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**CHERNOBYL SOURCE TERM, ATMOSPHERIC DISPERSION, AND  
DOSE ESTIMATION****P. H. Gudiksen, T. F. Harvey, and R. Lange****Prepared for presentation at the  
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# CHERNOBYL SOURCE TERM, ATMOSPHERIC DISPERSION, AND DOSE ESTIMATION

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## ABSTRACT

The Chernobyl source term available for long range transport was estimated by integration of radiological measurements with atmospheric dispersion modeling, and by reactor core radionuclide inventory estimation in conjunction with WASH-1400 release fractions associated with specific chemical groups. These analyses indicated that essentially all of the noble gases, 80% of the radioiodines, 40% of the radiocesium, 10% of the tellurium, and about 1% or less of the more refractory elements were released.

Atmospheric dispersion modeling of the radioactive cloud over the Northern Hemisphere revealed that the cloud became segmented during the first day, with the lower section heading toward Scandinavia and the upper part heading in a southeasterly direction with subsequent transport across Asia to Japan, the North Pacific, and the west coast of North America. The inhalation doses due to direct cloud exposure were estimated to exceed 10 mGy near the Chernobyl area, to range between 0.1 and 0.001 mGy within most of Europe, and to be generally less than 0.00001 mGy within the U.S.

The Chernobyl source term was several orders of magnitude greater than those associated with the Windscale and TMI reactor accidents, while the  $^{137}\text{Cs}$  from the Chernobyl event is about 6% of that released by the U.S. and U.S.S.R atmospheric nuclear weapon tests.

## INTRODUCTION

Evidence from environmental radiation measurements throughout the Northern Hemisphere and in particular, within Europe, indicate that the release of radioactivity from the Chernobyl-4 reactor accident in the Soviet Union on April 26, 1986 contained a wide spectrum of fission and activation products. The most important radionuclides of concern were I-131,  $^{137}\text{Cs}$ , and  $^{134}\text{Cs}$  due to their relative abundance as well as their radiological and chemical characteristics. Some fraction of the radioactivity, which was released by the initial explosion and the subsequent graphite fire, was apparently lofted into the middle troposphere as evidenced by its rapid transport over the Northern Hemisphere. The staff at the Atmospheric Release Advisory Center (ARAC), situated at the Lawrence Livermore National Laboratory, utilized a three-dimensional atmospheric dispersion model to derive the spatial and temporal evolution of the radioactivity distribution over local to hemispheric spatial scales (Di83). Comparison of these distributions with the radiological measurements made throughout the Northern Hemisphere permitted estimation of the source term for those radionuclides that were available for long range transport and were measured in the airborne debris collected outside the Soviet Union. The release rates for

radionuclides that were not generally measured, such as  $^{90}\text{Sr}$ , were derived by first estimating the core's radionuclide inventory with a nuclear physics model, and then using the measured radionuclides as tracers of the WASH-1400 designated chemical groups to obtain the release fractions for each group of radionuclides. This paper summarizes our estimates of the source term available for long range transport, the hemispheric dispersion of the radioactivity, and the resulting doses due to direct cloud exposure. Also included is a comparison of the activity released by the Chernobyl accident with those for previous nuclear events.

Most of the radiological measurements used in this work were acquired from numerous informal reports prepared by various scientific organizations in Denmark, Finland, France, Japan, Kuwait, Netherlands, Norway, Sweden, United Kingdom, United States, and West Germany as well as from the World Health Organization (WHO) and the International Atomic Energy Agency (IAEA).

### SOURCE TERM ESTIMATION

Estimates of the radionuclide specific release rates were obtained by integrating the radiological measurements with the results of hemispheric-scale atmospheric dispersion modeling. This involves optimizing the agreement between the model calculated radioactivity distributions and the radiological measurements. A three-dimensional particle-in-cell transport and diffusion model (PATRIC) driven by Northern Hemispheric wind fields supplied by the U.S. Air Force Global Weather Central was used to perform the analysis. The PATRIC model (La78a and La78b) utilizes the principle of generating a large number of marker particles to represent the radioactivity. The particles are injected at the source point and subsequently transported within the three-dimensional Eulerian grid mesh by means of a transport velocity applied to each particle. This transport velocity consists of a wind velocity provided at each grid point and a diffusion velocity. In addition, gravitational settling and dry deposition velocity vectors as well as radioactive decay may be applied to the particles, as appropriate. Summing the resulting distribution of particles over the grid mesh volumes allows determination of the three-dimensional radioactivity distributions. The calculations were based on computational meshes that ranged from the 50 km scale surrounding the reactor to hemispheric scale.

Measurements of airborne radioactivity over Europe, Japan, and the U.S. detected the presence of fresh fission products up to heights of about 7 km within a few days after the initial explosion. This suggests that some of the radioactivity released by the explosion and the subsequent fire within the reactor core must have been transported to heights well within the middle troposphere. This high altitude presence of the radioactivity may have been due to a variety of factors such as the thermal energy associated with the releases, rapid atmospheric mixing due to the presence of thunderstorms in the vicinity of the Chernobyl area during the first day of the accident or the possible uplifting of the radioactive debris over a warm front situated between Chernobyl and the Baltic Sea. Thus, the source term used in the model calculations included an upper level cloud simulating the initial explosion and a lower level cloud for the ensuing fire. By optimizing the agreement between the cloud arrival times and duration of peak concentrations measured at over 20 sites in Europe, Kuwait, and the U.S. and aircraft sampling missions over Europe, Japan

and the U.S. with the model calculations, it was possible by an iterative process to derive the best fit to the initial vertical distribution of radioactivity in the atmosphere. This optimization required the upper level cloud to be centered at 4500 m with a vertical extent ranging from 1500 to 7500 m and the lower cloud was centered at 1300 m and extended from the surface to 1500 m. Scaling of the calculated concentrations with those measured then permitted a derivation of the total amount of  $^{131}\text{I}$  and  $^{137}\text{Cs}$  released. The release rates of other radionuclides were acquired by calculating their ratios relative to  $^{137}\text{Cs}$  from measurements of airborne radioactivity within Scandinavia. This lead to the estimated releases given in Table 1.

## ESTIMATION OF CORE INVENTORY AND RELEASE FRACTIONS

To estimate the inventory of specific radionuclides in the RBMK-1000 Soviet reactor core at the time of the accident, we utilized the FORIG model (BI85), which is an LLNL version of the ORIGEN2 model (Cr80) that was developed at the Oak Ridge National Laboratories. These numerical models solve a large number of rate equations to simulate fission, radioactive decay, nuclear transformations, and neutron reactions in order to calculate the build-up and decay of the numerous radionuclides residing in a reactor core. Input to the model included data on the reactor's operating history obtained from the Soviet report presented at the IAEA Experts' Meeting in Vienna (USSR86).

The Chernobyl-4 reactor is thought to have started operation during December 1983 and operated continuously for about 875 days until April 26, 1986. At the time of the accident the core-averaged fuel burnup was 10.3 MWd/kg. The reactor is capable of being refueled during operation and utilizes 2% enriched fuel. To reconstruct its operating history, we assumed that (1) the average load factor was 80%, (2) an 8-month startup period for reaching full power, (3) a continuous refueling schedule of 3% fuel replacement per month commencing in August 1985 to give a core loading at the time of the accident of 75% original fuel, and (4) the core-averaged neutron energy spectrum was similar to that in a commercial PWR, but at a lower power density. On the basis of these assumptions, one obtains the fuel burn-up rate given in Fig. 1. This general description of the reactor's operating history permits estimation of the inventory of the long-lived radionuclides; however, a more detailed history of the reactor power level during the last month and even the last day of operation is needed to estimate the inventory of the short lived radionuclides having half-lives of the order of hours to weeks. Since such detailed information is not presently available, an iterative approach, utilizing isotope ratios obtained from measurements of airborne radioactivity collected in western Europe, was employed. Certain isotope ratios are sensitive to the recent average power levels. This approach involved varying the power level in our calculations for the last month of the reactor's operation to obtain agreement between the calculated and the measured isotope ratios of iodine, ruthenium, cesium, and cerium. Only isotope ratios of the same chemical elements were used in order to minimize fractionation effects. This agreement between the calculated and the measured values is illustrated in Table 2. Almost exact agreement was achieved except for ruthenium and  $^{134}\text{Cs}/^{137}\text{Cs}$  ratios. The ruthenium difference is thought to be due to the Ru-106 inventory being very sensitive to the choice of neutron spectrum, whereas the cesium difference may be due to the different specific activities between the original fuel and the new fuel. Note that the calculated  $^{134}\text{Cs}/^{137}\text{Cs}$  ratio is slightly higher than that measured. More detailed

inspection of the measurements revealed a time varying activity ratio that decreased with time. This variation is believed to be due to the older fuel being preferentially ejected early in the accident because of its greater specific activity and, thus, higher thermal density. On the basis of measurements of airborne radioactivity collected in the U.K. (Ca87), the older fuel appeared to have a ratio of about 0.66, while the newer fuel, which is believed to have been primarily released somewhat later from cooler fuel assemblies, exhibited a value of about 0.50. This iterative approach produced an estimate of the core's inventory of a large suite of radionuclides at the time of the accident. The inventory of the radionuclides of primary interest from an environmental impact point of view is given in Table 3. A review of the data in the table reveals that we estimate the core to have contained 1700 PBq (45 MCi) of  $^{131}\text{I}$ , 200 PBq (5.5 MCi) of  $^{137}\text{Cs}$ , and 130 PBq (3.5 MCi) of  $^{134}\text{Cs}$ . These values have been decay-corrected to 3 days after the accident.

From the releases given in Table 1, and the core inventories listed in Table 2, we obtained the fraction released and available for long range transport for those radionuclides that were measured in the airborne radioactivity sampled outside the Soviet Union. These release fractions may be extended to a host of unmeasured radionuclides by using those listed in Table 1 as tracers of their corresponding chemical transport groups; groups of radionuclides of similar volatility and chemical behavior as defined by the USAEC reactor safety study (Ri75). This is illustrated in Table 4 which depicts the Chernobyl release fractions. These reveal that essentially all of the noble gases, 80% of the iodines and about 40% of the cesium radionuclides were released. This was followed by the release of about 10% of the tellurium and 1% or less of the more refractory elements. These release rates are quite comparable to those estimated from the WASH-1400 study for a major meltdown of a light water reactor and are also similar to those observed during the Windscale reactor accident involving a meltdown of 8 tonnes of fuel. Thus, it is reasonable to assume that a major fraction of the Chernobyl core underwent a nearly complete meltdown.

On the basis of the release fractions given in Table 4 and the core inventories listed in Table 3, we obtained estimates of the total activity released for radionuclides of interest that were not generally measured. These are given in Table 5. Most notable is  $^{90}\text{Sr}$  whose release rate is estimated from Table 5 to be only about 2% of that for  $^{137}\text{Cs}$  due to its low volatility. This seems in reasonable agreement with a few measurements in western Europe.

## DOSE ESTIMATION DUE TO DIRECT CLOUD EXPOSURE

Using these source terms in conjunction with the Northern Hemispheric wind fields, the dispersion model derived the three-dimensional distributions of radioactivity as a function of time over Europe and the Northern Hemisphere. This is illustrated in Fig. 2 by the plan views of the cloud on the second, fourth, sixth, and tenth days after the initiation of the accident. Detailed analysis of these spatial distributions indicate that the cloud became segmented during the first day, with the lower section heading toward Scandinavia and subsequently over central Europe, while the upper part headed in an easterly direction across Asia to Japan, the North Pacific, and the North American continent. The distributions in Fig. 2 were translated to the radiation dose resulting from inhalation of the radioactive debris, and that due to external exposure to the airborne radioactivity. The inhalation

dose distributions over various spatial scales ranging from hemispheric to local scale are shown in Fig. 3. A review of Figures 3a and 3b reveals that within a region encompassing the western U.S.S.R., northeastern Poland, parts of Sweden and eastern Europe, the inhalation dose is 0.1 mGy. In most of central Europe, parts of northern Scandinavia and the remainder of eastern Europe, the inhalation dose is between 0.01 and 0.1 mGy. Denmark, the United Kingdom, and Spain received less than 0.01 mGy. By contrast, the corresponding dose within the U.S. is less than 0.00001 mGy. Figures 3b and 3c provide estimates of the inhalation doses closer to the Chernobyl area where the values range up to approximately 100 mGy. These values, however, appear to be about a factor of 3 lower than those reported by the Soviets. This may be due to the source term used in these estimates not including the radioactivity that contributed to local fall-out. About 80% of the calculated dose values reported here are due to the radioiodines, while the cesium, ruthenium, and tellurium radionuclides are the major contributors to the remaining 20%. Because the spatial distribution of the dose due to immersion in the radioactive cloud is essentially identical to those shown in Fig. 3, one may approximate the immersion dose received during cloud passage by multiplying the isopleths in Fig. 3 by 0.02. This factor, however, is spatially dependent within a factor of about 2 due to the time varying activity ratios of the radioiodine.

#### COMPARISON OF CHERNOBYL WITH OTHER NUCLEAR EVENTS

In order to place the radioactivity released from the Chernobyl reactor in perspective with radioactivity releases from previous nuclear events, it is useful to compare the Chernobyl release with the radioactivity estimated to have been produced by the U.S. and the U.S.S.R. atmospheric nuclear weapons testing programs, as well as with the releases associated with the TMI and the Windscale reactor accidents. However, one should be aware that a complete comparison of the radiological impact of the atmospheric weapon testing programs with that produced by the Chernobyl reactor accident is very difficult. This is partly due to the fact that weapon tests produce mixtures of radionuclides that are relatively more abundant in short lived radionuclides, with half-lives of less than a few days, in comparison with the mixture of radionuclides that may be found in the core of a reactor having been operated over periods ranging from months to years. In addition, the weapon tests injected a large fraction of the radioactivity into the stratosphere with the result that the activity was dispersed globally and did not reach the earth's surface for months and years allowing considerable time for radioactive decay. Furthermore, the weapon tests were conducted at several isolated test sites where, with only a few exceptions, no one was exposed to the immediate effects of the tests. This is in contrast to the Chernobyl event, which released principally noble gases and volatile radionuclides that were relatively more abundant in the long lived species, such as  $^{137}\text{Cs}$ . The accident exposed operating personnel and emergency workers to radiation doses causing fatal injuries and acute radiation sickness, as well as exposing adjacent population centers to dose levels requiring evacuation. Nevertheless, it is still useful to compare the data in Table 6, which shows for the most notable radionuclides of interest the amounts released by the weapon tests as well as the TMI and Windscale (Cl74) reactor accidents. Except for TMI, the release estimates have been decay-corrected to three days after the events for ease of comparison. A comparison of the data shows that with the exception of  $^{134}\text{Cs}$ , the activity

released by the Chernobyl accident is minor relative to the weapon test releases, which are based on 225 Mt of fission. The 200 PBq (2.4 MCi) of  $^{137}\text{Cs}$  released by the Chernobyl event is only 6% of that produced by the weapon tests, while the remaining radionuclide releases represent less than 1% of the corresponding weapon test releases. However, since  $^{134}\text{Cs}$  is not significantly produced in weapon tests, the Chernobyl contribution is at least 30 times greater than that produced by the weapon testing programs. One may also note that the Chernobyl releases were greater by at least several orders of magnitude than those associated with the Windscale and TMI reactor accidents.

## SUMMARY

The Atmospheric Release Advisory Center (ARAC) utilized long-range atmospheric dispersion modeling in conjunction with radiological measurements over the Northern Hemisphere to estimate the source term for the radionuclides measured in the airborne radioactivity. By using a nuclear physics model to calculate the expected core radionuclide inventory at the time of the accident and WASH-1400 release fractions associated with specific chemical groups, it was possible to estimate the source term available for long range transport for radionuclides that were largely unmeasured. Thus, we estimate that essentially all of the noble gases, 80% of the radioiodines, 40% of the radiocesium, 10% of the tellurium, and about 1% or less of the more refractory elements were released.

On the basis of this source term information and Northern Hemispheric wind fields, we utilized a three-dimensional atmospheric dispersion model to derive the evolution of the radioactivity distributions over Europe and the Northern Hemisphere. These indicated that the cloud became segmented during the first day, with the lower section heading toward Scandinavia and the upper part heading in a southeasterly direction with subsequent transport across Asia to Japan, the North Pacific, and the west coast of North America. Integrating these distributions revealed that the inhalation doses near the Chernobyl area exceeded 10 mGy, while for most of Europe the inhalation doses varied from 0.1 to 0.001 mGy. The corresponding doses within the U.S. were less than 0.00001 mGy.

The source term for the Chernobyl accident was several orders of magnitude greater than those associated with the Windscale and TMI reactor accidents, while our estimates of the  $^{137}\text{Cs}$  released by the Chernobyl event is about 6% of that released by the U.S. and U.S.S.R. atmospheric nuclear weapon tests.

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Table 1. Estimated releases of radionuclides from the Chernobyl event. The activities are decay-corrected to three days after the accident. 1 PBq =  $10^{15}$  Bq.

Radionuclide	Activity Released	
	PBq	MCi
<sup>95</sup> Zr	8.5	0.23
<sup>103</sup> Ru	28.	0.76
<sup>106</sup> Ru	5.9	0.16
<sup>131</sup> I	1300.	36.
<sup>133</sup> I	340.	9.1
<sup>132</sup> Te	200.	5.3
<sup>134</sup> Cs	48.	1.3
<sup>136</sup> Cs	17.	0.47
<sup>137</sup> Cs	89.	2.4
<sup>140</sup> Ba	37.	1.0
<sup>141</sup> Ce	8.5	0.23
<sup>144</sup> Ce	5.2	0.14

Table 2. Fission product activity ratios measured in environmental samples and predicted by the FORIG model. The latter are core averaged values.

Activity Ratio	Measured	FORIG Model Estimates
$^{103}\text{Ru}/^{106}\text{Ru}$	4.5	3.3
$^{134}\text{Cs}/^{137}\text{Cs}$	0.54	0.64
$^{136}\text{Cs}/^{137}\text{Cs}$	0.27	0.28
$^{134}\text{Cs}/^{136}\text{Cs}$	2.4	2.3
$^{131}\text{I}/^{133}\text{I}$	0.47	0.46
$^{141}\text{Ce}/^{144}\text{Ce}$	1.6	1.3

Table 3. Estimates of the core inventory of selected radionuclides as predicted by the FORIG model. The activities are decay-corrected to three days after the accident.

Radionuclide	Inventory	
	PBq	MCi
<sup>85</sup> Kr	19.	0.5
<sup>89</sup> Sr	2300.	63.
<sup>90</sup> Sr	150.	4.1
<sup>91</sup> Y	3000.	82.
<sup>95</sup> Zr	4100.	110.
<sup>99</sup> Mo	2200.	59.
<sup>103</sup> Ru	3100.	84.
<sup>106</sup> Ru	1000.	27.
<sup>127</sup> Sb	150.	4.0
<sup>127m</sup> Te	33.	0.9
<sup>129m</sup> Te	96.	2.6
<sup>131</sup> I	1700.	45.
<sup>131m</sup> Te	63.	1.7
<sup>132</sup> Te	1900.	51.
<sup>133</sup> I	440.	12.
<sup>133</sup> Xe	3700.	99.
<sup>133m</sup> Xe	89.	2.4
<sup>134</sup> Cs	130.	3.5
<sup>136</sup> Cs	48.	1.3
<sup>137</sup> Cs	200.	5.5
<sup>140</sup> Ba	3100.	84.
<sup>140</sup> La	3400.	92.
<sup>141</sup> Ce	3600.	96.
<sup>144</sup> Ce	2800.	76.
<sup>147</sup> Pm	440.	12.

Table 4. Estimated fractional releases of radionuclides available for long range transport from Chernobyl compared with the USAEC reactor safety study (WASH-1400) and the Windscale reactor accident.

Chemical Group	Tracers	Chernobyl Estimate (%)	WASH-1400 (%)		
			Range Estimate	Best Estimate	Windscale Estimate (%)
Noble Gases <sup>(a)</sup>	<sup>133</sup> Xe	100.	50-100	90	100.
Halogens <sup>(b)</sup>	<sup>131</sup> I, <sup>133</sup> I	80.	50-100	90	20.
Alkali Metals <sup>(c)</sup>	<sup>134</sup> Cs, <sup>136</sup> Cs, <sup>137</sup> Cs	40.	40-90	80	20.
Tellurium Group <sup>(d)</sup>	<sup>132</sup> Te	10.	5-25	15	20. <sup>(i)</sup>
Alkaline Earths <sup>(e)</sup>	<sup>140</sup> Ba	1.2	2-20	10	.2
Noble Metals <sup>(f)</sup>	<sup>103</sup> Ru, <sup>106</sup> Ru	0.9	1-10	3	2. <sup>(i)</sup>
Rare Earths <sup>(g)</sup>	<sup>141</sup> Ce, <sup>144</sup> Ce	0.2	.01-1	.3	.2
Refractory Oxides <sup>(h)</sup>	<sup>95</sup> Zr	0.2	.01-1	.3	.2

(a) Xe, Kr

(b) I, Br

(c) Cs, Rb

(d) Te, Se, Sb

(e) Ba, Sr

(f) Includes Ru, Rh, Pd, Mo, Tc

(g) Includes Y, La, Ce, Pr, Nd, Pm, Sm, Eu, Np, Pu, Cm

(h) Zr, Nb

(i) Sb was estimated at 0.2% by Clarke<sup>(8)</sup>

(j) Rh and Pd were estimated at 0.2% by Clarke<sup>(8)</sup>

Table 5. Estimated releases of radionuclides that were largely unmeasured in airborne radioactivity collected outside the Soviet Union. The activities are decay-corrected to three days after the accident.

Radionuclide	Activity Released	
	PBq	MCi
<sup>85</sup> Kr	19.	0.5
<sup>89</sup> Sr	28.	0.75
<sup>90</sup> Sr	1.9	0.05
<sup>91</sup> Y	5.9	0.16
<sup>99</sup> Mo	20.	0.53
<sup>127</sup> Sb	15.	0.40
<sup>127m</sup> Te	3.3	0.09
<sup>129m</sup> Te	9.6	0.26
<sup>131m</sup> Te	6.3	0.17
<sup>133</sup> Xe	3700.	99.
<sup>133m</sup> Xe	89.	2.4
<sup>140</sup> La	6.7	0.18
<sup>147</sup> Pm	1.1	0.03

Table 6. Comparison of Chernobyl with other nuclear events. Except for TMI, the activities are decay-corrected to three days after the events.

Nuclide	Radioactivity Released (PBq)			
	Chernobyl	Weapon Tests	Windscale	TMI
<sup>137</sup> Cs	89.	1500.	0.04	ND
<sup>134</sup> Cs	48.	<1.5	0.001	ND
<sup>90</sup> Sr	1.9	1300.	$2.2 \times 10^{-4}$	ND
<sup>133</sup> Xe	3700.	$2 \times 10^6$	14.	370.
<sup>131</sup> I	1300.	$7.8 \times 10^5$	0.6	0.001

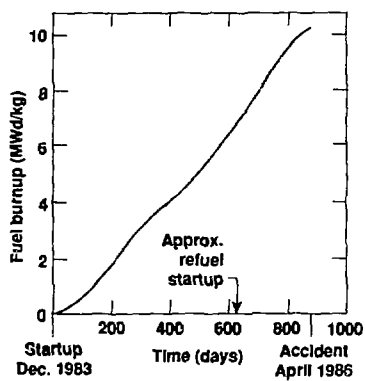
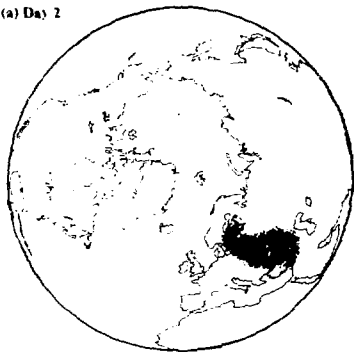
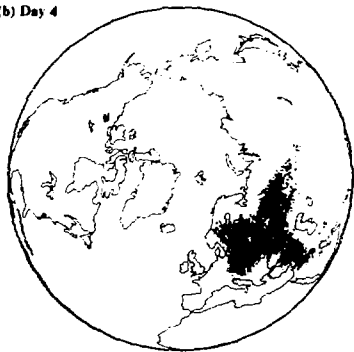


Fig. 1. The estimated fuel burn-up rate of the Chernobyl-4 reactor core.

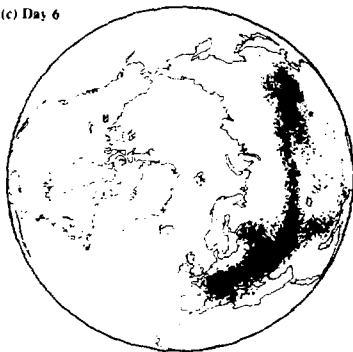
(a) Day 2



(b) Day 4



(c) Day 6



(d) Day 10

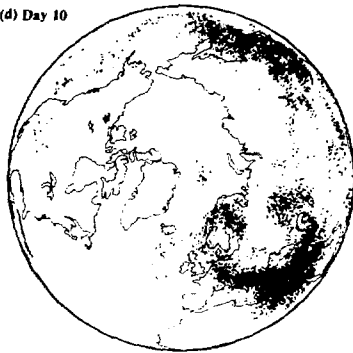


Fig. 2. The calculated spatial distributions of the radioactive cloud over the Northern Hemisphere on selected days after the explosion on April 26, 1986.



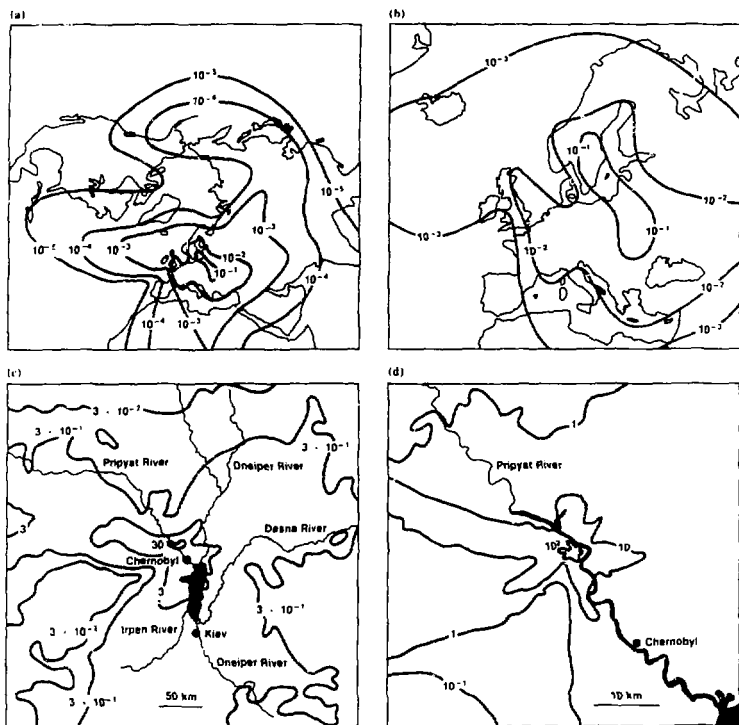


Fig. 3. The distribution of the calculated committed dose due to inhalation over (a) the Northern Hemisphere, (b) Europe, (c) a 400 x 400-km area around Chernobyl, and (d) a 50 x 50-km area around the Chernobyl area. The units are in mGy.