

A new Approach to Assess the Doses to the Population in the 30 km-Zone after the Chernobyl Accident

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INTRODUCTION

Very few data are available to assess the exposure of the population within the 30 km exclusion zone around the NPP Chernobyl. While gamma dose rate measurements were performed at regular intervals in most villages within the exclusion zone, virtually no measurements of the total activity concentration in air or the concentration of individual radionuclides relevant to the inhalation exposure were carried out due to lack of monitoring capacity. Also practically no data on activity concentration in foodstuffs are available to evaluate the exposure by ingestion of contaminated foodstuff in the various settlements. An estimate of the internal exposure of persons living in the settlements of this zone during the time of the accident and evacuated thereafter, therefore, is not possible on the basis of measured activity concentrations in air and foodstuffs for the calculations.

For some evacuees measurements of the thyroid were performed, but they cover only a limited number of individuals and therefore are representative only for certain settlements. They also comprise only ¹³¹I-intake and do not take into account other radionuclides some of which may deliver a significant contribution both to the inhalation and the ingestion dose, especially when the close distance to the Nuclear Power Plant is considered.

Therefore, to reconstruct the exposure in the various settlements for epidemiological purposes, in particular with regard to the internal exposure, a new approach had to be developed. For that purpose the fact was used that, apart from dose rate measurements in each settlement, the deposition of ¹³⁷Cs was extensively determined in the various settlements at a later stage after the accident. Also the radionuclide vector in deposition on ground is available from a number of measurements and for very few measurements also in air. Considering the fact that there was practically only dry deposition within the 30 km zone during the main fallout period, the integrated activity concentration in air of ¹³⁷Cs and the other radionuclides may be derived from ground deposition and thus the activity concentration integral in air as the basis of dose estimates reconstructed.

In the following the methodology and the problems associated with the reconstruction of some of the required parameters will be described and discussed.

METHODOLOGY

The exposure of individuals offsite the reactor site is caused by the four typical exposure pathways:

- external irradiation by the plume
- external irradiation from ground
- inhalation
- ingestion

The relative contribution of each pathway to the total exposure varies significantly according to the residence time of the individual in the contaminated zone and to the distance from the destroyed power plant. The inhalation and external dose dominate all other exposure pathways in the initial phase, while after a few days the ingestion dose contributes dominantly. In contrast to non-evacuated individuals for which the ingestion generally contributed more than 60 % of the total exposure (1,2,3), for an individual evacuated in the first days up to a about one week (depending on supply of local food) the inhalation dose supersedes both external and ingestion, after that the ingestion dose dominates the total dose. In both cases the internal exposure is the dominant contributor to the exposure of the individuals in the 30 km zone.

For the evaluation of the exposure only the following data are available:

- dose rate measurements in most settlements on a daily basis
- ¹³⁴Cs and ¹³⁷Cs deposition on ground measured at a later stage at a large number of sites
- measurements of soil samples (ratio of the various radionuclides to ¹³⁷Cs) at some sites in the zone
- very few filter measurements (particulate activity, ratio of the various radionuclides to ¹³⁷Cs in air)
- measurements of the activity ratio of the various radionuclides to ¹³⁷Cs in soil and air filters outside the 30 km zone

From these data the external exposure from the beginning of the activity release from the destroyed NPP until evacuation was evaluated for each settlement by interpolation of available dose rate data within the 30 km zone. These data were compared against dose rate curves derived from ¹³⁷Cs-deposition on ground and the

radionuclide vector as described later in this paper. From the derived dose rates in each settlement and an occupancy factor derived from average shielding factors of buildings and outdoor residence times evaluated by questionnaires, the individual effective dose of 12632 evacuees was estimated (4,5).

The evaluation of the inhalation dose requires the knowledge of the activity concentration integral of each radionuclide relevant to the inhalation dose for the whole passage of the plume. To reconstruct these activity concentration integrals, the ratio of activity of each radionuclide to the reference activity ^{137}Cs was first determined on ground. The methodology and the parameters on which these ratios depend, will be described in the next chapter. From the fact that the deposition of ^{134}Cs and ^{137}Cs on ground could be determined at a much later stage after the accident, the spatial distribution of the cesium-deposition is a well investigated parameter in the 30 km zone which is much better known than the deposition of other, in particular, short-lived radionuclides. From these ratios the activity deposition of the other radionuclides could be derived.

Since there was practically only dry deposition within the 30 km zone during the main fallout period, the integrated activity concentration in air of each radionuclide may be derived from the deposited activity on ground of this radionuclide by

$$C_i = \frac{A_i}{v_{g i}} \quad (1)$$

C_i activity concentration integral in air of i-th nuclide [$\text{Bq}\cdot\text{s}\cdot\text{m}^{-3}$]

A_i activity deposition of i-th nuclide on ground [$\text{Bq}\cdot\text{m}^{-2}$]

$v_{g i}$ deposition velocity of i-th radionuclide [$\text{m}\cdot\text{s}^{-1}$]

The deposition velocity depends strongly on the particle size, especially in the particle size range 1 - 10 μm (6). Although there are no measurements available of the size distribution of airborne particles in the 30 km zone during the time of the accident, estimates of these particle sizes were made from comparison of ground deposition to airborne activity at a few spots and from the decrease in activity ratios with increasing distance from the nuclear power plant (7). From this an AMAD of 1 μm was attributed to all volatile radionuclides (Te-, I-, Cs-, and Ru-isotopes), an AMAD of 5 μm to Sr, of 8 μm to Zr, Ba and Ce and of 16 μm to ^{239}Pu (7). The deposition velocities attributable to these particle sizes are 1 mm (volatile nuclides), 10 mm (Sr), 20 mm (Zr, Ba, Ce) and 150 mm (Pu). From this and the activity deposition of ^{137}Cs on ground as well as the ratio of the respective radionuclide to ^{137}Cs , the activity concentration integral in air was derived for each settlement and each radionuclide. Assuming the above AMAD-values for each radionuclide the inhalable fraction and thus the inhalation dose factor according to ICRP-71 (9) was determined and thus the inhalation dose for a person staying in the village for the whole time of the passage of the plume evaluated.

For most evacuees of the 30 km-zone, the residence time in the zone was shorter than the duration of the passage of the plume. To obtain the activity concentration integral in air until the time of evacuation, but also to correct for the changing ratios with radioactive decay, the fraction of the activity concentration integral responsible for inhalation at each day after arrival of the plume in the settlement was estimated from the dose rate measurements. Except for a short phase in the beginning, the external dose is dominated by the dose from ground deposition (1,10). This is particularly valid if a small fraction of noble gases is accompanied by a larger fraction of aerosol-bound radionuclides which are readily deposited. If the dose rate due to submersion is negligible compared to that from ground deposition, the change of dose rate is basically proportional to the ground deposition of activity as long as the deposition is not finished. According to formula (1), the activity concentration integral in air is proportional to the rate of change of deposited activity on ground and thus the increase in dose rate per time. By differentiating the dose rate and dividing by the maximum dose rate after deposition the daily activity concentration integral may be estimated (7).

In this way, the activity concentration integral in air for each day after intrusion of the plume in the respective village is derived and the respective inhalation dose fraction by each radionuclide estimated.

The evaluation of the ingestion dose requires the activity concentration in all foodstuffs relevant to ingestion and its time course. Since no measurements of these concentrations are available in the 30 km-zone during the days after the accident, another approach to evaluate these was adopted. The activity concentration in foodstuff may be estimated from the activity concentration integral in air, precipitation and fallout rate of the relevant radionuclides by radioecological models. The model structure is displayed in figure 1. For the main calculations the computer code ECOSYS (11) was used. This code had been validated with regard to measured activity concentrations in air and activity concentrations measured in a multitude of foodstuff samples after the Chernobyl accident in Austria and Germany and also validated in the VAMP exercise (12). Compared to inhalation dose estimates, for estimating the ingestion dose only a few radionuclides (mainly ^{131}I , ^{134}Cs , ^{137}Cs , ^{103}Ru , ^{106}Ru and ^{90}Sr) are relevant. Also the estimate is not as sensitive on aerosol particle size as with inhalation dose estimates since the influence of the deposition velocity is eliminated when first the activity concentration in air is determined from ground deposition and then the deposition on plants from that air-borne activity.

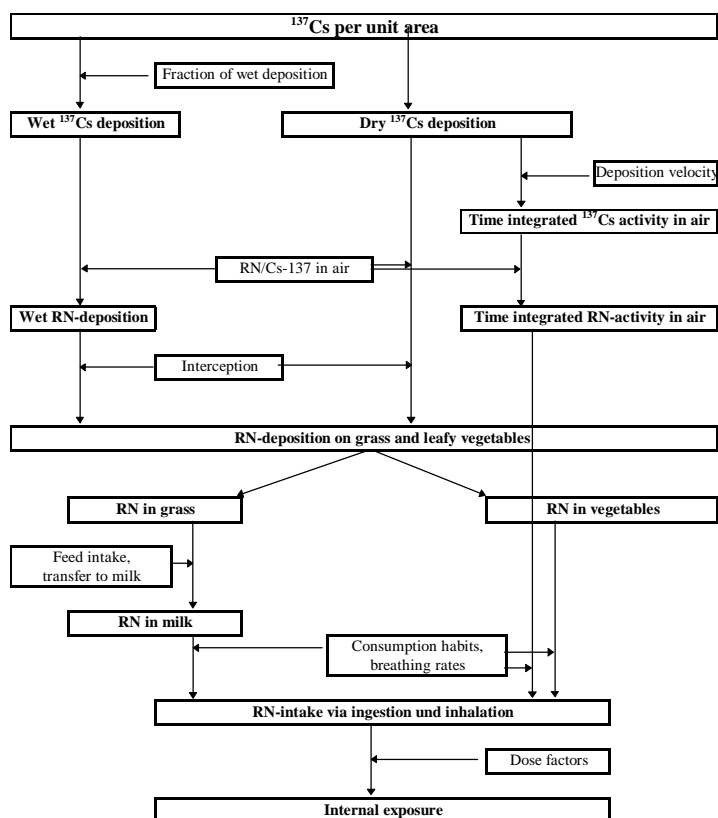


Figure 1 Model for estimating the internal exposure of evacuees

RECONSTRUCTING THE RADIONUCLIDE VECTOR

The evaluation of the ratios of the various radionuclides with regard to ^{137}Cs is separated into two groups:

- the determination of the ratio of isotopes to each other
- the ratio of different radionuclides other than isotopes

Being the same element, isotopes are characterized by the same release characteristics from the core and dispersion parameters. Radionuclides of different chemical elements may show different release characteristics to each other and to ^{137}Cs , but also different dispersion and fallout parameters. Taking into account radioactive decay, isotopes, therefore, should show constant activity ratios for the whole plume and fallout on ground and plants. In contrast, the ratio of radionuclides of different chemical properties in the release will vary according to release time (accident sequence, core temperature). They may also vary with distance according to different deposition rates associated with different particle sizes.

Therefore, on one hand, the ratio of the various isotopes to one specific "lead isotope" was evaluated from various data sets and, on the other, the ratio of these lead isotopes to ^{137}Cs was determined. It was assumed that the ratio of isotopes over the whole core is rather constant which seems credible due to the high burn-up levels of a large fraction of the core (13).

Table 1 Ratio of isotopes in the plume of the Chernobyl accident

radio-isotopes	$^{89}\text{Sr}/^{90}\text{Sr}$	$^{91}\text{Sr}/^{90}\text{Sr}$	$^{106}\text{Ru}/^{103}\text{Ru}$	$^{129\text{m}}\text{Te}/^{132}\text{Te}$	$^{133}\text{I}/^{131}\text{I}$	$^{134}\text{Cs}/^{137}\text{Cs}$	$^{136}\text{Cs}/^{137}\text{Cs}$	$^{141}\text{Ce}/^{144}\text{Ce}$
ratio of isotopes	7.7	11.8	0.216	0.066	1.6	0.55	0.23	1.70
stand.dev.	1.5	-	0.024	0.012	0.4	0.02	0.04	0.80

The ratio of isotopes was evaluated for each group from a large number of reported values of the activity in the core evaluated by various codes and calculations (7). Also from 14 different reports the ratios measured in the environment both near the NPP and at further down-wind distances were evaluated both in deposition and in air. A detailed description of these data and the methodology for the derivation of ratios is given in Pröhl et al. (7). The best estimates for the derived isotope ratio are given in table 1.

The investigation of the ratios of radionuclides of different chemical form showed that these ratios varied both with regard to distance and the direction from the NPP. This is basically caused by the different release directions at different stages of the accident and different particle sizes of the different radionuclides. However, the variation with the release direction is limited and may be characterized by a constant factor for the southern plume in comparison to the western and northern plume. This factor ranged from about 1 to 3 (14,15,16). The lower values are typical for volatile nuclides, the higher ratios for less-volatile nuclides, with the exception to $^{131}\text{I}/^{137}\text{Cs}$ which showed a ratio of 3. The differences for the western and northern plume were minor, in particular, with regard to radionuclides relevant to the inhalation dose. With regard to the purpose of dose reconstruction, therefore, the same ratios were applied for the western and the northern plume.

The ratio of the activity of the various radionuclides to that of ^{137}Cs with distance showed a more complex behavior. An evaluation of these ratios published by some 10 authors at short, medium and large distances (17, 18, 19, 20) and compared to each other in (7) showed significant variations in the ratios with increasing down-wind distances. For lack of space only some of these ratios are displayed in fig. 2 - 5, the others are given in Pröhl et al. (7).

The ratios may be described by either two exponential functions for one group of nuclides or by a linear function for the others. The decrease by two exponential functions obviously is typical for the ratio of fuel related particles such as ^{95}Zr , ^{99}Mo , ^{140}Ba and Ce-isotopes, while the linear function or even constant ratios with distance are typical for volatile radionuclides such as I, Te and Ru-isotopes. A mixture between the two appears to be typical for Sr-isotopes.

As no other process may be the cause for such a decrease in activity ratio versus distance, the only explanation for it is given by different deposition velocities for the different radionuclides. Except for iodine, different deposition velocities may be only explained by the different sizes of particles to which the various radionuclides are attached (7). According to this decrease, the radionuclides were divided into three groups of

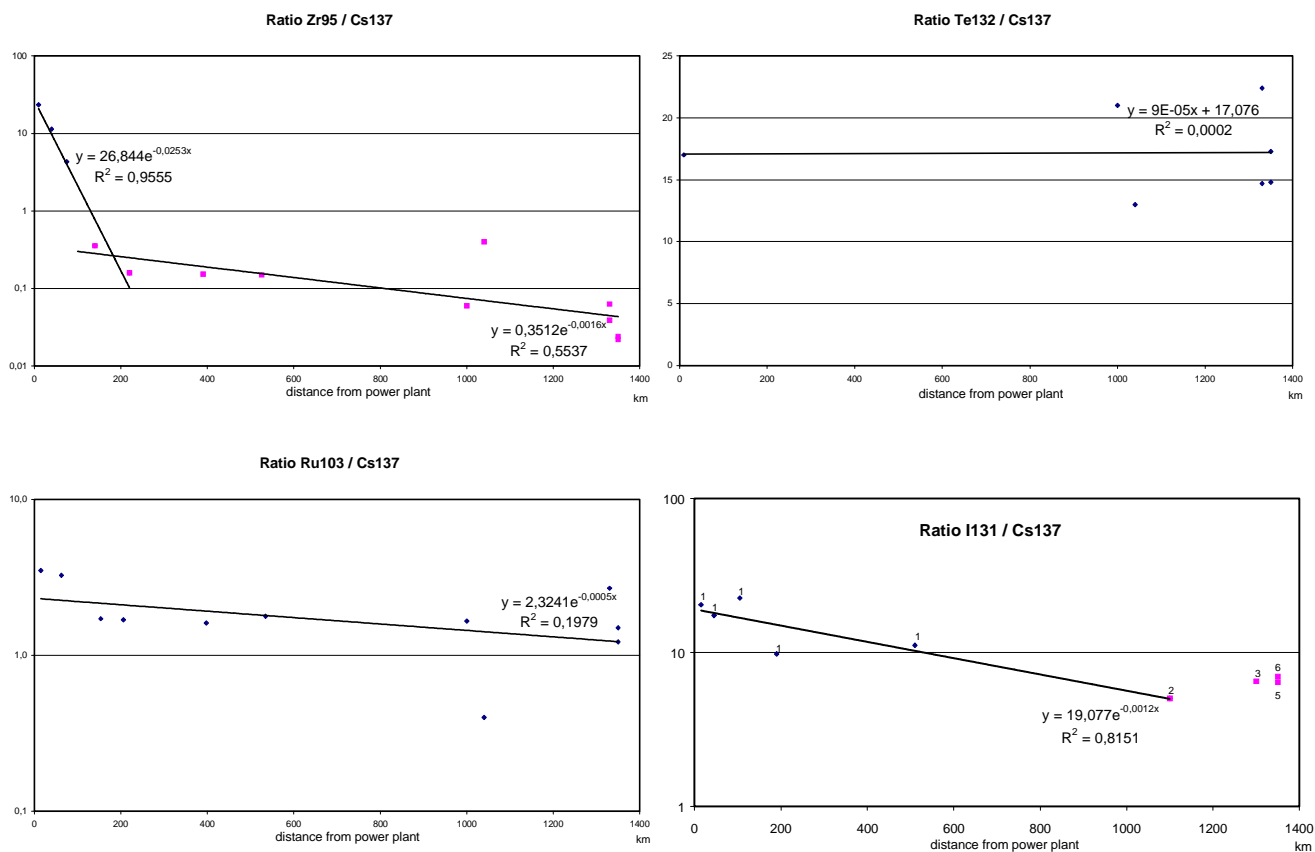


Figure 2 – 5 Ratio of the relevant radionuclides ⁹⁵Zr, ¹⁰³Ru, ¹³²Te and ¹³¹I to ¹³⁷Cs according to distance

Table 2 Ratios of guide isotope to ^{137}Cs and other relevant parameters for the derivation of the activity concentration integral in air

Radionuclide	correction factor for southern direction c_{dir}	medium particle size [μm]	deposition velocity v_d [mm s^{-1}]	ratio of lead isotope to ^{137}Cs with distance d [in km]
^{90}Sr	3.0	5	10	$0,368.e^{-0,0171 d}$
^{95}Zr	2.0	8	20	$26,84.e^{-0,0253 d}$
^{99}Mo	3.1	1	1	$3,808.e^{-0,006 d}$
^{103}Ru	3.0	1	1	$2,324.e^{-0,0005 d}$
^{132}Te	0.9	1	1	17
^{131}I	1.7	1	2^1	$19,08.e^{-0,0012 d}$
^{137}Cs	1.0	1	1	1
^{140}Ba	2.9	8	20	$27,50.e^{-0,0198 d}$
^{144}Ce	2.0	8	20	$6,199.e^{-0,0172 d}$
^{239}Np	5.6	16	150	$0,0015.e^{-0,0172 d}$
^{239}Pu	1	16	150	$0,0015.e^{-0,0172 d}$

¹ average deposition velocity for 30 % aerosol, 30 % gaseous and 40 % organic iodine

different deposition velocity:

- elements with a practically constant ratio or a decrease by less than one decade
- elements with a decrease in ratio to values of between 10 % to 2 % of near NPP ratios
- elements with a variation in ratio down to less than 2 % of near NPP ratios

The first group comprises the elements Mo, Te, I, Cs (highly volatile elements). Ru is very close to the lower boundary (9 %) and therefore may be treated as belonging to this group. An AMAD of $1 \mu\text{m}$ is attributed to these radionuclides which is the same AMAD as attributed to the Cs-isotopes.

The second group consists of Sr (less volatile), while the third group includes Zr, Ba, La, Ce and Np (low volatility). All of these radionuclides are, at least to a high degree, attached to fuel particles of larger sizes than the average aerosol particles in air.

In order to ensure that the particle size estimate and the derived deposition velocities are correct, a comparison of air-borne and ground deposition radionuclide ratios was performed (7). Although there are very few actual measurements of the activity concentration in air in the vicinity of the Chernobyl NPP, this comparison generally showed a good agreement for Zr-, Ru- and I-isotopes, but less agreement for the ratios of Te, Ce and Mo. Unfortunately, Te and Mo could only be compared in one filter measurement in the 30 km-zone, but good agreement was found in measurements at further distances (outside the 30 km-zone). In one filter measurement 20 km west of the reactor a good agreement was found for ^{141}Ce , in another filter sample a trend to underestimate the air concentration by the approach was found. Generally, for the relevant radionuclides the agreement is good proving the applicability of the method to dose reconstruction in the case of absence of activity measurements in air.

RECONSTRUCTING THE INHALATION DOSE

The inhalation dose H_{eff} of a resident of the 30 km zone is given by:

$$H_{\text{eff}} = \sum_i a_i (f + r(1-f)) \sum_j (I_j d_j) \int_0^{t_e} c_i(t) dt \quad (2)$$

H_{eff} inhalation dose [Sv]

a_i inhalation dose factor of i-th radionuclide for the appropriate age group [Sv.Bq^{-1}]

f fraction of daily outdoor residence time during passage of plume

r indoor reduction factor for aerosol activity concentration

I_j breathing rate associated with a given behavior [$\text{m}^3 \text{h}^{-1}$]

d_j duration of a certain behavior of residents (sleeping, resting, light or heavy work)

$c_i(t)$.. activity concentration of i th-nuclide in outdoor air [Bq.m^{-3}]

t_e end of plume or time of evacuation of resident

With the radionuclide ratio as given in table 2, the isotope ratio as listed in table 1 and the correction factor and deposition velocity from table 2 the evaluation of the activity concentration integral in air and the inhalation dose may be performed for each settlement.

From the fact that dwellings in the 30 km zone typically do not have tight windows or doors and that many residents stayed long periods outdoors during the period after the Chernobyl accident as a consequence of the prevalent warm weather and no information by the authorities, the indoor reduction factor r was assumed to be 1 (no reduction of aerosol activity intake due to staying indoors). A slight modification with regard to the breathing rate as compared to ICRP-71 (9) was introduced to take into account the higher fraction of time spent on heavy agricultural work by the residents in the villages around the Chernobyl NPP than the average standard man and a somewhat lower sleeping period of 8 h per day was assumed for the 12 - 17 year old child. To simplify for both men and women the same breathing rate (average of the two values) was assumed.

The results of the model calculations are given in figure 6 for the village which is situated at a distance of 4.5 km from the NPP site and was evacuated on the 3rd day after the accident. In the calculation the activity concentration integral in air was only extended until the third day. The main contribution to the inhalation dose comes from ^{131}I as observed in all other areas outside the 30 km-zone. For adults, ^{131}I contributes about 44 %, for 1-year infants about 55 % of the total inhalation dose. The second most important contribution comes from ^{132}Te (~ 19 % for adults or 18 % for the infant) while the Ru-isotopes contribute about 10 % (adults) or 8 % (infants), respectively. The contribution of those radionuclides where the activity concentration in air and thus the inhalation dose strongly depends on the correct modeling of the particle size, is low ($^{95}\text{Zr}+^{95}\text{Nb}$: 4 %, Ce: < 2 %).

However, assuming a smaller particle size diameter of 5 μm for ^{95}Zr , ^{95}Nb and the Ce-isotopes, the inhalation dose contribution of these radionuclides would amount to more than 10 % increasing the inhalation dose by about 6 %. For the very unrealistic case of an AMAD of 2 μm for these nuclides, their contribution to the inhalation would be about 73 % resulting in a substantial increase in the inhalation dose by about a factor of 2.8. The correct particle size evaluation, therefore, is very important for a correct estimate of the inhalation dose.

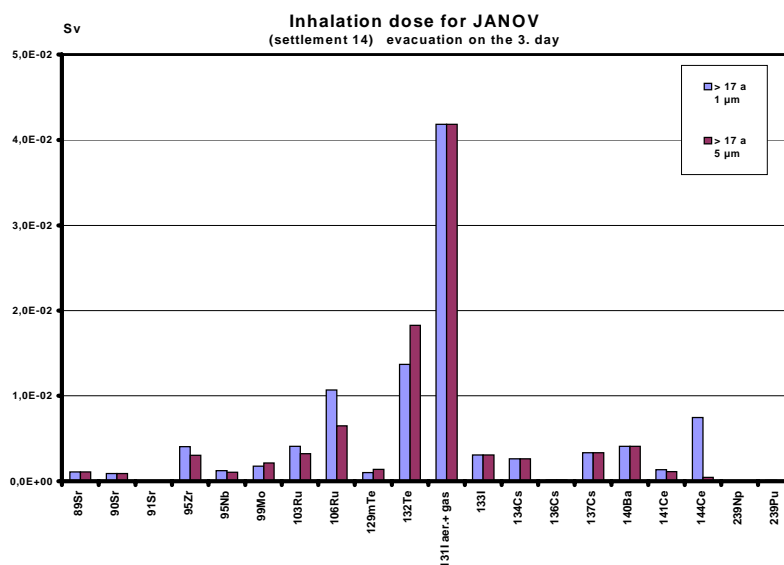


Figure 6 Contribution of the radionuclides to the inhalation dose in Janov

RECONSTRUCTING THE INGESTION DOSE

To reconstruct the ingestion dose, the model as described in figure 1 was used. For the calculations basically the prediction model ECOSYS (11) was used which had been validated with a large number of activity concentration measurements in foodstuff and fodder after the Chernobyl fallout before (12,21). In this case the deposition of ^{137}Cs and - derived via the radionuclide vector - the deposition of the other relevant radionuclides on ground were the input parameters for the calculations. From that the interception by plants were estimated based on the approach described by Pröhl and Hoffman (22). For the calculation of the transfer into various foodstuffs parameters as described in Müller and Pröhl (11) are used. For site specific intake by cows a feeding by only grass at 40 kg d^{-1} was assumed (7,23).

The calculations were performed for milk and leafy vegetables only since other foodstuffs could not contribute significantly to the ingestion dose of evacuees at that time of the year. Food consumption rates were assumed to be 0.6 L d^{-1} for milk and 0.010 kg d^{-1} for leafy vegetables for adults. Ingestion dose factors were taken from ICRP (24).

The derived effective dose in 5 selected settlements is given in table 3. Values for the total effective

dose range from 38 to 350 mSv according to distance and evacuation day. For settlements close to the site and early

Table 3 Dose in selected settlements of the 30 km-zone and the relative contribution of the various pathways

Settlement	distance	evacuation day	dose of adult [mSv]				
			total dose	ratio total / ext.	external dose	inhalation dose	ingestion dose
Janov	4.5	3	130	13.3	9.5 (7.5 %)	92 (73 %)	25 (19.5 %)
Nova Krasnitsa	18.2	8	352	15.3	23 (6.5 %)	150 (42.6 %)	180 (50.9 %)
Lelev	9.8	8	215	9.3	23 (10.7 %)	8.7 (4.0 %)	180 (85.3 %)
Chistogalovka	7.2	7	280	4.0	70 (25.0 %)	120 (42.5 %)	91 (32.5 %)
Chernobyl	15.2	7	38	6.3	6 (15.8 %)	20 (52.9 %)	12 (31.3 %)

evacuation the inhalation dose contributes mostly to the dose (40 - 70 %). For settlements at greater distances the ingestion contributes 50 - 80 % of the total dose.

Table 4 shows the contribution of the various radioisotope groups to the total effective dose. As expected, the main contribution comes from the iodine isotopes (50 - 70 %). The second most important is caused by the Ru- and Te-isotopes (5 - 12 %, and 7 - 14 %, respectively). All other radionuclides contribute only insignificantly (< 7 %) to the total effective dose. This is also true for the Cesium-isotopes. This demonstrates the significance of reducing releases of iodine in case of severe accidents, in particular, for the population at highest exposure risk close to the site.

The thyroid doses resulting from ^{131}I and ^{133}I would be roughly 14 times the values given for inhalation plus ingestion, i.e. for Janov the thyroid dose for adults is 1.44 Sv on average, for Nova Krasnitsa about 4.64 Sv. For 1-year infants the thyroid doses in these villages amounted to 5.68 Sv and 28.7 Sv, respectively.

Table 4 Relative contribution of the various isotope groups to the total internal effective doses in two selected settlements

Isotope group	Contribution to effective dose (%)	
	Janov	Lubianka
$^{89/90/91}\text{Sr}$	0,8	1,6
^{99}Mo	1,0	0,5
$^{95}\text{Zr}/^{95}\text{Nb}$	3,7	1,9
$^{103/106}\text{Ru}$	12	5,2
$^{131/133}\text{I}$	53	71
$^{134/136/137}\text{Cs}$	6,0	6,6
$^{129\text{m}/132}\text{Te}$	14	7,5
$^{141/144}\text{Ce}$	6,4	3,1
$^{140}\text{Ba}/^{140}\text{La}$	3,0	2,4
^{239}Np	0,0	0,0
^{239}Pu	0,1	0,0

CONCLUSIONS

A new approach to estimate the dose to the evacuees of the 30 km-zone around the Chernobyl NPP was developed. It comprised the evaluation of the ratios of the various radionuclides to ^{137}Cs as guide radionuclide. These ratios were determined in a two-level approach in which in a first step an evaluation of isotopic ratios for each isotope group was performed and in a second step the ratios of one guide isotope of each element group

relative to ^{137}Cs was determined in dependence on distance and direction of the plume. From the fact that practically only dry deposition occurred in the first days within the 30 km-zone, the activity concentration integral in air was derived from ^{137}Cs ground deposition values via the deposition velocity.

Preliminary results indicate that the effective dose due to inhalation amounted to 8 - 13 times the external exposure and the exposure caused by ingestion to about 2 - 2.5 times that of the external exposure in a village close to the reactor site and evacuated early or exposed early in the passage of the plume. Under those circumstances, the total effective dose is more than 10 - 15 times greater than the external exposure. In villages at further distances from the site or predominantly exposed by a longer passage of the plume and evacuated at later stages of the accident the contributions to the total effective dose were approximately: 15 - 25 % from external exposure, 30 - 50 % by inhalation and 30 - 40 % by ingestion for the adult, but 5 - 8 % from external exposure, 15 - 45 % by inhalation and 50 - 80 % by ingestion for the 1 y-infant.

As an example, for Janov at a distance to the reactor of 4 km and evacuation on day 3, the estimated total effective dose is 130 mSv, of which 92 mSv are due to inhalation. Of this dose 40 mSv are caused by ^{131}I and ^{133}I equivalent to a thyroid dose of approx. 800 mSv by inhalation. Together with ingestion this gives about 1.44 Sv organ dose for the thyroid. For Chernobyl (distance to the reactor 15 km and evacuation on day 7) the total effective dose to adults is assessed at 38 mSv. About 45 % of this dose is due to iodine from inhalation and ingestion resulting in a thyroid dose of approx. 340 mSv.

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