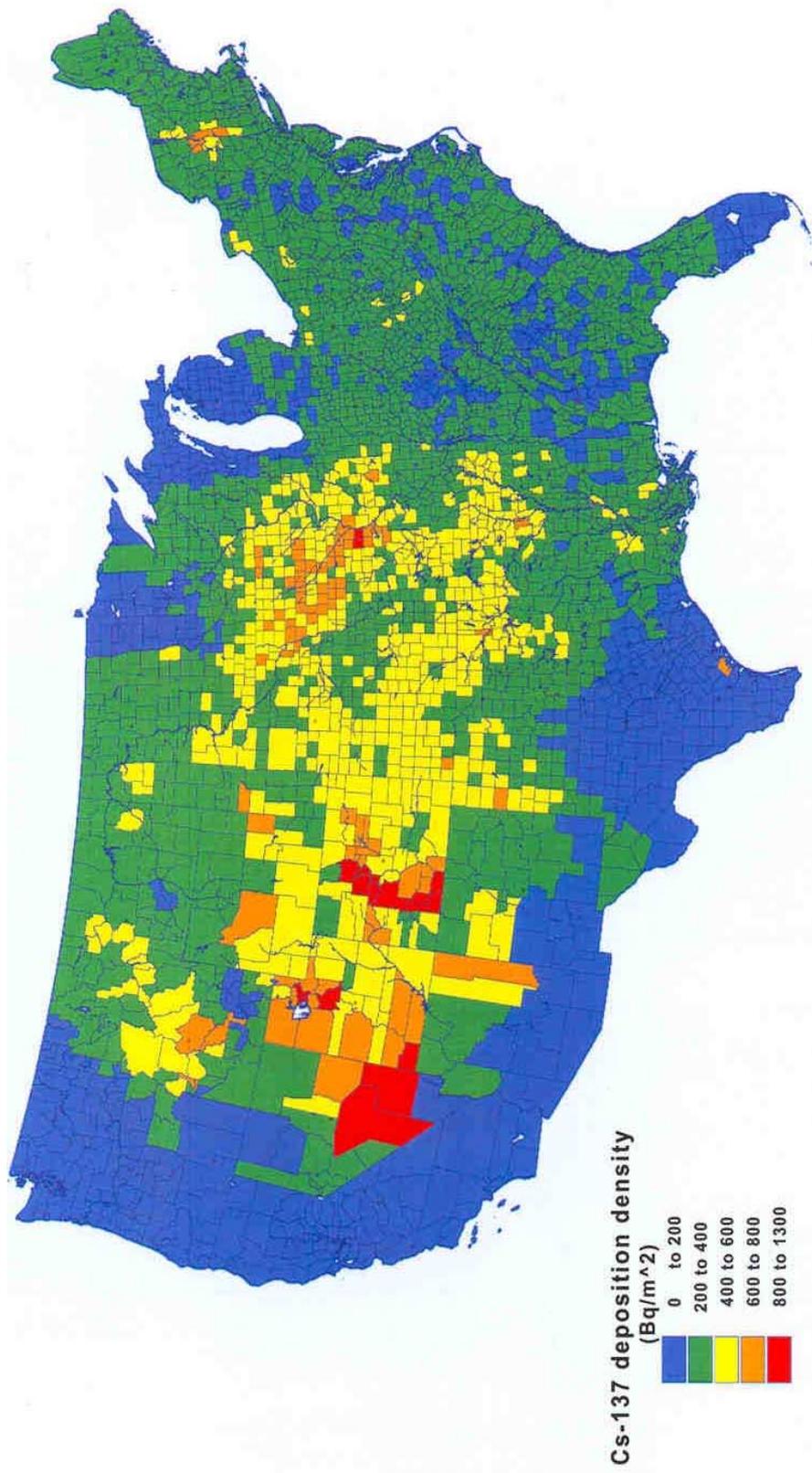


FIG. 2. Map of county estimates of <sup>137</sup>Cs deposition density (Bq/m<sup>2</sup>) from all tests conducted at the Nevada Test Site.

### 3. THE ORERP STUDY

The Off-Site Radiation Exposure Review Project (ORERP) sponsored by the U.S. Department of Energy was the first major study to address the necessary methodology needed to estimate credible doses to the U.S. public from NTS fallout at a variety of locations. The project, established in the late 1970s, utilized expertise of scientists from a variety of organizations including government laboratories, government agencies, universities, etc. Considerable new methodology developed by the ORERP later contributed to the success of the other studies to be described. In particular, the ORERP developed tables of factors to convert exposure rates following deposition of fallout to relative amounts of all important fission and activation products (Hicks 1981). These detailed tables were calculated for each individual nuclear test.

In addition, the ORERP developed computer models to estimate both external and internal dose (Henderson and Smale 1990; Whicker and Kirchner 1987). The latter modelling effort was particularly innovative in that it described different lifestyle scenarios, different pathways of exposure, and incorporated parameter values for lifestyles, age-dependence, etc. The ORERP also developed databases of deposition densities at many locations in the western United States (Beck 1984). The ORERP computed a variety of different dose estimates, pertaining mainly to collective dose and for representative persons in the western states. The reader is referred to Anspaugh et al. (1986, 1990), Whicker et al. (1996), and to Bouville (1996). ORERP estimates of dose following test event HARRY are provided in Table 1 below. The location, St. George, UT, is downwind (east) of the NTS.

TABLE 1. ORERP STUDY: ESTIMATES OF INTERNAL AND EXTERNAL DOSES IN ST.GEORGE, UT FROM TEST EVENT HARRY (19 May 1953)

Organ/tissue	Infant dose (mGy)	Adult dose (mGy)
Internal irradiation		
Thyroid	840	51
Lower intestine	large 25	5.0
Upper intestine	large 8.8	2.0
Bone surfaces	7.6	1.1
Total Body	1.1	0.48
External irradiation		
Total Body	~10	~10

### 4. THE UNIVERSITY OF UTAH LEUKAEMIA CASE-CONTROL STUDY

The two studies, conducted by the University of Utah and funded by the U.S. National Cancer Institute, were also in response to public and Congressional interest. The Utah studies differed considerably from that of the ORERP, however, in that they were epidemiological investigations whose purposes were to test hypotheses posed by earlier investigators concerning possible health effects among residents of Utah.

The study of leukaemia among Utah residents used methods and data from the ORERP study as well as personal residence histories of all persons dying in Utah with leukaemia after 1 November 1958 and matched control subjects. Residence histories were obtained from records of the Church of Jesus Christ of Latter-day Saints and other sources. Because the tissue of interest was the active bone marrow, external irradiation by fallout was the primary contributor to the radiation exposure of the subjects. Previous investigations show that internal dose from fallout was only a small contribution. Methodology for the leukaemia study and estimated doses were discussed by Simon et al. (1995) and by Lloyd et al. (1990), and are summarized in Table 2 below. Results of the epidemiological analysis were reported in Stevens et al. (1990).

TABLE 2. UTAH CASE-CONTROL STUDY; SUMMARY OF ACTIVE BONE MARROW DOSES (MGY) FOR 6,507 SUBJECTS (Simon et al. 1995)

	<b>CASES</b>	<b>CONTROLS</b>	<b>OVERALL</b>
Mean	2.9	2.7	2.8
Median	3.2	3.1	3.2
Minimum	0	0	0
Maximum	26	29	29

## 5. THE UNIVERSITY OF UTAH THYROID COHORT STUDY

The Utah study of thyroid disease also used data and methodology for estimating deposition from the ORERP. However, independent methods for estimating individual doses and uncertainty were developed specifically for this study by the Utah investigators (see Simon et al. 1990). Doses were estimated for 3,545 subjects of which 3,122 were re-examined for evidence of thyroid disease, both benign and cancerous.

The dosimetry methodology accounted for all major pathways of exposure including external and internal irradiation (ingestion and inhalation). The primary source of  $^{131}\text{I}$  to the study subjects was consumption of milk though fresh leafy vegetables were also included in the calculations. The source of milk (i.e., commercial or backyard cow) was important because of differences in feeding habits and in time delays between production and consumption. In addition, whether or not the subjects drank goat's milk was important because of the higher transfer factors from ground deposition to milk contamination for goats. Milk consumption rates for each study subject during the years of fallout deposition were determined from interviews with the parents of each subject.

Individual doses ranged from near zero to 4600 mGy. Average thyroid doses in the three counties where subjects lived at the time of exposure were 72 mGy (Washington County, Utah), 3.6 mGy (Graham County, Arizona) and 28 mGy (Lincoln County, Nevada) (see Table 3).

Doses to milk drinkers greatly exceeded those to subjects that did not drink milk. Similarly, consumers of goat's milk had higher predicted doses than those to subjects drinking only cow's milk (see Table 4).

A summary of the dosimetry findings for the Utah thyroid cohort study is presented in Till et al. (1995) and findings of the epidemiological analysis of the Utah thyroid cohort study are presented in Kerber et al. (1993) and in Stevens et al. (1992).

TABLE 3. UTAH THYROID COHORT STUDY: SUMMARY OF THYROID DOSES (mGy) FROM UTAH THYROID COHORT STUDY (Till et al. 1995)

	Washington County, Utah	Graham County, Arizona	Lincoln County, Nevada	Overall
Number of subjects	1896	1369	280	3545
Mean dose	170	13	50	98
Median dose	72	3.6	28	25
Minimum dose	0	0	0	0
Maximum dose	4600	450	840	4600
Mean GSD <sup>a</sup>	2.7	3.0	2.7	2.8

<sup>a</sup>Geometric Standard Deviation.

TABLE 4. COMPARISON OF THYROID DOSES (mGy) BETWEEN MILK DRINKERS AND NON-MILK DRINKERS (Till et al. 1995)

	Non-milk drinkers	Cow's Goat's drinkers	and/or milk	Goat's drinkers	milk
Number of subjects	120	3337		155	
Mean dose	12	100		300	
Median dose	0.5	30		39	
Maximum dose	25	4600		4600	

## 6. THE NATIONAL CANCER INSTITUTE <sup>131</sup>I FALLOUT STUDY

The study of the National Cancer Institute (NCI 1997) was the first study to estimate thyroid doses from NTS fallout to representative persons in all 3,000+ counties of the United States. The NCI study also used certain methodologies developed by the ORERP though the NCI study relied heavily on extensive review of deposition data and interpolation strategies of deposition information derived from the gummed film monitoring network of the Atomic Energy Commission's Health and Safety Laboratory in New York City (Beck et al. 1990). All major pathways of exposure were considered as well as a set of consumption and age-dependent scenarios (Bouville et al. 1990).

The collective thyroid dose to the population of the contiguous United States from all atmospheric bomb tests detonated at the Nevada Test Site was estimated to be about  $4 \times 10^6$  person Gy, corresponding to a per capita thyroid dose of about 20 mGy. The greatest contributions to the collective thyroid dose are estimated to have been due to the Plumbbob test series in 1957, the Tumbler-Snapper test series in 1952, and the Upshot-Knothole test series in 1953. Thyroid doses to representative individuals were found to vary mainly according to age, origin and consumption rate of milk, as well as to place of residence at the time of the tests.

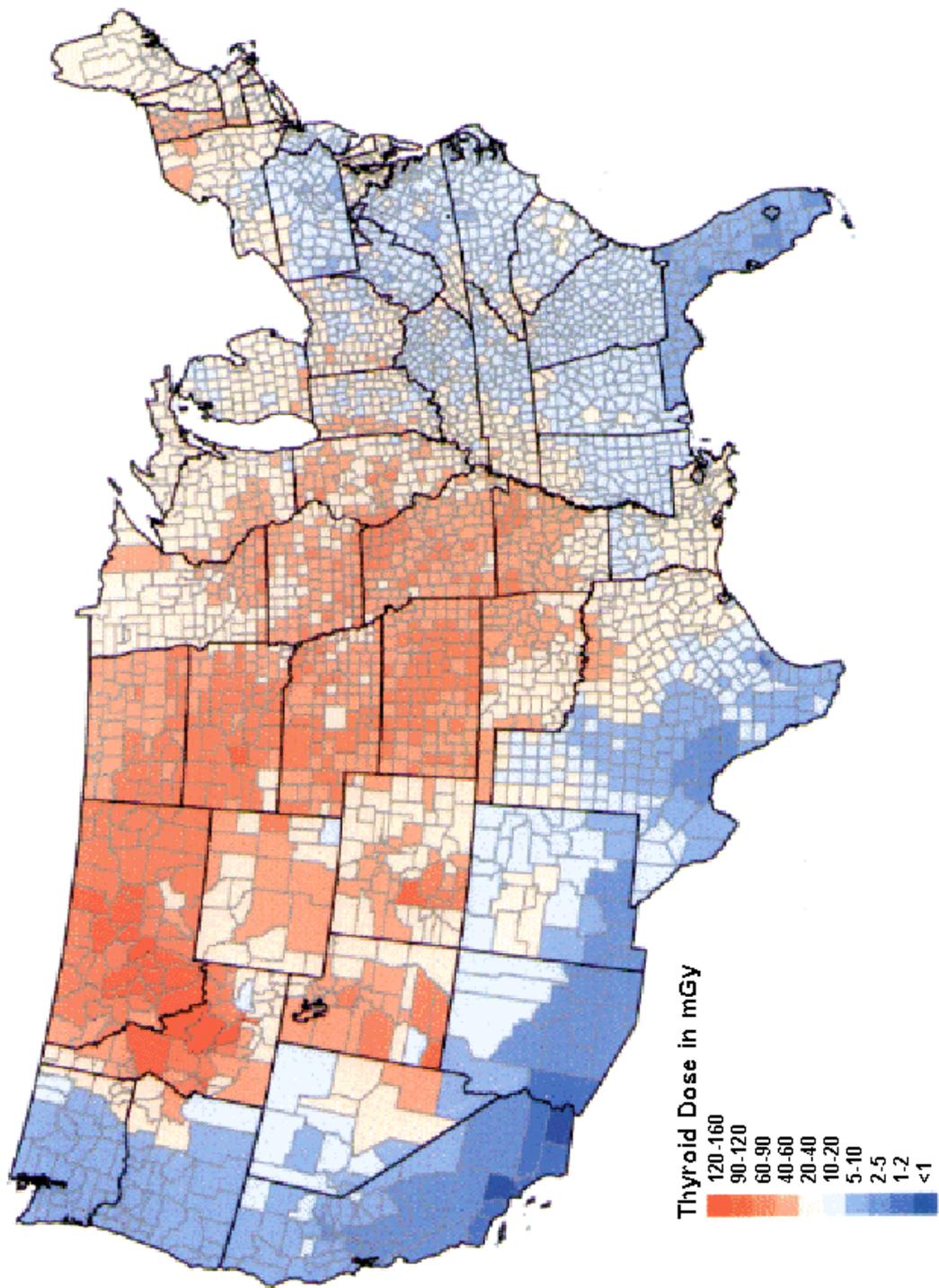


FIG. 3. Map of county estimates of thyroid dose (mGy) from  $^{131}\text{I}$  released from all tests conducted at the Nevada Test Site, averaged over all age groups.

Interested readers are referred to the website of the National Cancer Institute (<http://rex.nci.nih.gov/>, see "About Radiation Fallout") for the entire report and a series of colour coded maps giving the results of the extensive calculations. Figure 3 presents a map showing the estimated thyroid dose received in each county averaged over all groups.

Because the risk of thyroid cancer following exposure to  $^{131}\text{I}$  is not known with certainty, the expected number of excess cases had to be predicted based on what is known about childhood exposure to external radiation. The predicted number of lifetime excess thyroid cancer cases associated with the exposure from testing is dependent on the relative biological effectiveness of  $^{131}\text{I}$ , the effects of gender and age at exposure on radiation risk, the risk coefficient, and the statistical model used. The 95% confidence limits on the predicted number of excess thyroid cancer cases resulting from NTS exposures have been estimated to be 11,300 and 212,000 (IOM and NRC 1999). It is estimated that over one-third of the predicted cases have already been diagnosed.

## 7. THE CDC/NCI FEASIBILITY STUDY ON ESTIMATING TOTAL DOSES FROM NTS FALLOUT

In October 1998, the United States Congress directed the Department of Health and Human Services to conduct a study of health consequences to the American people from radioactive fallout resulting from nuclear weapons tests conducted by the United States and other nations. Following this mandate, the Centres for Disease Control and Prevention (CDC) and the NCI collaborated to determine feasibility and to produce preliminary dose estimates.

The CDC/NCI feasibility study, now being completed, has crudely estimated doses resulting from the tests conducted at the NTS and considered, at least in a preliminary fashion, all important radionuclides and radiation exposures resulting from external irradiation and from consumption of contaminated foodstuffs. In this framework, preliminary doses to representative persons in all counties of the continental United States have been estimated for the first time for a set of the most important radionuclides produced as a result of nuclear weapons testing between 1951 and 1963 by the U.S. and other nations. Only preliminary doses resulting from NTS fallout are discussed here. They are provided for illustration purposes of the type of results obtained in the study, as the extensive report prepared by CDC and NCI is currently under scientific review. It is stressed that the numerical values of the dose estimates may change before the publication of the report.

Doses were calculated for 61 of the most significant events that occurred at the NTS during 1951, 1952, 1953, 1955, 1957, and 1957. Most of the radiation exposure from external irradiation was from gamma rays emitted by fission products deposited on the ground; the external dose was mainly due to short-lived radionuclides and occurred within the first 3 weeks after each test. On an annual basis, the time dependence of doses to the U.S. population followed the same basic time-pattern as the deposition of  $^{137}\text{Cs}$  (see Figure 1 and Table 5). The total collective external dose was about 84,000 person-Gy corresponding to an average external dose from all NTS tests of about 0.5 mGy, equivalent to about one year of external radiation exposure from natural background. Residents in the states immediately downwind from the NTS received much higher exposures than the average while people in the western and north-western U.S. and some areas of the Midwest and of the Southeast received much less than the average. The actual dose received by any individual depended on the fraction of time he/she spent outdoors during the first few weeks after fallout and the degree of shielding

provided by his/her dwelling. The most exposed individuals at any particular location would have been outdoor workers or others who spent most of their day outdoors.

Doses from internal irradiation were calculated to a variety of organs and tissues, in particular, red bone marrow and the thyroid. Table 6 summarizes the population-weighted dose to these tissues for those citizens who were adults at the time of fallout and for those were born 1 January, 1951, that is, just before the first tests that were conducted at the NTS.

Internal radiation doses were dominated by the thyroid dose, the most important contributor being  $^{131}\text{I}$ . Other than the doses from  $^{131}\text{I}$  to the thyroid, doses to other organs are much smaller and are less than the dose that was estimated to have resulted from external exposure to NTS fallout. This included liver, red bone marrow, colon, etc. The more important contributors to internal dose from NTS fallout, other than  $^{131}\text{I}$ , were the short-lived radionuclides  $^{89}\text{Sr}$ , and  $^{140}\text{Ba}$ . The population weighted effective dose received by the U.S. population from ingestion was estimated to be about 0.3 mSv.

TABLE 5. COLLECTIVE EXTERNAL DOSE AND COUNTRY-AVERAGE DOSE FROM NTS FALLOUT AS A FUNCTION OF YEAR OF TESTING

YEAR	Test Series	Cumulative Collective Dose ( $10^3$ Person- Gy)	Country-Average Dose (mGy)
1951	Ranger and Buster- Jangle	6.8	0.039
1952	Tumbler- Snapper	16	0.093
1953	Upshot- Knothole	20	0.12
1955	Teapot	13	0.072
1957	Plumbbob	23	0.12
1962	Storax	5.0	0.029
Total NTS		84	~0.5

TABLE 6. POPULATION-WEIGHTED RED MARROW AND THYROID DOSES FROM ALL NTS TESTS (MGY).

Population Subgroup	Organ Dose (mGy)	
	Red marrow	Thyroid
Child born in 1951	0.12	30
Adult in 1951	0.1	5

The preliminary results provided here establish that a reconstruction of external and internal (i.e., ingestion) doses from all important radionuclides NTS fallout is feasible.

## CONCLUDING REMARKS

The five dose reconstruction studies described here have added considerable knowledge about the doses received by the U.S. public as a consequence of nuclear testing in Nevada. These dose estimates have enabled the health impacts to be estimated (e.g., for thyroid cancer) to a limited degree. There are, however, still gaps in knowledge with respect to estimating doses to the U.S. population from NTS fallout. These include lack of knowledge about fission neutron energy spectra and  $^{137}\text{Cs}/\text{Pu}$  ratios as well as information about individual's habits and lifestyle. In addition, estimated doses could be improved by acquiring additional data to validate dose estimates. Validation data could take the form of deposition densities, concentrations of radionuclides (in particular  $^{131}\text{I}$ ) in food products or in thyroid glands. A higher resolution grid of historical precipitation data could be used to further improve the interpolations. Finally, dose estimates are yet to be completed for Hawaii and Alaska and for the territories. Correcting these various deficiencies in availability of data and completing the identified refinements in models remain as challenges within the immediate future.

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## DISCUSSION AFTER THE PRESENTATION OF S.L. SIMON

**E.D. STUKIN (Russian Federation):** The Nevada test site and the Semipalatinsk test site are of roughly the same size, and there were other similarities (for example, the height of the towers at the top of which the nuclear-weapon-related devices were installed was about the same). However, the individual and collective doses due to the tests carried out at the Semipalatinsk test site are 20-25 times higher than those due to the tests carried out in Nevada, and those high doses have given rise to great concern among people living in the Altay region. To what do you attribute the higher doses resulting from the tests carried out at the Semipalatinsk test site?

**S.L. SIMON (USA):** The only explanation I can offer for the higher individual and collective doses is differences in living habits; people in the Altay region probably consume more locally produced food and milk than people living near the Nevada test site. However, I understood that there is some controversy about how high the individual and collective doses are near the Semipalatinsk test site, and in my view it is important to resolve that issue.

**B.I. OGORODNIKOV (Russian Federation):** When did the United States start taking meteorological conditions into account in order to reduce the doses from nuclear weapon test fallout?

**S.L. SIMON (USA):** I don't know whether there were ever any major efforts to reduce such doses. With the great importance attached to nuclear weapons development at the time, our Government did not acknowledge the resulting contamination-and restrictions on radiation measurements meant that it was a long time before the general public became aware of the contamination levels.

# MODEL OF EXTERNAL EXPOSURE OF POPULATION LIVING IN THE AREAS SUBJECTED TO RADIOACTIVE CONTAMINATION

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## *Abstract*

*In the paper, we formulated the general approach to assessment of external doses to population living in contaminated areas (the model equation and the set of parameters). The model parameters were assessed on the basis of results of monitoring in the environment, phantom experiments, and social and demographic information obtained on the contaminated areas. Verification of model assessments performed by comparison with measurement results of individual external doses in inhabitants within the thermoluminescent dosimetry method have shown that differences in dose assessments within both methods does not exceed 1.5 times at a confidence level of 95%. In the paper, we present the results illustrating specific features of external dose formation in population living in the areas of Russia subjected to radioactive contamination due to nuclear tests at the Semipalatinsk test site, radioactive releases from the Mayak enterprise, and the Chernobyl accident.*

## 1. FORMULATION OF THE MODEL OF EXTERNAL EXPOSURE

External exposure is one of the most significant possible ways of population exposure due to broad-scale releases of radioactive substances into the environment. In any situation of human external exposure, the following three blocks of data are necessary for assessment of the effective dose:

- parameters of external gamma radiation field;
- parameters of human behaviour in this field;
- conversion factors from parameters of the gamma radiation field to the effective dose in a person.

The basic model for human exposure in case of radioactive contamination of the environment is the model for exposure above an open plot of virgin soil, and the absorbed dose in the air at the height of 1 m above the surface is used as the parameter for the radiation field. In this case, its value is influenced, besides the surface activity of deposited radionuclides, only by such natural factors as the initial deepening of radionuclides in soil, their radioactive decay, vertical migration of long-lived radionuclides, and the presence of snow cover.

The parameters of radiation field are different in case of population exposure in the anthropogenic environment. In the model, this fact is taken into account by means of location factors  $f_j$  defined as the ratio of the dose rate in the air in point  $j$  inside a settlement and in its vicinity, attributed to gamma radiation of radioactive depositions, to the similar value above a plot of virgin soil.

Human behaviour in the radiation field is described by means of occupancy factors  $p_{ij}$  which are the part of the time spent by representatives of the  $i$ -th population group in the  $j$ -th point of the settlement.

The third block of data necessary for assessment of the effective external dose is represented by the conversion factors that relate the actually measured values (the absorbed dose in the air) with the criterion of radiation impact – the effective dose—being assessed.

On this basis, we write the model equation for assessment of the effective external dose rate  $E_i$  for representatives of the  $i$ -th population group in the following way:

$$E_i(t) = d(t) \cdot k_E \cdot k_S \cdot \sum_j f_j \cdot p_{ij} ,$$

$$d(t) = r(t) \cdot \sum_k A^k \cdot g_0^k \cdot \exp(-\lambda^k \cdot t) ,$$

where:  $d(t)$  is the absorbed dose rate in the air at the height of 1 m above the open plot of virgin soil;

$k_E$  is the transfer factor from the absorbed dose in the air to the effective dose;

$k_S$  is the factor of influence of snow cover on the value of the effective dose;

$f_j$  is the location factor;

$p_{ij}$  is the occupancy factor;

$A^k$  is the surface activity of the  $k$ -th radionuclide as of the date of termination of radioactive deposition;

$g_0^k$  is the specific rate of the absorbed dose in the air from gamma radiation of the  $k$ -th radionuclide in the geometry of a plane isotropic source located at the air-soil interface;

$r(t)$  is the function that describes the influence of radionuclides migration in soil on the absorbed dose rate in the air, equal to the ratio of the dose rate at the time moment  $t$  above soil with the observed distribution of radionuclides in soil to the dose rate from a thin source with the same surface activity located at the air-soil interface;

$\lambda^k$  is the constant of radioactive decay for the  $k$ -th radionuclide;

$t$  is the time elapsed since the moment of termination of the radioactive depositions.

Numeric values of the parameters listed above were determined on the basis of long-term dosimetric investigations in the most contaminated regions of Russia, by performing phantom experiments, and on the basis of population polls [1, 2]. Verification of model assessments on the basis of measurement results for individual doses in inhabitants within the method of thermoluminescent dosimetry has shown that dose assessments obtained according to the model calculations did not differ from experimental results with the error up to factor 1.5 times at a confidence level of 95% [3]. The model was taken as the basis for official documents on assessment of current [4] and reconstruction of accumulated external doses to population living in the areas contaminated as a result of the Chernobyl accident [5]. Below we present the results illustrating the specific features of external dose formation in population after different radiation accidents that took place on the territory of the former USSR.

## 2. SPECIFIC FEATURES OF EXTERNAL DOSES FORMATION IN POPULATION DURING DIFFERENT RADIATION ACCIDENTS

Fig. 1 presents the calculation results that reflect dynamics of accumulation of effective external dose on the trace of radioactive depositions after a surface nuclear explosion and after the Chernobyl accident. In the first case, the initial data were activities of 41 radionuclides in the non-fractionated mixture of products of instant  $^{239}\text{Pu}$  fission by neutrons of fission spectrum, normalised to the  $^{137}\text{Cs}$  activity release as of the moment "x + 3 hours". The effective dose rate was calculated for  $t > 3$  hours from the explosion moment, which corresponds to the minimum time period of approach of the radioactive cloud to the boundaries of the Altay Kray. The value of the effective dose was obtained by integrating the dependence of the effective dose rate within the given time limits. In the second case, the initial data were activities of 15 radionuclides in the proportion determined for the far zone (over 100 km from the Chernobyl NPP) of the north-east Chernobyl trace [6]. Calculation of the effective dose was performed beginning from 48 hours after the moment of the accident, i. e., the time of beginning of main radioactive depositions on the territory of the Bryansk region. The dynamics of accumulation of the effective external dose in settlements of the Altay Kray located approximately at the distance of 150 km from the test site is such that about 60% of the dose during 50 years was formed during the first week after the radioactive depositions (without consideration for future changes in configuration of the source), and during the first year the dose was formed almost completely. In contrast to this case, in the settlements of the Bryansk region also located at the distance of 150 km from the Chernobyl accident, only 25% of the effective dose value during 50 years was formed during the first year after the accident.

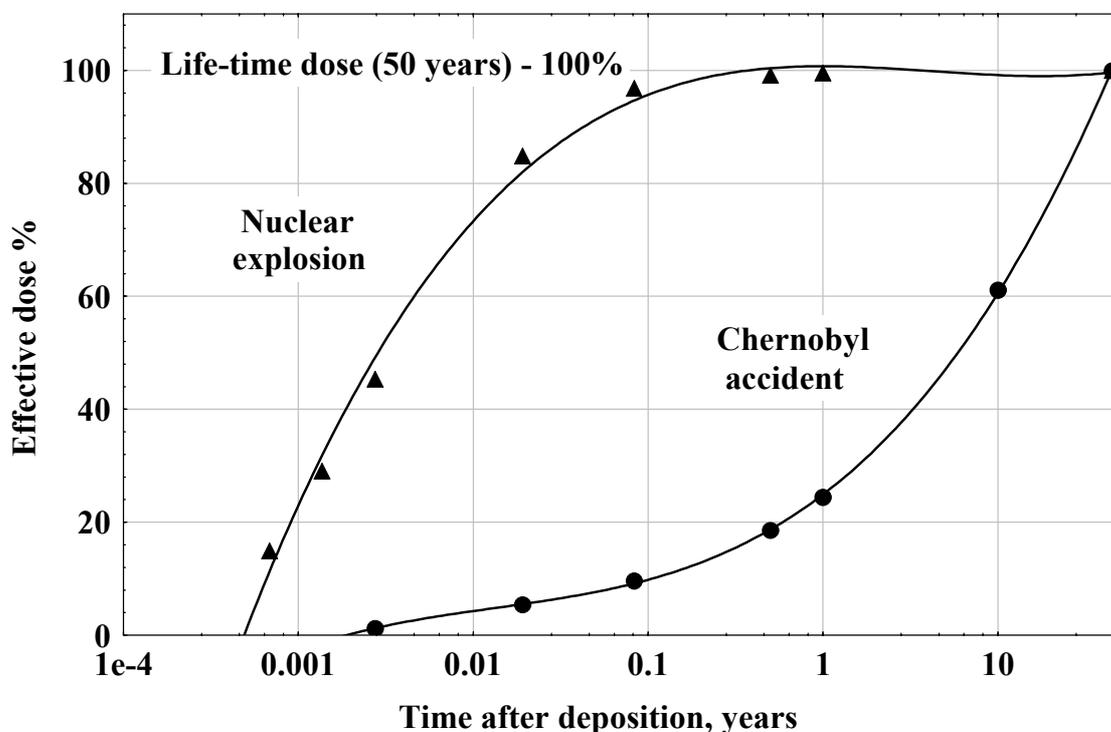


FIG. 1. The dynamics of accumulation of the effective external dose after nuclear explosion and after the Chernobyl accident.

The long-term dynamics of the gamma radiation dose rate is primarily connected with migration of the long-lived  $^{137}\text{Cs}$  radionuclide in the environment. During 1987-1999, in the most contaminated south-west part of the Bryansk region, over 300 samples of virgin soil were taken to the depth of 20 cm. Each core was separated in layers 2-5 cm thick with subsequent determination of  $^{137}\text{Cs}$  content within the gamma-spectrometry method. After that, the determined distribution of  $^{137}\text{Cs}$  activity in the upper soil layer was used to calculate the dose rate in the air at the height of 1 m above the soil surface [7]. The results of pre-Chernobyl investigations of caesium migration [8, 9] indicated that the function of influence of caesium deepening in soil on the decrease of absorbed dose rate in the air can be represented in the two-exponential form with a short ( $\sim$  some years) and a long ( $\sim$  some tens years) periods. The short time of investigations after the Chernobyl accident did not permit to assess correctly the value of the long period. Therefore, we included in the analysis the results of investigations of caesium migration during 24 and 30 years [10]. The joint analysis of the results permitted to assess the parameters of the  $r(t)$  function with lesser error, at least for the “age” of radioactive depositions up to 30 years (see Fig. 2).

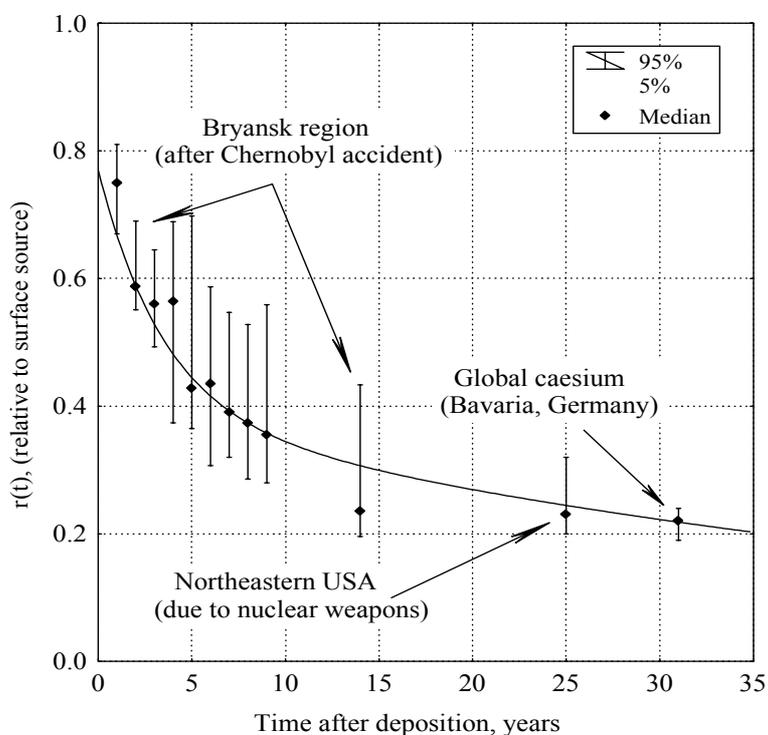


FIG. 2. Long-term dynamics of the absorbed dose rate in air from  $^{137}\text{Cs}$  gamma radiation over virgin land.

The analysis of over 5000 individual external doses measured within the method of thermoluminescent dosimetry in inhabitants of the Bryansk region has shown that the distribution of individual doses can be approximated by a lognormal law [11]. Moreover, if we plot the distribution of ratios of individual doses to the average value of the dose in the settlement, then the parameters of such distribution will be very close for all settlements, and the lognormal distribution with the geometric mean  $\sim 0.9$  and geometric standard deviation  $\sim 1.5$  can be assumed typical for the areas contaminated as a result of the Chernobyl accident. Fig. 3 presents the results for comparison of distributions of individual doses obtained on the basis of measurements within the thermoluminescent dosimetry (565 measurements) in three villages of the Bryansk region in summer 1993, and within stochastic simulation. It is directly

seen that both distributions actually have the same parameters, thus confirming the adequate character of the model structure and numeric values of its parameters.

The conditions of external exposure to population living in the drainage area of the Techa river significantly differ from those on other contaminated areas of Russia, because here the main source of radiation is a small part of the river water-meadow. The territory of settlements proper was subjected to considerably lesser radioactive contamination. Thus, for example, in village Muslyumovo, the average value of the  $^{137}\text{Cs}$  surface activity in soils of river water-meadow is approximately by 60 times higher than the similar value for soil plots in the settlement proper or in its vicinity outside the river water-meadow. In contrast to the “Chernobyl” variant of the model, in this case the parameters of the gamma radiation field that we used were the results of direct measurements of dose rates in the air at the height of 1 m above the surface both in the Techa river water-meadow and in settlements proper obtained during investigations of radiation situation in the region in the middle of the nineties [12].

To find out the character and duration of contacts of the population with the river water-meadow, we performed on site observations by the river in the village Muslyumovo [13]. Processing the observations results permitted to assess both the average values and scatter for values of duration’s of staying in the river water-meadow for representatives of different population groups. As an example, Fig. 4 presents the results for assessments of individual external doses distribution in two population groups in the village Muslyumovo obtained within the method of stochastic simulation. Representatives of the first population group correspond to adult inhabitants of the village with the average value of duration of staying in the river water-meadow (about 1.5 hours per day in summer). The duration of staying of another, critical population group (herdsmen) in the river water-meadow in summer can reach 10 hours per day. Both distributions of individual doses, as in the Chernobyl case, correspond to the lognormal law. However, these distributions have greater positive asymmetry, which is quantitatively confirmed by the greater value of the geometric standard deviation ( $\sim 2$ ). The presented results indicate that in a part of representatives of the critical population group the external dose even taken separately can exceed the dose limit set for population, 1 mSv.

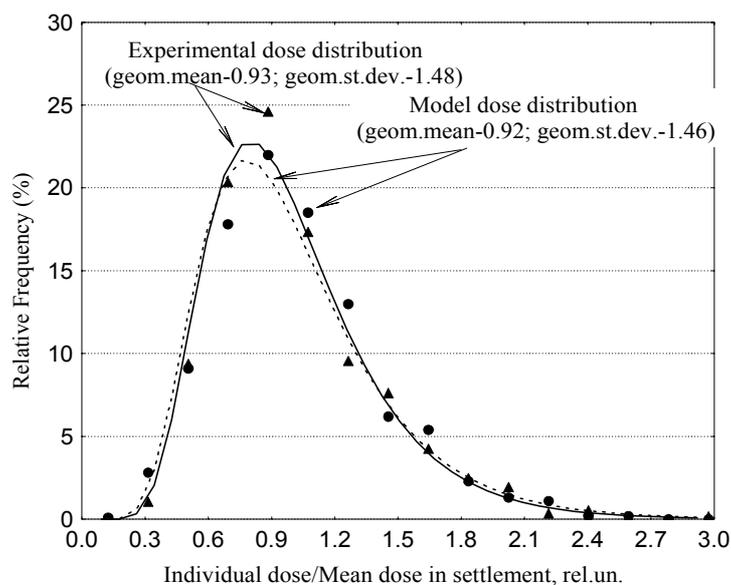


FIG. 3. Frequency distributions of monthly effective doses measured in summer 1993 with TL-dosimeters in three villages of the Bryansk region and calculated by stochastic model.

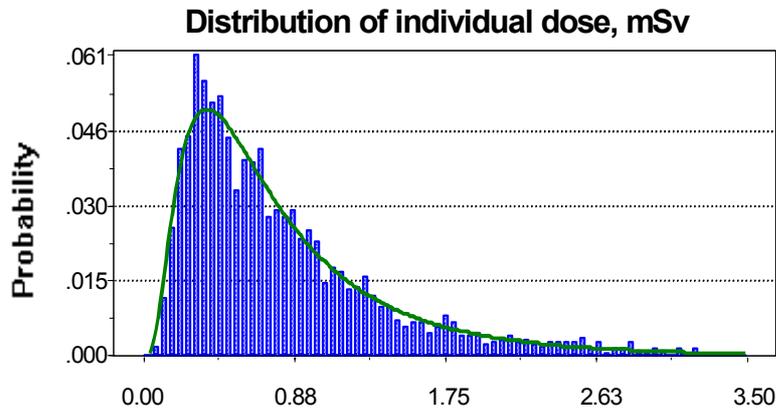
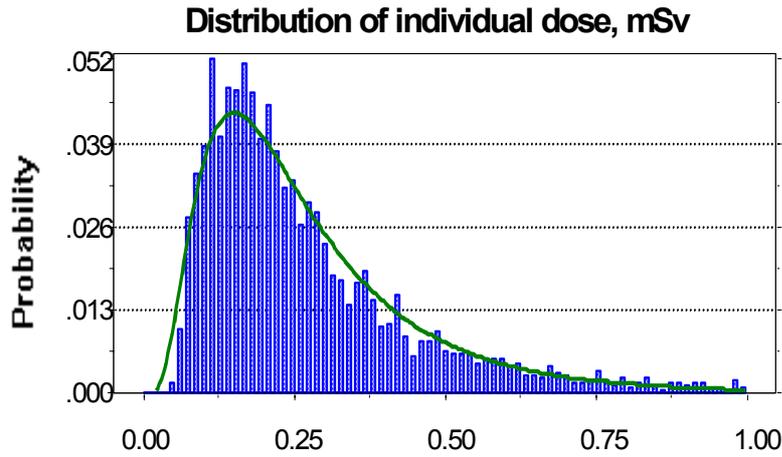


FIG. 4. Individual external doses distribution in two population groups in the village Muslyumovo. The upper figure correspond to adult inhabitants of the village with the average value of duration of staying in the river water-meadow, the lower figure correspond to critical population group.

The methodology for external dose assessment described above was used as the official procedure for determination of current and accumulated doses in inhabitants who live in the areas subjected to radioactive contamination due to the Chernobyl accident. To conclude, we present in Fig. 5 the calculation results for the collective effective external and internal doses (excluding the thyroid dose) in inhabitants of 18 regions of Russia accumulated during 15 years after the accident. The total collective dose in 36 millions inhabitants in 20 regions of European part of Russia was 66200 pers. $\cdot$ Sv during 15 years after the Chernobyl accident. Of this value, the external collective dose was 23380 pers. $\cdot$ Sv, or 35%. The relation between the external and internal doses depends mainly on soil type in the region and on the efficiency of countermeasures, which were more efficient with respect to reduction of internal, but not external exposure to population.

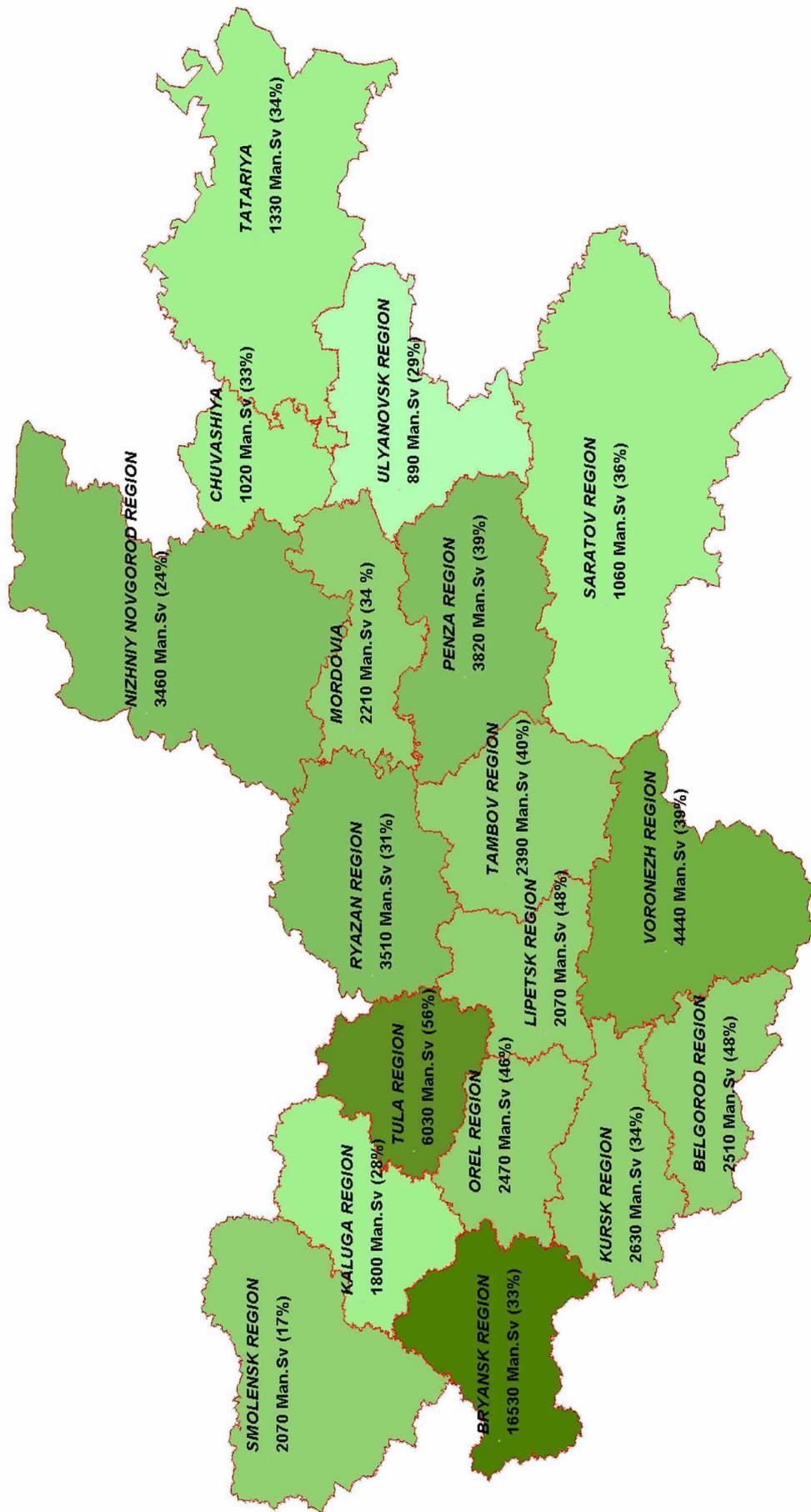


FIG 5. Cumulative effective dose during 15 years after the Chernobyl accident (man.Sv) in different regions of Russia. The contribution of external dose in the total dose is given in brackets in %.

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#### DISCUSSION AFTER THE PRESENTATION OF V.Y. GOLIKOV

**Yu.A. IZRAEL (Russian Federation-Session Chair):** When estimating the doses resulting from nuclear explosions, did you take account of fractionation?

**V.Y. GOLIKOV (Russian Federation):** No, we did not.

**Yu.A. IZRAEL (Russian Federation-Session Chair):** In that case, zirconium-95 will not show up in your calculations.

**E.D. STUKIN (Russian Federation):** According to calculations performed by us, one and a half years after the Chernobyl accident the contribution of  $^{134}\text{Cs}$  to the dose exceeded that of  $^{137}\text{Cs}$ . What do you think the reason for that was?

**V.Y. GOLIKOV (Russian Federation):** The specific dose rate from  $^{134}\text{Cs}$  is about 2.7 times that from  $^{137}\text{Cs}$ . With a fallout ratio of about 0.5, for one and a half years the  $^{134}\text{Cs}$  accounted for more of the gamma radiation dose than the  $^{137}\text{Cs}$ .

**E.V. KVASNIKOVA (Russian Federation):** There is usually a direct correlation between deposition and gamma dose rate. In some situations, however, as the deposition increases owing to erosion the maximum caesium concentration decreases and there is a corresponding decrease in the dose rate-and perhaps a fairly significant decrease also in the external gamma dose. To what extent did you take such processes into account in your calculations, and can they significantly affect the total external dose from region to region?

**V.Y. GOLIKOV (Russian Federation):** Yes, they can. The curve on the slide which I showed relating to the long term migration of  $^{137}\text{Cs}$  is a function of time. It reflects the dose rate decline due to migration. We can calculate the dose over any profile. Here is reflected the natural process of migration in virgin, open soil-for the Bryansk region. This decrease in dose is taken into account very correctly in that it is calculated over the real caesium profile in the soil.

# CHARACTERISTICS OF PARTICULATE RADIONUCLIDES IN THE ATMOSPHERIC SURFACE LAYER OF THE 30-km ZONE OF CHERNOBYL

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## **Abstract**

*For the inhalation dose assessment, reliable estimations are necessary of the average volume concentration of particle bound radionuclides in the atmosphere specifying the nuclide composition, the activity distribution in the different particle size ranges and the solubility characteristics of the nuclides. For that purpose, the analysis of measurement series is presented of the daily average activity concentrations of  $^{137}\text{Cs}$  and  $^{144}\text{Ce}$  and their temporal and spatial variability. From 1986 till 1994, samples were taken with high-volume samplers and multicascade impactors; the number concentrations of aerosol particles and the number concentrations of "hot" particles were determined for different conditions with an Aerosol Particle Sizer and a Rotating Arm Impactor. The data demonstrate a decrease the atmospheric concentrations of  $^{137}\text{Cs}$  and  $^{144}\text{Ce}$  with time larger than caused by radioactive decay alone. A statistical analysis showed a high level of fluctuations in the concentration of radionuclides in air with maxima exceeding the annual average by 10 to 20 times. The analysis of 88 experimental radioactivity size distributions at Zapolie and Pripjat for wind-driven resuspension conditions have shown that the measured distributions are generally very wide and differ from the log-normal distribution in the most cases. At Zapolie, the mean air concentrations of  $^{137}\text{Cs}$ , discriminated in four size ranges, showed an increasing part of inhalable particles with time since the accident. In 1993, the inhalable fraction was about 48% of the total concentration. The size distribution of atmospheric  $^{137}\text{Cs}$  particulate activity during these periods of enhanced resuspension showed a similar common shape with two maxima, the first in the 2-4  $\mu\text{m}$  range, and the second in the 12-20  $\mu\text{m}$  range. The estimated radioactive loading of particles showed an enrichment of resuspended radionuclides compared with soil particles. The highest enrichment factor was found for large particles, the lowest for fine particles. The investigations on the solubility of the radionuclides  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$  in selected Petryanov air filter samples according standardized tests give first data of the solution rates and the respective contribution of the fine radioactive particles and the coarse "hot" particles to the total activity of the samples.*

## 1. INTRODUCTION

After the Chernobyl accident, the 30-km zone is a permanent source of radioactive aerosol, including hot particles, due to the high level of soil contamination. Knowledge of the characteristics of the radioactive aerosol will be required for a long time for a more detailed dose assessment and estimation of processes of redistribution of the contamination of the surface soil. The results of the measurements of the resuspended radioactive aerosol of the Chernobyl area were obtained in the three scientific programmes. One was launched by the Hydrometeorological Committee of the USSR, the second ("Contamination of surface by resuspended material", ECP1) by the European Parliament, the third by the collaboration between the two reporting institutes.

In these studies, aerosol measurements are analysed in the surface layer of the atmosphere generally within the 30-km zone. They were conducted from July 1986 to September 1994 with high-volume samplers in the towns of Chernobyl and Pripjat and with the multicascade

impactors in the town Pripjat and the village Zapolie. In 1993, agricultural activities and operations of different trucks were simulated. In these experiments the size distribution of radioactive particles was measured by a cascade impactor, a Rotating Arm Impactor (RAI), the mass concentration with a Berner impactor, the number concentration of airborne particles in the size 0.6-30  $\mu\text{m}$  was measured in a size-resolved manner by an Aerodynamic Particle Sizer (APS). More details of the measurements sites, instruments and experiments are given in [1,2].

## 2. RESULTS AND DISCUSSION

Material, which has been deposited after an accident may be, resuspended by wind only, or by wind and disturbances at the ground surface by anthropogenic activities, for example, agricultural activity or movement of vehicles. The data presented in [3] demonstrate a decrease of the atmospheric concentrations of  $^{137}\text{Cs}$  and  $^{144}\text{Ce}$  with time greater than due to radioactive decay alone. There must be further processes which are responsible for the reduction of the activity concentration in the air, mechanisms for instance which reduce the source term of resuspension such as vertical migration in soil, run-off with rain or melting water and snow cover. If the atmospheric concentrations are presented as a rolling mean, any seasonality and long-term features in the data can be seen more clearly. The atmospheric concentrations of  $^{137}\text{Cs}$  shortly after the accident, averaged over a seven-month cycle, measured at Chernobyl and Baryshevka (150 km south-east of Chernobyl), are shown in Fig. 1. The figure shows that the atmospheric concentrations decreased in two phases. First, there was a very rapid fall over a period of a few months. Secondly a much slower decline in the atmospheric concentrations followed. Overlying these features, there is evidence of seasonal changes in the atmospheric concentrations and other possible resuspension effects.

A statistical analysis [3] of the series of measurements in the years 1987-1991 of the daily average concentration of  $^{137}\text{Cs}$  and  $^{144}\text{Ce}$  activity in air close to the ground showed a high level of fluctuations in the concentration of radionuclides in the air with maxima exceeding the annual average by 10 to 20 times. Periods when the concentration exceeded the mean lasted from 1 to 14 days. During all years, the intensity of fluctuation (determined as the ratio of the standard deviation to the average concentration) exceeded unity. The strong asymmetry of the distribution of the  $^{137}\text{Cs}$  and  $^{144}\text{Ce}$  concentration is reflected by the large ratio of the average concentration to the median and the values of the skewness. The frequency distributions of the activity concentrations, which are above the annual average, are not described satisfactorily by single analytical distributions over the whole period of measurements.

Apparently, several processes are responsible for the formation of the concentration field of radioactive aerosol: local resuspension by the wind, transport from a distant area and anthropogenic resuspension. Accordingly, the observed distributions would result from the combination of several simple distributions evolving in time. The influence of the averaging period  $T$  on the estimation of the standard deviation  $\sigma(T_2) = \sigma(T_1) [T_2/T_1]^{-m}$  was determined. The mean value of the empirical exponent  $m$  is  $0.33 \pm 0.08$  [3]. The above relation can be used to compare different averaging periods  $T_1$  and  $T_2$ . It is permitted to estimate the affect of increasing the internal averaging time by sampling, for example, 3(10) days instead of 1 day: the standard deviation will decrease to 69% (47%) of the daily value. Therefore, the averaging period is of importance in the assessment of activity concentrations.

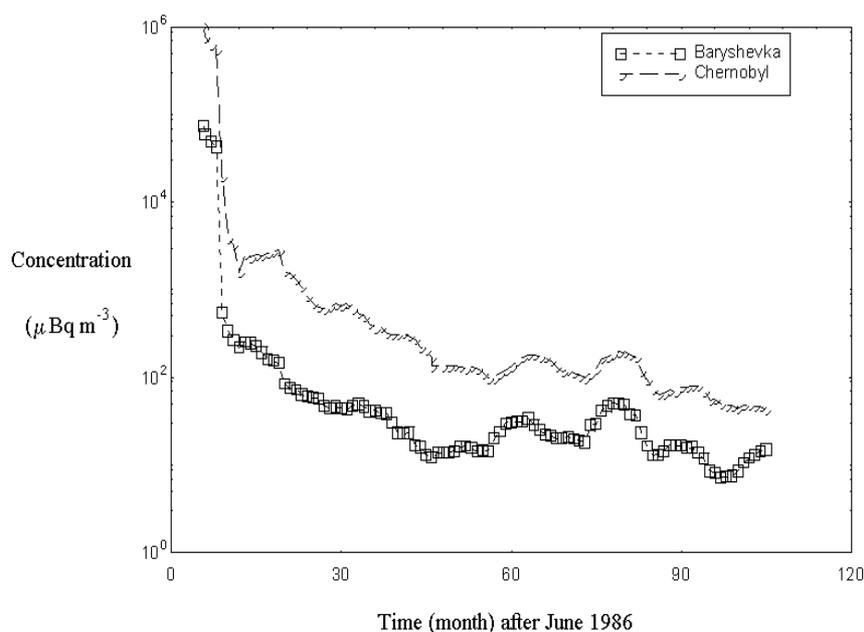


FIG.1. Rolling seven month mean atmospheric concentrations of Cs-137 at Baryshevka and Chernobyl (June 1986 to August 1994).

Detailed analysis of the radioactivity size distributions has shown that five typical shapes of distributions can be distinguished (see Fig. 2). The analysis of 88 samples has shown [4] that at Zapolie a bimodal distribution of  $^{137}\text{Cs}$  was observed in 91% of all cases which was formed by two processes: the local resuspension and the advective transport of radioactive aerosol from highly contaminated territories, e.g. from the Chernobyl NPP. However, at Pripyat, which is situated within a highly contaminated area, the shapes of size distributions were representative for local resuspension with only a weak transformation. The observed variability of the radionuclide size distributions in the air of the 30 km zone makes it difficult to calculate resuspension parameters taking into consideration only local processes (e.g., the estimation of the resuspension factor or the estimation of the airborne concentration using the radioactive loading of soil particles). During the measurement period the log-normal radionuclide size distribution was observed in only 3% of all cases in Zapolie. At Zapolie, the mean air concentrations of  $^{137}\text{Cs}$  discriminated in four size ranges showed an increasing part of inhalable particles with time since the accident. In 1993 the inhalable fraction was about 48% of the total concentration.

During forest fire, the activity size distribution was measured at a distance of 17 km from the burning area. The main part of radioactivity was connected to sub micrometer particles with the median diameters in the range  $0.28\ \mu\text{m}$ - $0.50\ \mu\text{m}$ . Anthropogenic enhanced resuspension was measured during different simulated agricultural activities and operations of different trucks at the site Zapolie [1,5]. For that purpose two different soil surfaces free of vegetation were prepared on which several tractor types were driven simulating soil management such as harrowing. The soil surface strips represented fixed line sources. In a typical experiment, the tractor started at one end of the strip, passed the sampler at a certain fixed distance, drove to the other end of the prepared surface and returned. For a certain experiment, the strip was chosen for which the wind trajectories pass the sampling equipment after crossing the prepared surface.

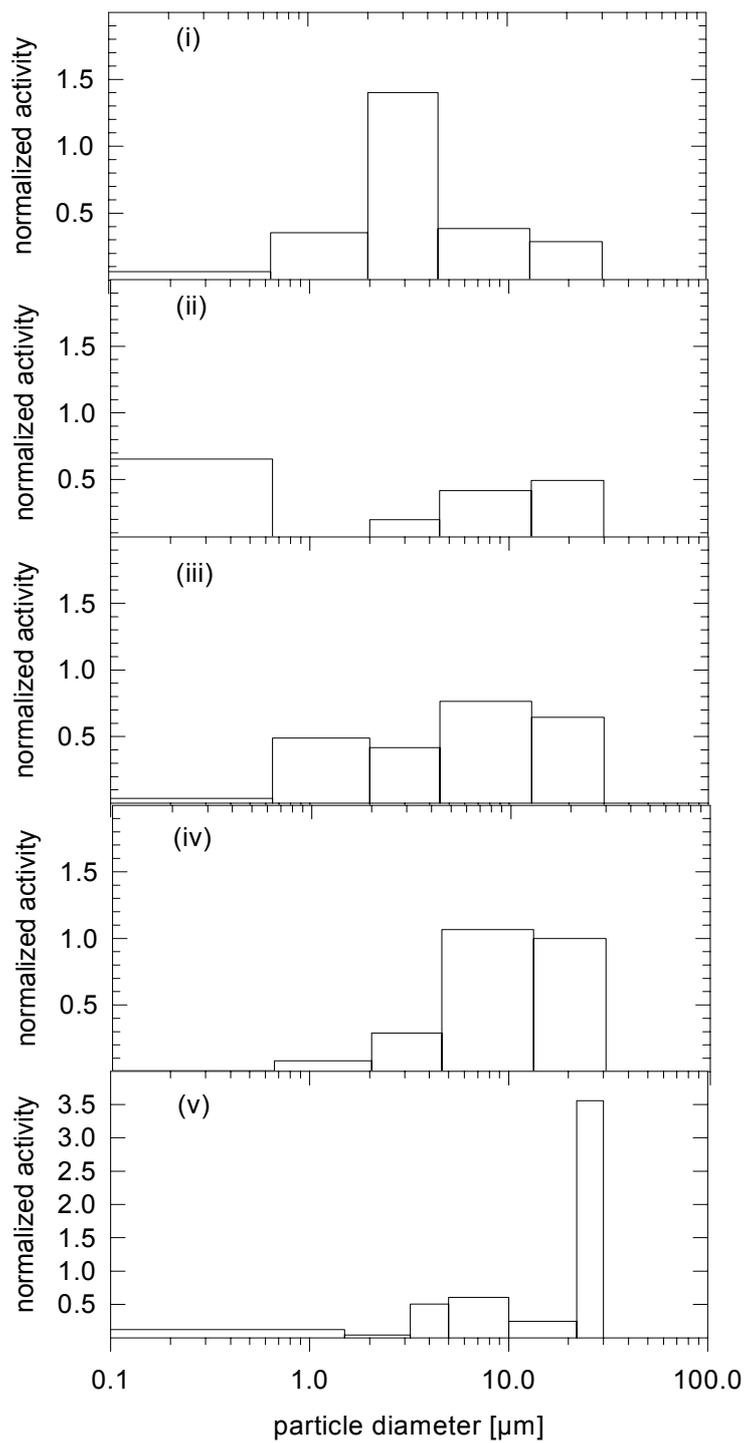


FIG.2. The five most frequent shapes of the activity size distribution in the Chernobyl area.

Results of the project measurements show that the total airborne radionuclide concentrations and deposition rates were increased considerably during anthropogenic enhanced resuspension. Depending on the experimental conditions, the increase was by a factor of several thousand in comparison to the concentrations occurring during wind resuspension at distances about 20-30 m from the dust sources and a factor 10-100 at distances of about 100 m or more [1]. The measurements of the number concentrations of hot particles show an increase of 3 orders of magnitude, reaching 0.7-1.0 hot particles / m<sup>3</sup> with a maximum activity of 1.5-2.0 Bq / particle. The size distributions of atmospheric <sup>137</sup>Cs particulate activity during these kinds of enhanced resuspension showed a similar common shape with two maxima, the first in the 2-4 μm range, and the second in the 12-20 μm range. In the fine particle size range (0.1-2.0 μm) 33% ± 6% of <sup>137</sup>Cs activity was found in the mean of all experiments. Measurements of the coarse particles (particle diameter d > 10 μm) have shown that the activity concentration of <sup>137</sup>Cs was not uniform distributed: in the size range 10-20 μm 44%, in the range 20-28 μm 10% and in the range > 28 μm 46% of the activity is found. Microscopic analysis of the samples (d > 20 μm) proved a linear relation between particle number and activity [6].

An estimation of the airborne plutonium concentrations in two experiments of anthropogenic enhanced resuspension showed a significant part of activity in the fine particle size range (36-40%) and in the large particle range (approximately 20%). The measurement of the number concentrations of particles has shown that the means for large particles (3-10 μm) and giant particles (10-30 μm) are, respectively, 4 times and 2 times larger than the mean number concentration of fine particles (0.6-1.0 μm) during periods of soil management (see Fig. 3). The increase of the mean number concentration in periods of soil management has been a factor of about 40 for large and giant particles, but only a factor of approximately 3 for the fine particles. The variability of the concentration is higher for giant particles. During anthropogenic enhanced resuspension predominantly large particles are injected in the atmospheric surface layer. The estimated radioactive loading of particles showed an enrichment of resuspended radionuclides compared to soil particles. The highest enrichment factor was found for large particles, the lowest for fine particles [5].

In the scientific investigations of solubility processes, there are only few works devoted to the study of dissolution of <sup>239+240</sup>Pu, <sup>90</sup>Sr, <sup>137</sup>Cs from Chernobyl origin in the atmospheric aerosol. There are investigations of solubility of hot particles from soil and bottom sediments near the Chernobyl NPP and from the Chernobyl nuclear reactor [7, 8]. But these data cannot be directly used for the inhalation dose assessment.

Generally, the absolute activities in the filter samples are small even in the 30 km zone. Only our high-volume samplers "Typhoon" (4200 m<sup>3</sup>h<sup>-1</sup>) gave us the possibility to estimate *in vitro* the solubility of the radionuclides <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>238+240</sup>Pu in the Petryanov filters exposed in 1987 and 1988 at Pripjat. Before dividing the filters in equivalent segments for the solubility study, the filters were tested with an autoradiography method. The nuclide concentration in the segments was measured by gamma-spectrometry. The segments of the Petryanov filters were confined within two 0.14 μm pore size membrane filters MFA (Dubna, Russia) as a filter sandwich [9] and exposed a dissolution fluid. The composition of the fluid was according to the Gamble biological fluid simulated [9].

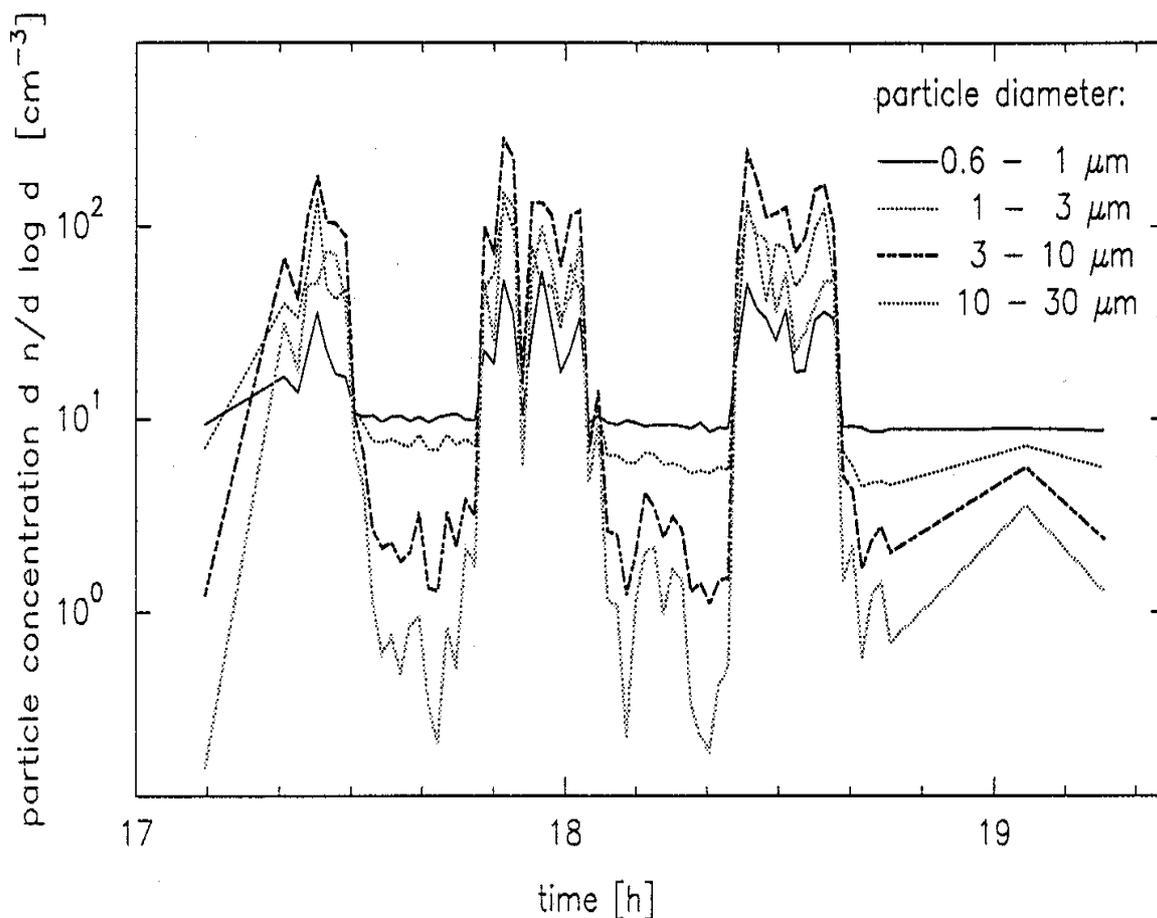


FIG. 3. Time record of the normalized particle number concentration in four different size ranges as measured by an Aerodynamic Particle Sizer during the resuspension experiment on 13 May 1993 at Zapolie. The periods of operation of the big tractor can be identified by the periods of increased particle concentrations. The increase of large particles is much higher than the increase of fine particles.

Dissolution data of  $^{137}\text{Cs}$  from five fragments of the Petryanov filter 12.10-13.10. 1987 are given in Table 1. From the 6-th fragment, an area of  $(1 \times 2) \text{ cm}^2$  was selected, where the  $^{137}\text{Cs}$  activity was  $(17.4 \pm 1.8) \text{ Bq}$  after 21 days of exposition into the Gamble solution. The rest part of that fragment had an activity of  $(5.36 \pm 0.54) \text{ Bq}$  after the same time period. The 7 and 8 fragments had activities below detection limit. In Table 1 is shown the mean and the standard deviation of the filter, the solution and the total activities. These data demonstrate the sharp heterogeneous of the activities at these filter areas (the ratio between max. and min. activity was 10.5 times). Unlike from filters, the solutions are more uniform. In the last column of Table 1 the relative  $^{137}\text{Cs}$  activity at the filter is shown. The dependence between the total values of activities and the solution fractions are obvious. The explanation may be the inverse connection of the activity with the particle size [10].

In Table 2 the dissolution data of  $^{139+140}\text{Pu}$  for the same fragments is presented. In spite of the higher sensitivity of the alpha detection than beta detection by LB 770–PC 10 Channel  $\alpha$ – $\beta$  Counter, the uncertainty for these solution measurements is much larger.

TABLE 1. DISSOLUTION OF  $^{137}\text{Cs}$  FROM FIVE FRAGMENTS OF THE PETRYANOV FILTER 12.10.-13.10.1987

Number of fragment and time for dissolution	Filter Activity Time of Measurement (Bq/sample) (s)		Solution Activity Time of Measurement (Bq/sample) (s)		Total Relative* Activity (Bq/sample) (s)	
	1,28 days	$2.53 \pm 0.29$	9029	$0.73 \pm 0.07$	79047	$3.26 \pm 0.26$
2,28 days	$2.64 \pm 0.22$	15442	$0.98 \pm 0.07$	85126	$3.62 \pm 0.19$	72.9
3,28 days	$5.23 \pm 0.43$	6648	$0.82 \pm 0.14$	28403	$6.05 \pm 0.40$	86.4
4,25 days	$7.16 \pm 0.07$	53084	$0.80 \pm 0.05$	146457	$7.96 \pm 0.07$	90.0
5,7 days	$26.5 \pm 0.32$	303444	$1.05 \pm 0.17$	12087	$27.55 \pm 0.32$	99.4

\* Ratio of the filter activity / the total activity in%.

TABLE 2. DISSOLUTION OF  $^{239+240}\text{Pu}$  FROM FIVE FRAGMENTS OF THE PETRYANOV FILTER 12.10.-13.10.1987

Number of fragment and time for dissolution	Filter Activity Time of measurement (Bq/sample) (s)		Solution Activity Time of measurement (mBq/sample) (s)		Total Relative* Activity Activity (Bq/sample) (%)	
	1, 28 days	$0.064 \pm 0.004$	48160	0.66	96660	0.06466
2, 28 days	$0.034 \pm 0.002$	73199	4.65	96660	0.03465	98.6
3 28 days	$0.178 \pm 0.098$	73034	0.28	96659	0.17828	99.92
4, 25 days	$0.200 \pm 0.011$	20922	0.13	351896	0.20013	99.94
5, 7 days	$0.610 \pm 0.031$	20922	0.79	135218	0.61079	99.87

\* Ratio of the filter activity / the total activity in%.

The contents of  $^{239+240}\text{Pu}$  activity in the air filters were two orders of magnitude less than the contents of  $^{137}\text{Cs}$  or  $^{90}\text{Sr}$  activity. The available Petryanov air filters contain a wide radioactivity distribution of aerosol particle sizes. Available experimental methods allow estimating a contribution to the fine radioactive particles and coarse hot particles too. The investigations on the solubility of the radionuclides  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{239+240}\text{Pu}$  in selected Petryanov air filter samples according standardized tests give first data of the solution rates and the respective contribution of the fine radioactive particles and the coarse “hot” particles to the total activity of the samples.

## CONCLUSION

The determined variability of the characteristic properties of the resuspended radioactive material as radionuclide concentration, particle size distribution and solubility in simulated lung fluid makes it necessary to determine these properties over a long period. Only with sufficient experimental data of the most significant characteristics, a reliable inhalation dose assessment will be possible.

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# A STATE-OF-THE-ART METHODOLOGY FOR IMPACT ASSESSMENT OF COVERED URANIUM MILL TAILINGS

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## *Abstract*

*An impact assessment methodology is being developed that integrates several advanced modelling and characterisation techniques for the purpose of assessing the current and future environmental and health impact of a surface repository containing wastes from uranium milling and radium processing. The former radium processing plant at Olen, Belgium, accumulated during nearly half a century considerable amounts of radium-containing wastes. Also present at the site are uranium mill tailings. These wastes were disposed of in a heavily engineered surface repository at the occasion of a remediation plan carried out in the mid eighties. The repository contains several concrete bunkers covered with a multi-layer hydraulic barrier. In the current impact assessment study the only exposure pathway discussed is by contamination of groundwater. For this purpose we calculated variably-saturated water flow in the multi-layer barrier and the underlying waste zones and used geochemical modelling to estimate the chemical species and their solubility's in the aqueous phase of the various waste forms. The assessment further includes modelling of contaminant leaching from the tailings towards the groundwater, contaminant transport in the surrounding groundwater towards a water well, and evaluation of the doses for ingestion, inhalation and external irradiation resulting from use of groundwater from the well. Details of the waste and site characterisation as well as contaminant modelling are discussed.*

## 1. INTRODUCTION

The company Union Minière (UM) at Olen (Province of Antwerp, Belgium) has a long history in the production of radium. The radium production, for instance, started in 1922 and ended in 1969. Half of the total world production<sup>1</sup> of radium has been produced by UM at Olen [1]. During more than half a century of industrial activities, a variety of radioactive wastes have been generated and are presently disposed of in a heavily engineered surface repository at Olen. The wastes include radium rich uranium mill tailings, residues from the radium production plants, radium needles, contaminated building materials and soils, etc.

The present study deals with the long-term impact assessment of the surface repository at Olen. It considers an important pathway by which radionuclides can reach man and the environment, i.e., leaching from the repository towards the groundwater and consecutive transport through groundwater into wells and rivers. For the purpose of assessing the impact of radiation on man, radionuclide concentrations and doses will be calculated using state-of-the-art computer models together with appropriate parameters representative for the different physical and chemical processes that play a role in the behaviour of the radionuclides.

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<sup>1</sup> The total radium production until 1989 is estimated at 4.5 kg.

## 2. METHODOLOGY

In the impact assessment calculations leaching towards the groundwater is considered to be a major pathway through which contaminants present in the disposal structure can reach man and environment. A schematic overview of such pathway is seen in Figure 1. Three steps can be distinguished along the travel path of the contaminants:

Dissolution of the waste form into the pore water of the silo or bunker, followed by predominantly vertical transport towards the unsaturated soil as a result of molecular diffusion and advection, where the latter is due to rain water infiltrating the multi-layer cover (step 1 in Figure 1). The contaminant transport is strongly retarded by adsorption of chemicals onto mineral phases of soil and concrete.

In the unsaturated soil, vertical transport is governed by advection, molecular diffusion, and adsorption onto mineral phases of the soil (step 2 in Figure 1).

Once the contaminant plume reaches the groundwater, predominantly horizontal transport takes place with advection, hydrodynamic dispersion, and adsorption being the most important processes (step 3 in Figure 1).

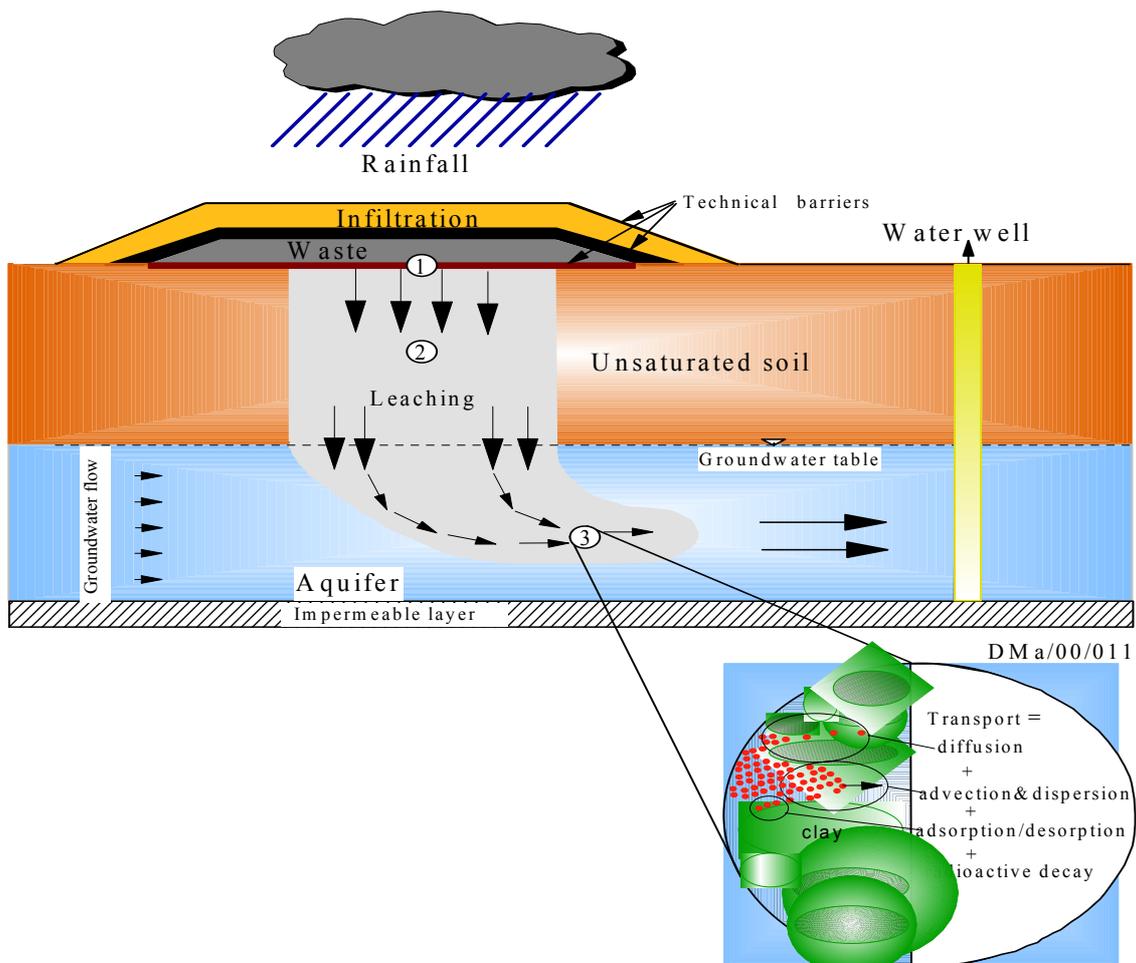


FIG. 1. Schematic of processes considered in impact assessment calculations in case of contaminant migration from surface repository to groundwater.

The degree of pollution of the groundwater by leaching of contaminants from the surface disposal site was assessed in a two-step approach. First, a highly conservative screening calculation was performed to assess the hazard of all elements present in the different wastes. Elements were screened on the basis of four criteria to decide whether or not they further required detailed assessment. The first two criteria are quantitative whereas the next two are more qualitative. These criteria were: (i) comparison between elemental concentration in the waste zone and drinking water standards, (ii) comparison between estimated concentrations in groundwater and drinking water standards, (iii) abundance in the geosphere, and (iv) miscellaneous criteria. These four criteria were applied in this order, i.e. from (i) to (iv), because more weight was given to the quantitative criteria. In a second, more detailed calculation, the groundwater concentrations of the remaining elements (i.e., those that could have a potential health effect) were assessed using numerical flow and transport models. For most waste types the majority of elements did not require such a detailed analysis.

Elemental concentration in the liquid phase of the waste is a useful criterion in the screening process since it is the maximum concentration in the entire pathway of a given element. When no information about the pore water composition was available, geochemical calculations were carried out as a way to estimate the elemental concentrations (e.g., for the radium-rich uranium mill tailings, see further). As will be discussed further, several elements were solubility limited under the assumed geochemical conditions of pH and Eh. The solubility of a given element is mainly determined by its stable mineral phase. Since the geochemical calculations revealed many possible mineral phases, we always selected that mineral with the highest solubility. This conservative approach was necessary, as no detailed information on the mineralogical composition was available.

In the conservative screening calculation it was further assumed that the entire inventory of all elements would be released to the groundwater in a very short period of time. In other words, the retarding effects of the engineered barriers on the release are neglected. In this first screening criteria the limited solubility of the elements was also excluded from the calculations. Another assumption was that an equilibrium partitioning of the chemicals in the groundwater and the solid materials would exist and that this partitioning could be well described by means of the linear Freundlich sorption model. Concentrations in groundwater are then compared with drinking water standards, and are expressed as the ratio of groundwater concentration to drinking water standard. Elements with ratios smaller than one are not considered further in the detailed calculations because of their negligible health effect.

Several elements are ubiquitous in man's natural environment and can be excluded from further assessment. This is true for calcium, iron, magnesium, phosphorus, and sulphur. These elements are not considered as contaminants and are excluded from the list for which detailed calculations are required. The same reasoning can be used for aluminium and silicium, because they are the principal constituents of soils and rocks. Certainly their presence in a surface disposal should not result in a significant increase in existing levels of toxicity. Furthermore, the main source for aluminium and silicium would be the waste matrix, i.e. the sand fraction.

The screening analysis revealed that radium and uranium did require further analysis for nearly all waste types. An exception are the calcium silicate wastes, for which radium had a sufficiently low concentration. Other elements which needed further analysis include lead, cobalt, and manganese, among others. A discussion of the heavy metal behaviour is beyond the scope of the present paper.

### 3. DATA COLLECTION

#### 3.1 Hydrogeological characterization

The sequence, thickness, and physico-chemical properties of geological layers is important information in an impact study, for they determine the extent the radionuclides will spread in the groundwater as soon as they migrate below the disposal installation.

The description of the geological layers together with vertical variation in physico-chemical properties was based on a set of drillings. The general stratigraphy can be deduced from Figure 2. A fine sandy top layer (Kasterlee sand) extends from the surface to a maximum depth of approximately 3 m. Between 3 and 5 m we note the presence of a dark green clay layer with an irregular thickness (Kasterlee Clay). At some locations it is only present as a fairly thin clay lens. The following layer is again Kasterlee sand, but now contains a small amount of glauconite and has a typical light green colour. The lowest sand layer contains about 25-30% glauconite and is known as Diest sand, which has an approximate thickness of 90 m. The presence of glauconite is of particular importance because of its high sorption capacity for cations ([2], see further). Together with the Kasterlee clay it is an effective barrier which retards the further spreading of radionuclides migrating from the disposal site.

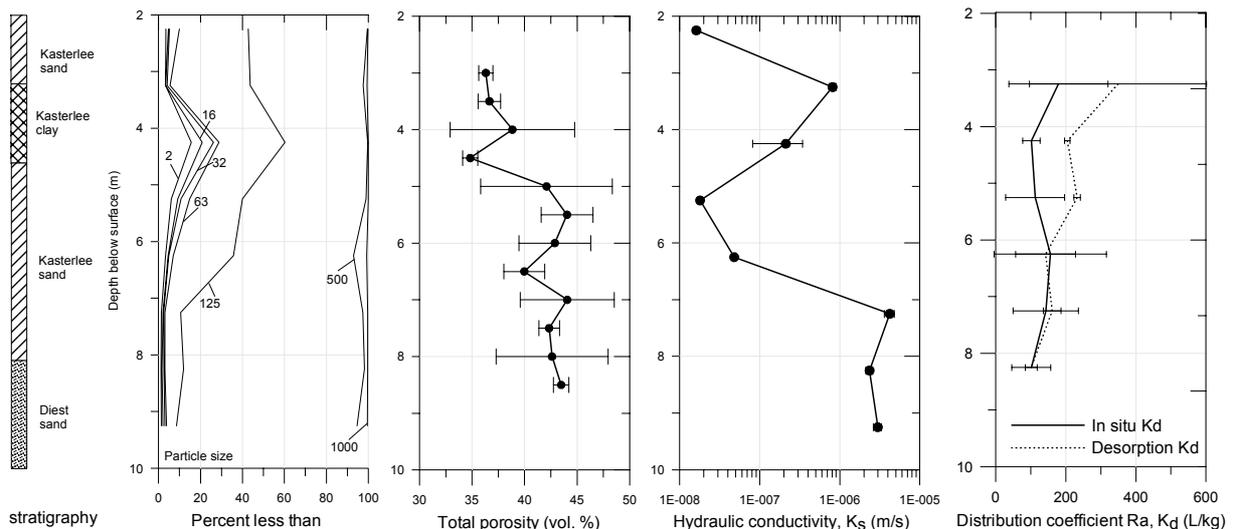


FIG. 2. Vertical variation in physical and chemical parameters at the Olen site. Error bars indicate one standard deviation around the mean.

A summary of some key parameters is shown in Figure 2. Inspection of the particle size distribution confirms the presence of Kasterlee clay layer between 3 and 5 m depth, with a maximum percentage of clay between 15-20%. Total porosity is around 37% in the top layer and 43% in the deeper layers. For transport calculations effective rather than total porosity is needed. For sandy materials total and effective porosity do not differ that much [3], and we assumed them to be identical. Higher fraction of fine particles (say  $< 125 \mu\text{m}$ ) in the first 5 m presumably explains the lower saturated hydraulic conductivity values at those depths (with  $K_s$  around  $10^{-7}$  m/s). In the deeper Diest sands  $K_s$  is approximately  $3 \times 10^{-6}$  m/s.

## Sorption of radium and uranium on aquifer sediments

The long-term performance of radioactive waste disposal facilities to a large extent depends on the degree of sorption onto the technical barriers and to a lesser degree on the sorption in the geosphere. Ideally, sorption will retard the migration of contaminants leading to a contaminant release that is spread over long times. Several sorption mechanisms have been identified in well-defined systems, including equilibrium reactions (Freundlich and Langmuir models are perhaps the most commonly used equilibrium models) and kinetic or time-dependent types of reactions (e.g., the first-order kinetic reversible reaction for describing time-dependent adsorption/desorption). The Freundlich model is the simplest approach to quantify the behaviour of retention of reactive solutes with the soil matrix. It has been used widely to describe solute retention by soils ([4]; among others). It has been used as an approximation of the dilute end of the adsorption isotherm where concentrations are low and linear sorption can be assumed.

In this study, solid-liquid distribution coefficient ( $K_d$ ) values of a large number of elements present in the waste are proposed for the screening calculations and the detailed performance assessment calculations. Most of the data were compiled from the literature for porous materials and conditions that are supposed to be representative of those expected in the technical barriers (e.g., a cementitious environment in the bunkers containing Ra-sources), soils, and the aquifers [5]. Additional measurements were done to determine  $K_d$  for the major radionuclides of concern, i.e. radium and uranium, for the aquifer sediments.

Both in-situ and laboratory batch methods were used to determine  $K_d$  for radium and uranium. The in-situ method consisted of measuring the in-situ pore water Ra and U concentration on solutions obtained after centrifuging sediment material collected at different depths in several boreholes. The total sorbed concentrations were obtained by digestion of soil material using standard techniques. Distribution coefficients were then calculated as the ratio of total sorbed to in-situ pore water concentrations. This resulted in an average  $K_d$  of 132 L/kg for Ra and 68 L/kg for U. In the laboratory batch method contaminated soil solutions were allowed to equilibrate on a shaker during one day. Radium concentrations were then measured in the solution obtained after centrifuging. The radium concentrations thus obtained by desorption from the solid phase are considered to be the exchangeable fraction. In a separate experiment the radium concentration in the solution was determined. The  $K_d$  was then determined as the ratio of exchangeable radium to soil solution radium. Average values were 197 L/kg for Ra and 204 L/kg for U.

Vertical variation in  $K_d$  for radium considering the two measurement techniques is shown in Figure 2. Note that up to 6 m depth the in situ  $K_d$  is smaller than the desorption  $K_d$ . Parameter values obtained from both measurement techniques together with literature values were used in a sensitivity analysis with the transport model but final results are always given for the most conservative values.

### 3.3 Waste characteristics

A variety of wastes originating from the production of radium and uranium have been disposed of in the heavily engineered repository. An overview of the different types of waste is provided in Table 1. Six major types of waste have been identified. Some wastes contain mainly radium (e.g., the radium sources), while others are a mixture of various chemical substances, such as radium, uranium, heavy metals, iron and aluminium oxides. Only for the

radium sources and one type of uranium mill tailings (referred to as tailings D) concrete was used to better confine the waste. The major solid phase for the other wastes depends on the waste type: tailings are mainly composed of sands and fines such as clays. Radium sources, uranium mill tailings and other Ra-rich tailings are disposed of in reinforced concrete bunkers. All other wastes are located in silos. A multi-layer cover is present on bunkers and silos to prevent infiltration of rainwater and to avoid dispersion by human intrusion and biotic forces.

TABLE 1. RADIUM CONTENT AND CONCENTRATION (ONLY RANGE IS INDICATED) FOR WASTE TYPES DISPOSED OF IN ENGINEERED DISPOSAL STRUCTURE (n.a. = not applicable)

General waste type	Detailed waste type	Ra content (10 <sup>9</sup> Bq)	Ra conc. (Bq/g)
Ra-sources	Ra-Be sources	730	n.a.
	Needles	5 600	n.a.
Uranium mill tailings	Ra-salts	870	n.a.
	Tailings A	3 200	5000-10000
	Tailings B	11 000	20000-30000
	Tailings C	3 000	10000-20000
	Tailings D	8 900	10000-20000
Other tailings: Ra-rich	HCl leach tailings	44	500-1000
	BaCl <sub>2</sub>	1 500	5000-10000
	BaCl <sub>2</sub>	11	1000-5000
	BaSO <sub>4</sub>	570	1000-5000
	Carbonatation tailings	7,4	5000-10000
Other tailings: Ra-low	Rare earth's	3,7	500-1000
	Green iron hydroxide	41	100-500
	Brown iron hydroxide	960	500-1000
	Lead carbonate	3,7	100-500
	Lead sulphide	150	100-500
Diverse wastes	Pb residues	140	500-1000
	CaSiO <sub>3</sub>	320	100-500
	Waste water treatment residues	350	500-1000
Contaminated soil & building material		890	<100

Understanding the composition of the tailings is of paramount importance to assess how the radium and other radionuclides are sorbed onto the different materials. Tailings chemistry and mineralogy will determine the strength of sorption onto the solids. In general, tailings contain the following minerals [6]:

- Quartz comprises more than 70% of the tailings sands and 20 to 60% of tailings fines;
- Tailings fines contain 10 to 30% of clays (mainly illite and kaolinite);
- Feldspars are a major constituent of fines;
- Other minerals are calcite ( $\text{CaCO}_3$ ), barite ( $\text{BaSO}_4$ ), chloroapatite ( $\text{Ca}_5(\text{PO}_4)_3\text{Cl}$ ), and -hematite;
- The sulphuric acid leached tailings also contain gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) and jarosite ( $\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$ ).

Chemical properties of uranium mill tailings relevant to migration (i.e.,  $K_d$ , solubility) were determined by geochemical calculations. On the basis of the elemental composition and the very low pH, geochemical calculations were carried out to determine the mineral phases, the chemical species present in the liquid phase and their concentration. This was done using the geochemical code The Geochemist's Workbench [7]. Sorption onto mineral phases is only considered for the neutralized waste (tailings D), with the main adsorption assumed to occur onto Hydrous Ferric Oxide (HFO). No sorption is expected under the extremely acidic conditions for other waste types (tailings A, B, and C).

Distribution coefficients  $K_d$  for Ra and U present in the radium-low wastes were estimated on the basis of leaching tests [8]. This was done by expressing  $K_d$  as the ratio of solid to liquid phase concentration. For radium,  $K_d$ s are between 2700 L/kg for contaminated soils and 710000 L/kg for iron hydroxides (see Table 2), whereas for U considerably lower  $K_d$  values were obtained. For other elements present in the different waste types we conservatively assumed that the  $K_d$  for sand could be used (using data from [5]).

## 4. CONCEPTUAL AND NUMERICAL MODEL

### 4.1 Multi-layer cover

The engineered barriers comprise a set of concrete bunkers for the high-activity waste, and a multi-layer cover which isolates the bunkers and the low-activity waste present in silos from man and environment. At the bottom of the disposal system a concrete liner reduces infiltration of contaminated water to the groundwater. A schematic view of the different components of the disposal system is shown in Figure 3.

The entire site, approximately 13,600 m<sup>2</sup> including bunkers and silos, was isolated from the environment by means of a multi-layer cover. This cover was constructed using the following materials (from top to bottom), as is also shown in Figure 3:

Top soil (0.4 m thickness);

- Coarse sand (0.3 m thickness);
- Gravel (0.4 m thickness, composed of three different types of gravel);
- Coarse sand (0.3 m thickness);
- Boom Clay (ranging between 1.1 m and 1.2 m thickness);
- Porphyric rubble stone to cover the slopes (this layer will not be considered in the modelling of water infiltrating the multi-layer cover, because it has no water holding capacity and does not constitute a barrier against infiltrating water. It is therefore not shown in Figure 3).

TABLE 2. DERIVATION OF DISTRIBUTION COEFFICIENT  $K_D$  (L/KG) FOR RADIUM, AND URANIUM USING LIQUID ( $C_{AQ}$ ) AND SOLID ( $C_{SOL}$ ) PHASE CONCENTRATIONS

Waste Type	Ra			U		
	$C_{aq}$ [mg/l]	$C_{sol}$ [kg/kg]	$K_d$ [L/kg]	$C_{aq}$ [mg/l]	$C_{sol}$ [kg/kg]	$K_d$ [L/kg]
Iron hydroxides	2,0E-08	1,4E-08	7,1E+05	8,5	3,9E-03	4,6E+02
Rare earth	5,0E-07	1,7E-08	3,3E+04	10,4	9,2E-03	8,8E+02
Water treatment	1,6E-07	1,8E-08	1,1E+05	<1 <sup>#</sup>	n.d.	n.a.
Lead sulphide	1,0E-07	5,8E-09	5,8E+04	<1 <sup>#</sup>	2,7E-03	2,7E+03
Calcium silicates	2,0E-07	4,5E-09	2,3E+04	<1 <sup>#</sup>	9,0E-04	9,0E+02
Contaminated soils	1,9E-07	5,0E-10	2,7E+03	<1 <sup>#</sup>	n.d.	n.a.

n.d : not determined; n.a. : not applicable; <sup>#</sup> : detection limit used

The spatial arrangement of the different soil types in the multi-layered barrier system is defined in Figure 3. Note that only one side of the barrier is shown, as both sides are identical. The slope of the layers situated above the waste is approximately 8°. Layers used for side construction have a slope of 35°. Hydraulic properties of unsaturated materials were in part calculated from particle size distribution data and in part taken from [8].

The water flow boundary conditions applied in the present modelling study are a specified flux top boundary and a unit gradient (or free drainage) bottom boundary condition (Figure 3). The former equals a long-term average net rainfall rate (total rainfall minus runoff and evapotranspiration) of 0.00074 m/d at the Olen site. Two alternative net rainfall rates are also considered, namely for very dry (0.000255 m/d) and very wet (0.00141 m/d) conditions.

The numerical model used for flow calculations is shown in Figure 3, with the unstructured finite element mesh of the 34-m-wide and 8.5-m-high flow domain. The numerical model is

composed of 7725 triangular elements in total, with the smallest elements in regions where highest fluxes are expected. HYDRUS-2D [9] was used to calculate water flow in the multi-layer cover (Figure 3). The average water flux at the bottom of the multi-layer cover was about 1% of the net rainfall rate, for all three rainfall rates considered.

## 4.2 Bunkers and silos

Calculation of contaminant migration from the bunkers was done by considering the following two-dimensional conceptual model. Each bunker is composed of several equally sized cells; therefore calculations were done for one cell for each waste type. The computational domain is composed of four materials: the sediment underneath the cell, the soil used to fill the space between bunkers at one side of the cell, the concrete floor and wall, and the waste zone itself (Figure 4). The thickness of the sediment considered in the calculations is one meter, whereas the soil extends up to one meter from the bunker wall in the horizontal direction. The concrete floor is 0.15 m thick and the wall is 0.45 m thick. A uniform vertical Darcy water flux  $q$  of  $4.53 \cdot 10^{-4}$  m/a was calculated in a separate calculation and is assigned to the entire computational domain.

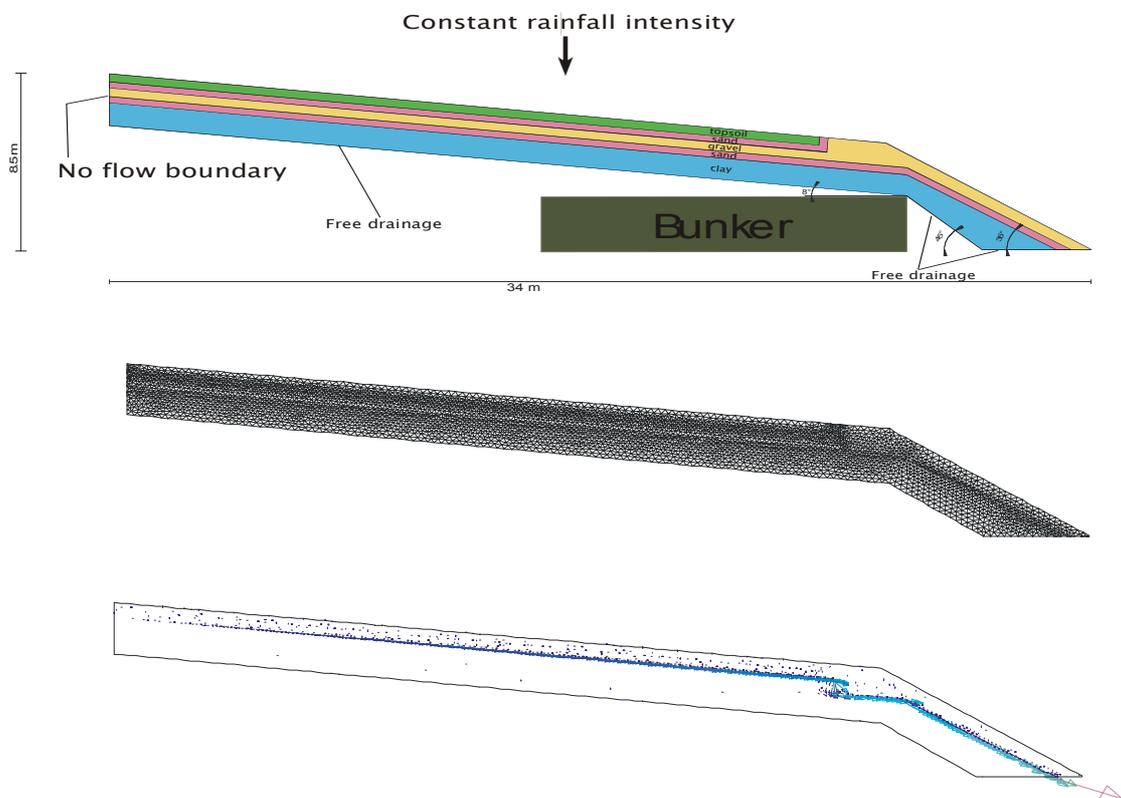


FIG. 3. Conceptual model of multi-layer cover (top), finite element mesh (middle), and simulated flow (bottom).

Boundary conditions for solute transport are as next: zero flux at left and right boundaries and at the top, and a zero gradient at the lower boundary. For each element appropriate transport parameters were assigned to the four materials considered. The PORFLOW code [10] was used to calculate the leaching from the waste zone to the groundwater. Leaching of contaminants from the silos was done using a one-dimensional model because of the much simpler geometry (not discussed here).

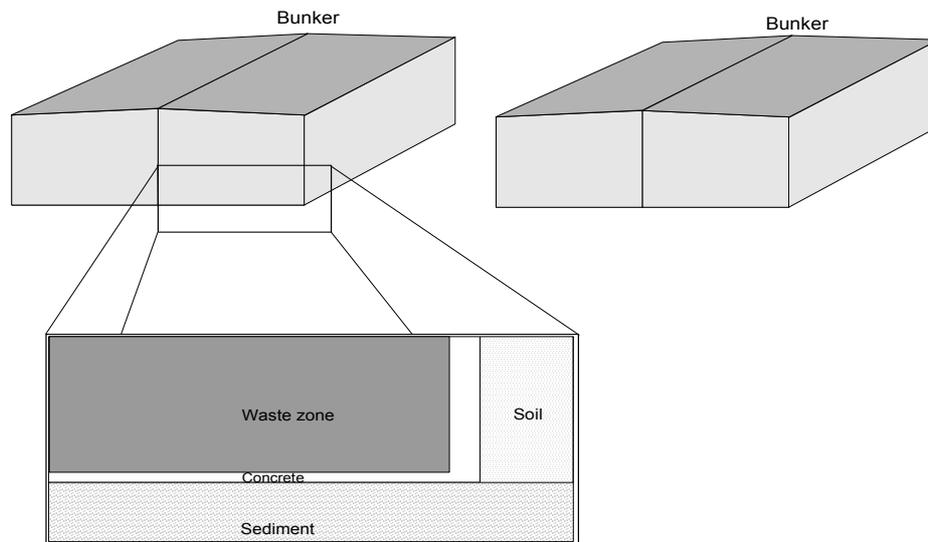
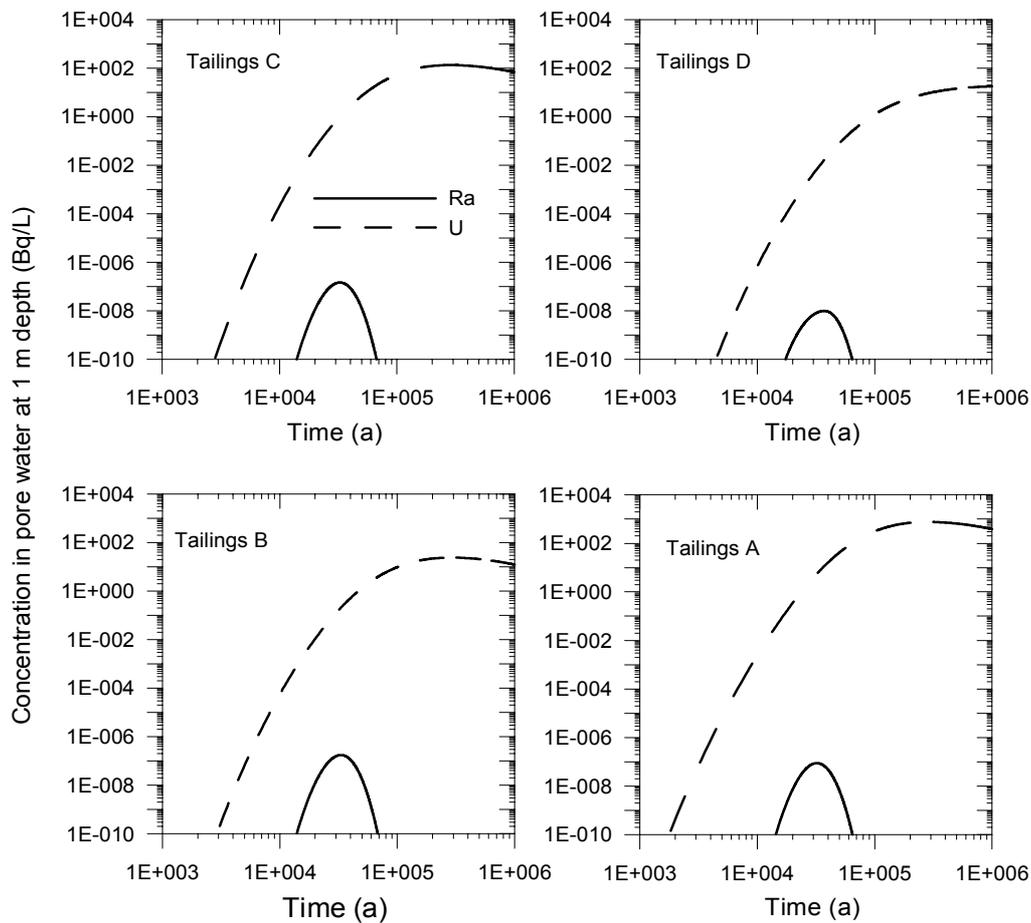


FIG. 4. Conceptual model used to calculate leaching of radium and uranium present in tailings.

## 5. CALCULATED RADIUM AND URANIUM LEACHING

Calculated pore water concentrations  $C_{liq}$  at 1 m below the concrete floor are shown in Figure 5 for all four types of uranium mill tailings. The results clearly illustrate that the radium concentration is negligible by the time it reaches the depth of 1 m below the bottom of the bunker. The results given in Figure 5 demonstrate that radium concentrations are always smaller than  $10^{-7}$  Bq/L (or  $6.5 \cdot 10^{-8}$  Bq/g soil, using  $C_{liq} \times R \times \eta / \rho_b$ , where retardation  $R = 2954$ , total porosity  $\eta = 0.367$  and dry bulk density  $\rho_b = 1666$  g/L). Considering a natural background concentration of about 0.015 Bq/g soil in the Kempen, the radium concentrations at the top of the aquifer owing to leaching are negligible. We also calculated the radium concentrations in case the oxygen would be depleted in the waste zone, i.e., under reducing conditions. In such case, radium is not solubility limited as no sulphates are present to form the  $RaSO_4$  precipitate. This represents the most pessimistic case. In this case the maximum concentration is approximately one order of magnitude higher compared to the solubility limited case. The concentrations in the pore water will also be one order of magnitude higher, but are still very low and hence negligible compared to the background value. Calculated radium concentrations for other waste types are always smaller than those for the uranium mill tailings.

Considerably higher concentrations are observed for uranium, but these concentrations obtain their maximum value only after more than 100 000 year, and are always less than 1000 Bq/L (Figure 5). Furthermore, dilution and dispersion in groundwater has not yet been considered in the present calculations. When considering consumption of this water as drinking water, at an annual rate of  $0.4 \text{ m}^3$ , an annual individual dose of 0.006 Sv/a is calculated (from a dose-conversion factor of  $1.9 \cdot 10^{-8}$  Sv/a per Bq/m<sup>3</sup> for  $^{238}\text{U}$ ). Although this dose rate is higher than the internationally accepted dose limit for the public of  $10^{-3}$  Sv/a, it is reached after a very large time, after more than 10 000 year. For such large time scales, uncertainties about the biosphere, the source term, the hydrogeology, etc. will be large and hence predictions will be unreliable. For instance, forecasts of the future climate, based on Milankovitch's orbital theory [11], indicate that a moderate glaciation might occur within approximately 25 000 years and a more drastic one after 55 000 years. A time cut-off at 10 000 years is therefore considered.



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FIG. 5. Calculated radium and uranium concentrations (Bq/L) at 1 m below the bottom of the bunker for radium rich tailings.

## CONCLUSIONS

An impact study is being carried out to assess the long-term effects of the disposal of radium containing wastes, including uranium mill tailings, on the environment and human health. Given the large number of waste types originating from different waste streams, a two-step procedure was adopted in which all elements (radioactive and non-radioactive) were first screened on their potential health effects using simple and conservative calculations (step 1), followed by a detailed calculation of the leaching behaviour (step 2) of those elements that were found potentially harmful in the first step. Unlike the first step which required only a limited data input, the second step included 1) detailed physico-chemical characterization of the waste forms, the site and the engineered barriers, and 2) detailed modelling of water flow and contaminant transport through various barriers. The results showed that the majority of the contaminants present in the various wastes did not require detailed calculations (i.e., would not lead to groundwater concentrations above health standards), and that detailed characterization and modelling efforts could be restricted to a few elements. The results further revealed that all waste types gave negligible concentrations of  $^{226}\text{Ra}$  in the groundwater and that  $^{238}\text{U}$  concentrations also were negligible the first 10 000 years.

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## DISCUSSION AFTER THE PRESENTATION OF D. MALLANTS

**M. GOLDMAN (USA):** As regards the Olen cleanup, to what extent was the public consulted?

**D. MALLANTS (Belgium):** The Olen cleanup led to a clear improvement, so that the public was very happy about it. Moreover, the fact that many people living in or around Olen worked at the plant facilitated the taking of remediation actions.

