

Global Fallout after the Chernobyl Accident

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# GLOBAL FALLOUT AFTER THE CHERNOBYL ACCIDENT

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## Kurzfassung

Die aus dem zerstörten Reaktorblock 4 in Tschernobyl freigesetzten Radionuklide und deren weltweite Verbreitung sowie regional unterschiedliche Ablagerung wird beschrieben. Die Ablagerung in verschiedenen Ländern wird gegenübergestellt und mit der im UNSCEAR-Bericht 1988 ermittelten, mittleren Bevölkerungsdosis für diese Ländern verglichen, wobei sich erwartungsgemäß viel höhere Werte pro Einheitsdeposition in südlichen Ländern im Vergleich zu nördlicheren ergibt.

Der Fallout aus dem Tschernobylunfall wird mit dem Fallout aus den Kernwaffenversuchen verglichen, und zwar sowohl in bezug auf den globalen Fallout als auch den in einigen ausgewählten Ländern. Die Strahlenexposition im ersten Jahr wird mit dem natürlichen Strahlenpegel in Beziehung gesetzt und die Entwicklung der Strahlenexposition in den folgenden Jahren wird beschrieben.

### Abstract

The radionuclides released from the destroyed reactor bloc 4 in <u>Tschernobyl and</u> the worldwide distribution as well as the regionally varying deposition are described. The deposition in various countries is contrasted and compared to the average population dose in these countries as evaluated by the UNSCEAR-Report 1988. As expected, much higher dose values per unit deposition result for more southerly countries than for northerly ones.

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The fallout from the Chernobyl accident is compared to the fallout from nuclear weapons testing, both with regard to the global fallout and the fallout in some selected countries. The radiation exposure in the first year is contrasted to natural background level and the trend of the radiation exposure in the following years is described.

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## **1. Introduction**

On April 26th, 1986 the most disastrous accident in a nuclear power plant ever occurred in unit 4 of the Chernobyl power station. This accident caused by a series of design disadvantages (positive reactivity coefficient, no fast shutdown system, strong instabilities in the core, etc.) and a series of human errors (trespassing of safety margins, overriding of several safety systems, etc.) led to a complete destruction of the reactor core and any barriers against releases of the radioactive inventory to the environment. Due to the lack of any containment structure with that reactor type a release of radionuclides to the environment occurred which is considered to be the maximum possible release quantity in any type of serious accident in a nuclear power plant of that size.

This release of radionuclides amounted to practically 100 % noble gases, about 20 % iodines and caesium isotopes and various other radionuclides of lesser amount. A detailed estimate of the radionuclides released is given in table 1.

Had there been any type of release mitigating features such as a containment structure typical for Western nuclear power plants, the release and therefore the consequences to the environment would have been lower by several orders of magnitude (1). Due to the lack of such mitigating features and due to the extremely high degradation of the core, the release by this accident may be considered as a maximum possible release in any extreme accident at a large nuclear power plant, though it may not be considered as typical for other power reactors.

Therefore, it may be of great interest to investigate the local and global contamination due to that accident in order to get a better understanding of the maximum contamination by severe accidents at nuclear power plants. This paper, therefore, focusses on the release and dispersion of radionuclides to the environment, their worldwide transport, their transport through food chains to man and the consequent radiation exposures and the longterm behaviour of longlived radionuclides in the environment.

## 2. Release and dispersion of radionuclides after the accident

The activity inventory of a 1000  $MW_e$  power reactor like the Chernobyl reactor is given in table 1. Also in this table the estimated release fraction for the individual radionuclide groups are given. As may be seen from this table, the major releases occur for gaseous and volatile nuclides (gases, iodine, tellurium, caesium) while nuclides with less volatility such as strontium, barium and in particular transuranium nuclides show very small releases even under most severe conditions.

nuc 11de	Half-15	fe	Inventory (EBq)	Percentage
		_		
Kr-85	10.72		0.033	- 100
Xe-133	5.25	٥	1.7	~ 100
1-131	8.04	٥	1.3	20
Te-132	3.26	đ	0.32	15
Cs-137	30.0		0.29	13
Cs-134	2.06	a i	0.19	10
Sr-89	50.5	đ	2.0	
57-90	29.12	4	0.2	
Zr-95	64.0	٥	4,4	i
Ma-99	2.75	đ	4.8	2
Ru-103	19.1	đ	4.3	3
Ru-106	36B	٥	2.1	i
Ba-140	12.7	٥	2.9	6
le-141	32.5	đ	4.4	ž
Ce-144	284	٥	3.2	i
1p-239	2.36	٥	0.14	j
Pu-238	87,74	a	0.00)	3
Pu-239	24065	a -	0.0008	i
Pu-240	6537	a –	0.001	3
241	14.4	ð.	0.17	3
	163	đ.	0 026	

 Table 1:
 Core inventory and release fractions for all significant radionuclides (2)

The release from the destroyed power plant lasted for seven days with a slight decrease after the initial explosion, but a continuing increase in release due to the heating up of the not sufficiently cooled corium in the next nine days. Only after that period a sufficient cover of the core by borium, lead and concrete lead to a significant decrease in release rate with only a minor release thereafter. Due to the long release period and the usual changes in climatic conditions over such prolonged periods, changes in the wind direction and speed are to be expected during the release leading to a transport of the radionuclides in various directions.

Figure 1: Radionuclide transport in the initial phase of the release from the power plant



Descriptive plume behaviour and reported initial arrival imas of detectable activity in air. Plumes A, B, and C correspond to air mass movements originating from Chemobyl on 28 April, 27-28 April, and 29-30 April, respectively. The numbers 1 to 8 indicate initial arrival immes: 1 (28 April), 2 (27 April), 3 (28 April), 4 (22 April), 5 (30 April), 8 (1 May), 7 (2 May), 8 (3 May).

Due to the heating-up of the non-cooled core and partially also due to the graphite fire, the released radionuclides were lifted to an altitude of more than 500 m where they could be transported for long distances. This lead to a global dispersion of the released radionuclides not anticipated to that amount up to now. The major three plumes carrying radionuclides from the power plant in the first 6 days are shown in Fig. 1. The first plume transported the nuclides in northern direction to Finland and Sweden where the environmental monitoring systems of the Swedish reactors gave the first hint in a western country that some major release in the USSR must have occurred. After 2 days a change in wind direction transported the radionuclides released by that time towards the west – Poland, Czechoslovakia, Austria, while the third major plume with radionuclides released at the end of the release period headed towards the South – Romania, Bulgaria, Greece.

The plume direction gives, however, only a first indication whether an area may be exposed or not. The amount of exposure depends not only on the radionuclide concentration in the plume which decreases with increasing distance from the source, but to a very high degree on the amount of radionuclide deposition on the ground. While under dry condition this fallout is rather low, with rainfalls a "washout" with up to three orders of magnitude higher deposition values may occur. This results in possibly much higher deposition values at further downwind distances than at closer distances to the source.

Phenomena such as these were, of course, also observed after the reactor accident at Chernobyl. Thus, much higher deposition values were observed in north-central Sweden than in Finland or even in the USSR north of Chernobyl. Similarly, the plume traversing to central Europe gave rise to low contamination values in Czechoslovakia und Hungary, but to higher levels in Austria, although further downwind. In Austria significant rainfalls at that time led to contamination levels about 2 - 3 times higher than in neighbouring Hungary or Czechoslovakia.



Figure 2: Average values of the total deposition of  $^{134}Cs + ^{137}Cs$  in the OECD countries

The result of these effects was a very heterogeneous deposition of radionuclides all over Europe. A country-averaged estimate for the OECD countries in Europe is displayed in Fig. 2 (3). The  $^{134}Cs + ^{137}Cs$ -deposition values for countries outside the USSR range from 0.004 and 0.08 kBq/m<sup>2</sup> for Spain and Turkey to 23 kBq/m<sup>2</sup> for Austria, the country with the highest deposition values outside the Soviet Union. Norway, a country with more than 1700 km distance showed higher deposition values than Sweden or Finland, both being only 1200 – 1400 km distant.

## 3. Distribution within individual countries

Heterogeneous deposition values were not only observed on nation wide scale, but also inside individual countries. Fig. 3, 4 and 5 show local variations in some countries of Europe. As obvious, even within one country large variations in the radionuclide deposition are observed. Depending on precipitation they may deviate for more than one order of magnitude. Thus in the highlands of Great Britain a deposition of more than 100 times the one observed in the vicinity of London was recorded (3). In Northern Germany the deposition values were about 30 times lower than in Southern Bavaria, the area with the highest deposition values in Germany (4).





## Figure 4: <sup>137</sup>Cs deposition in United Kingdom



Figure 5: <sup>137</sup>Cs deposition in Sweden



The aerial transport of radionuclides from the destroyed power plant had, of course, no halt at the boundaries of Europe. The contaminated air masses continued their way across the Asian part of the Soviet Union and across the seas. Thus, also countries very far away from the site in the Ukraine were hit by the radioactive plume. A few days later the plume passed Japan and shortly there after the high altitude part of the plume was detected over the USA. Similar to the European countries, due to varying rainfalls different deposition values were also observed there. Deposition values of up to 0.7 kBq/m<sup>3</sup> were observed in the northern Rocky Mountain States and in the northernmost part of the Appalaches. But in most parts of the country a deposition about five times lower and in the Eastern and Southeastern part of the US values about 10 times lower were measured (4).

Compared to Europe the average deposition in the United States was, however, lower by more than a factor of 100 and more than a factor of 1000 compared to the country with highest deposition values in Europe outside the Soviet Union (4).



Figure 6: <sup>131</sup>I-deposition in the United States of America

Countries outside these main jet streams circling the globe were of course much less contaminated. The average deposition in India, for instance, amounted to about  $0.035 \text{ kBq/m}^3$  while in the Southern hemisphere virtually no deposition was observed. Australia as an example reported no detectable <sup>137</sup>Cs on soils or vegetation.

## 4. Deposition within the Soviet Union

Strongly varying deposition patterns were, of course, also observed within the USSR. Fig. 7 shows the deposition pattern in the 200 km-vicinity of the Chernobyl plant in the republics of Ukraine and Byelorussia. In the close vicinity of the power plant a pattern as caused by the three main wind directions during the time of release is clearly visible. This pattern is, however, only observable in a range of up to 50 - 70 km, in an area slightly larger than the exclusion zone (5). Outside this area the deposition shows strong variations independent from wind directions. Those were also caused mainly by differences in precipitation during the time of the plume.

Figure 7: <sup>137</sup>Cs deposition in the Ukraine and Byelorussia



In Figure 7 a second significant deposition area apart from the immediate vicinity of the plant at a distance of about 150-250 km in NNO-direction is visible. This area around the town of Gomel shows a deposition with peak values in the range of the highest values outside the exclusion zone (5). On the other hand, the city of Kiev shows a very low caesium deposition. The values there are lower than in the high deposition areas of Austria, more than 1000 km away.

It should be noted that in the area outside the immediately affected zones of Northern Ukraine and Southern Byelorussia depositions are significantly lower. They are comparable to values observed in Central Europe and in most areas lower than deposition values observed in Northern Europe, for instance. In the majority of the country, i.e. in the Asian part, deposition values are comparable or less than in Western Europe (France, UK).

These strong variations in deposition which are caused by differences in precipitation during the passage of the plume require a close observation by public authorities in order to properly protect the public in case of such accidents. A thorough and comprehensive survey of large areas is required during a period of a few weeks after such an accident in order to take the appropriate decisions with regard to longterm countermeasures (measures on foodstuff, dislocation, etc.) in the various regions affected.

## 5. Comparison to the fallout after the atmospheric nuclear weapons tests

The radioactive fallout after the Chernobyl accident was not the first major fallout of radionuclides. Apart from three former reactor accidents with significant core degradation, but only minor or virtually no release to the environment in two cases and a minor release compared to the Chernobyl accident in the third case, a major fallout was observed after the nuclear weapons testing of the fifties and sixties. As this fallout is well documented and, in particular, the environmental behavior, exposure pathways and biological halflives of the major radionuclides are well known from it, a comparison of the Chernobyl fallout with that from the weapons testing may be of great interest, in particular, with regard to the longterm environmental trends of radionuclides with long physical halflives.

The worldwide transport of debris from nuclear weapons is similar to that from the Chernobyl accident. As an example, figure 8 shows the path of the radioactive cloud after the Chinese test of October 1980. The trajectory via northern China, Japan, the USA and finally Europe is clearly visible and was easily detected by the monitor networks of the countries affected (6).

Figure 8: Trajectories derived from meteorological data and confirmed by ground-level activity measurements of the atmospheric nuclear explosion of 16 Oct. 1980



There is, however, a significant general difference between the nuclear weapons fallout and the Chernobyl fallout. While as a consequence of the Chernobyl fallout more or less only a single deposition occurred, the number of nuclear tests amounted to more than 50, each one causing a fallout trajectory over the globe. Also the injection into the atmosphere occurred into higher layers which caused a much longer circulation and prolonged deposition around the globe. Due to that the contamination by the nuclear weapons testing is much more homogeneous worldwide than due to the Chernobyl fallout. The deposition virtually represents the longterm precipitation levels, not the single rainfall situation at a given date as with the Chernobyl accident. A global pattern is observable in which two peaks are visible, one in the northern and one in the southern hemisphere, the maximum of both being at approximately 40° latitude (see Fig. 9).

Figure 9: Latitudinal distribution of <sup>137</sup>Cs deposition density and the concentration of <sup>137</sup>Cs in milk, 1972 (6)



There are further significant differences in the radionuclide distribution between weapons and fallout the Chernobyl fallout. While in weapons fallout both Sr and Cs isotopes, but also Pu isotopes are released with high probability, the release from a molten core differs significantly for these nuclides. Caesium with a rather low melting temperature is more volatile than Sr, while Plutonium is only released at very high temperatures and even there only in minor amounts. Therefore, in the release from a molten core Cs always will show a much higher fraction than Sr, and Pu would be expected to be orders of magnitude lower. This was also observed after the Chernobyl accident. The major dose contribution with this accident came from the Cs isotopes (and <sup>131</sup>I), while the major dose contributor with the weapons testing was <sup>90</sup>Sr and <sup>89</sup>Sr and even more so <sup>239</sup>Pu.

A question often addressed to environmental radiation scientists is which of the two fallouts – that after the nuclear weapons tests or that after the Chernobyl accident – was bigger. Let us consider first the worldwide situation. This is shown in table 2 in which the total global deposition after the weapons testing and the accident both for  $^{90}$ Sr and  $^{137}$ Cs are given. As is clearly visible, the global fallout after the weapons testing was approximately 60 times higher for  $^{90}$ Sr and about 25 times higher for  $^{137}$ Cs than for the Chernobyl accident.

	ТОТ	TOTAL DEPOSITION (10 <sup>16</sup> Bq)			
		<sup>90</sup> Sr	<sup>137</sup> Cs		
Nuclear	Global Northern	60.4	96.6		
Weapons Testing	Hemisphere Southern	46.0	73.6		
	Hemisphere	14.4	23.0		
Chernobyl	Global	1.0	3.8		
Accident	Europe	0.8	3.0		

Table 2:Total global deposition of 90Sr and 137Cs after weapons testing and the<br/>Chernobyl accident (2, 6)

Due to the large local variations in the Chernobyl fallout and the wide spectra of fallout in different countries worldwide, a comparison of the global deposition is not a good description for the situation in individual countries. Therefore, in table 3 a comparison of the Chernobyl to the weapons fallout is shown for a few specific countries. For Austria the <sup>137</sup>Cs fallout after Chernobyl was on the average about 4 times higher than after the weapons tests, while in Great Britain it amounted only to one third of the weapons tests' fallout. In Turkey, on the other hand, the Chernobyl fallout was only about 2 % of that of the weapons testing period and in the United States only about 1 %.

Cumulative deposition [kBq/m <sup>2</sup> ]							
Country	<sup>137</sup> Cs Chernobyl weapons testing		<sup>90</sup> Sr Chernobyl weapons testing				
Austria	23	5.2	0.9	3.3			
UK	1.4	4.1	0.04	2.6			
Turkey	0.08	~4	0.003	~2.5			
USA	0.03	2.6	0.001	1.6			

Table 3:Regional cumulative deposition of 90Sr and 137Cs after weapons testing and the<br/>Chernobyl accident

In all countries considered and also in the USSR, except for the immediate area around the destroyed power plant, the fallout of  ${}^{90}$ Sr was lower than after the weapons testing; in Britain it amounted only to 1.5 %, and in Turkey to about 0.1 % of the weapons test fallout. Because of the high radiotoxicity of  ${}^{90}$ Sr, a significantly lower radiation burden to the population, especially infants, therefore, was observed after Chernobyl.

## 6. Dose estimates

Equal deposition values do not necessarily result in equal dose values to the population. Only with regard to external radiation, equal deposition levels would result in approximately equal exposures. A major part of the exposure is, however, caused by uptake of radionuclides via inhalation and more so by ingestion. The dose received by the population in this case depends significantly on the contamination of foodstuffs which in turn is primarily a result of the interception of radionuclides by plants. This interception increases with plant size and thus the contamination level of all foodstuff directly or indirectly derived from plants. This concerns vegetables and fruit as well as milk and meat.

As the plant size depends largely on the growth stage which is more advanced in a southern than a northern country in May, a comparison of the contamination levels of foodstuff in various countries is of great interest. Due to lack of space this comparison shall not be performed for all relevant foodstuff, but rather by comparing the actual ingestion dose received by the population in different regions worldwide. This comparison includes all types of contaminated foodstuff contributing to the ingestion dose.



Figure 10: Country-wide average adult thyroid dose equivalents from the Chernobyl accident

A comparison in the thyroid dose due to ingestion of <sup>131</sup>I is given in Fig. 10. A wide range of dose values for different countries is seen, with Yugoslavia and Greece at the upper end and non-European countries as well as Spain and Portugal, as expected, at the lower end. The data clearly demonstrate that countries closer to the USSR and with higher fallout rates showed higher incorporation values than those at farther distance or less fallout. The reason for the two southern European countries to head the <sup>131</sup>I-ingestion dose is that plants in these countries show an already more advanced growth stage at the beginning of May and therefore higher interception of <sup>131</sup>I by the plants causing higher iodine doses via milk and fresh vegetables.

The data in Figure 10 show the thyroid dose of a population without any countermeasures adopted. Taking into consideration that a considerable number of countries adopted some countermeasures in the early phase with regard to milk and fresh vegetables, the actual <sup>131</sup>I doses should be lower.

Figure 11 shows a comparison of the effective dose equivalent for all radionuclides contributing in the Chernobyl fallout in the first year after the accident. Because of rainfalls Bulgaria and Austria show the highest ingestion dose values, while countries such as Czechoslovakia or Poland, although closer, show lower ingestion dose values. The data represent country wide averages, which puts the USSR in the middle of the scale.

Again the values represent doses to be expected without countermeasures. In the case of Austria, for instance, where quite significant efforts to reduce the dose had been undertaken, a reduction of about 0,3 mSv or 1/3 of the expected value was observed (7).

Figure 11: Country-wide average first-year committed effective dose equivalents from the Chernobyl accident



## 7. Comparison to other radiation burdens

How high was the radiation exposure? If we consider, for example, Austria as a country with one of the highest exposures outside the USSR, we find an average exposure (including countermeasures) of 0.5 mSv(8). This includes all exposure pathways (direct radiation from the plume and deposition on ground, inhalation and ingestion of contaminated foodstuff (see Fig. 12)). The exposure varied for different parts of the country, but the lowest and highest exposures did not deviate by more than a factor of 1.5 from the average (9).







If we compare this to the radiation exposure by natural radiation as displayed in the right part of the figure, we find that the total radiation dose in this country due the Chernobyl accident was less than one fifth of the average annual natural radiation exposure perannum.

This is, however, only true for the first year after the accident. In the following years as we will see in the next chapter, the dose due to ingestion will significantly decrease as Cs is readily fixed to the soil. The natural radiation exposure will, on the other hand, remain constant as it had been for millions of years.

It is obvious that the radiation exposure in all countries outside the Soviet Union was significantly less than the annual natural radiation level and the life time dose of persons exposed in these countries will not exceed 1 % of the natural radiation exposure.

## 8. Longterm Caesium exposure

<sup>137</sup>Cs possesses a halflife of 30 years. However, the exposure due to this radionuclide does not decrease only according to its physical halflife. After one year caesium is well fixed to the soil and this process progresses as time goes on. Thereby, Cs will be less available to plant roots and thus for the future uptake into the plant. This causes a progressing elimination of <sup>137</sup>Cs from the environmental biocycles. It will, therefore, be less available to the human diet in the following years. Also Cs slowly drains into the soil thus causing less and less external exposure in the coming years.





Both effects were observed after the nuclear weapons testing more than twenty years ago. The reduction in availability of Cs and Sr throughout the years after the tests is best illustrated by the annual activity intake of these radionuclides by the population. This is displayed in Fig. 13. A progressing decrease in the <sup>137</sup>Cs and <sup>90</sup>Sr intake over the years is observed. For <sup>137</sup>Cs this decrease is by a halflife of 4.5 years, for <sup>90</sup>Sr of 8.5 years (10). The extrapolation of these values may be somewhat conservative to be transferred to the Chernobyl accident as even after the major test period's end in 1964 a number of smaller bombs were exploded by China and France injecting further radioactive material into the atmosphere and causing a prolonged, but smaller fallout.

Figure 14: Ingestion dose due to Chernobyl in the years following the accident (10)



Considering this reduction after the weapons testing, a significant reduction of the ingestion dose should also be expected after the Chernobyl fallout in the years after the first exposure. In Fig. 14 we see the reduction in the ingestion in the first 5 years following the reactor accident. While <sup>131</sup>I contributes only in the first year and causes no contribution in the following years because of its short half life, the Cs-isotopes remain due to their long halflives. But they also show a considerable decrease in the following years. For <sup>134</sup>Cs with a halflife of 2.06 years the reduction is more significant, but also with <sup>137</sup>Cs (t<sub>1/2</sub>=30 a) as expected a substantial reduction is observed throughout the following years.

The reduction in Cs intake and in the consequent ingestion dose is, of course, caused by the reduction in <sup>137</sup>Cs-activity concentration in virtually all foodstuff, both plants and animal products, which is caused by progressing fixation of Caesium in the soil. This reduction may be seen in the following figures. The time course for milk is given in Fig. 15. In winter 1986/87 a second rise to about half of the peak values of May is observed which is





caused by the feeding of hay produced in May and June of that year. With the feeding of fresh grass in May 1987 a significant decrease is observed which progresses throughout the following years as the Cs in the grass roots is more and more fixed to the soil and becomes less available to the roots.

Similar effects are observed for cereals. In Figure 16 data on activity concentrations in the first and second year after the accident are given. The differences between summer and winter cereals in the first year are caused by different growth stages of the plant at the time of accident. Similar reductions are observed for all other farmers'products. As an example I would like to show the activity concentrations in Turkish hazelnuts as they were exported to Austria and measured in our laboratory. As expected, also these show a significant reduction of the activity content over the years with values that are barely measurable today.









These reductions in activity concentrations result in very low longterm exposures from the Chernobyl accident in all countries outside the area in the vicinity of the power plant. The doses in the following years worldwide will be less than 0.001 mSv or a small fraction of the natural radiation exposure. Even in the countries with the highest exposure outside the USSR the future annual exposure will be less than one fiftieth of natural exposure levels.

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