Performance of the Environmental Monitoring Program for Nuclear Facilities in Germany: Possibilities of Unrecognized Exposures

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1. Introduction

During the last years several cases of leukaemia of children occurred in the nearby of the nuclear power plant of Krümmel in northern Germany, which is a Boiling Water Reactor of KWU construction line 69. No reasons for these could be identified definitely by expert groups of both countries Lower Saxony and Schleswig-Holstein at whose border Krümmel is located, but it was assumed, that only radioactivity might be able of causing these kinds of massively increased rates of leukaemia. [5] This was confirmed by an incidence study [2] and by investigations on chromosome aberrations of parents and brothers and sisters of the affected children, [4] although other analysis on chromosome aberrations of adults and children in northern Germany did not lead to conclusive answers [8].

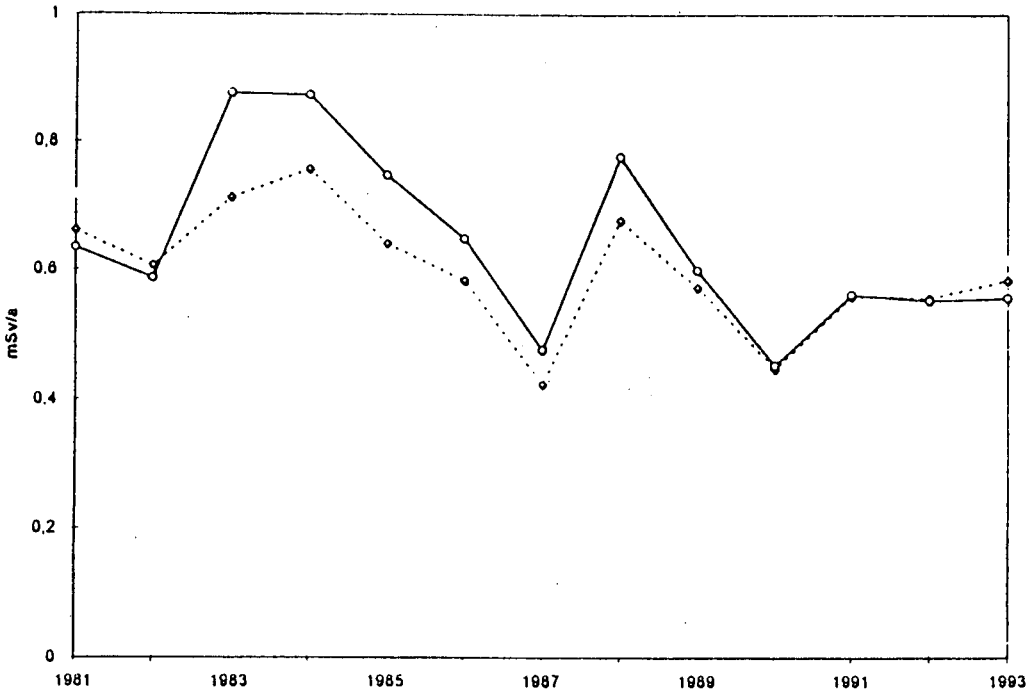
A quite extensive study on emission control of the nuclear power plant of Krümmel for the period of 1983 to 1993 came to the conclusion, that this plant had not emitted amounts of radioactivity, sufficient to lead to the observed leukaemia [3]. On the other hand, artificial nuclides had been found in the environment that could not be explained by atomic bomb fallout or the Chernobyl accident, as there were corrosion products, Cs-137 without or with an ill ratio to Cs-134, Sr-90 and hints on Ce-141 [5]. In this view a theory was introduced that tried to explain these foundings in the way that the nuclear power plant of Krümmel should emit mainly noble gases through some unknown paths, whose daughter products then could be found in the environment [5].

Further on it was suspected that Sr-89 might play a role, since it is a daughter of the noble gas Kr-89, indeed is able to damage the red bone-marrow, and since it does not emit γ-rays and due to its short half life of 50 days might stay undetected by the environment monitoring.

In the present analysis an attempt is made to determine the amounts of radioactivity that might be emitted with the exhaust air by the nuclear power plant leading to evidences in the environment monitoring no other than have been observed actually. The analysis focused on emission of noble gases and their impact on the remote monitoring system (KFÜ) which is a net of online γ-sensitive measuring systems in the surrounding of the power plant. Possible impacts on health of the people are γ-radiation due to noble gas decay and inhalation and ingestion of emitted aerosols and daughter products of the noble gases.

In a first step the dispersion parameters were calculated for dry and wet fallout as well as the γ-radiation according to the General Principle of Calculation for the Radiation Exposure (AVV) [1]. Second the data of the environmental monitoring are used to define the bounding conditions of possible emission scenarios and special attention was payed to the vector of the nuclide mixture. Third, the resulting radiation exposure is determined and finally the results are discussed in view of some conspicuous results of the environment monitoring.

Fig.2: Mean annual gamma dose measured by TLD dosimeters in 2 radial zones around the nuclear power plant Krümmel
drawn line: 2.5-5 km
broken line: 5-15 km



2. Dispersion parameters

2.1. Fallout

The dispersion of the radioactive cloud in case of no rain in the German AVV is performed according to the model of Gauß. Necessary coefficients that are deemed to be representative for German locations of nuclear power were determined by experiments [7]. The short term fallout factor is dependent on diffusion class (A-F), emission height and wind velocity. In cooperation with the breathing rate and the dose factor for inhalation it determines the inhalation dose and multiplied by the sinking velocity the fall out soil contamination is calculated. In fig. 1 and 2 isolines of the short term dispersion coefficient are shown for a windvelocity of 1 m/s, and for emission heights of 150 m and 50 m. Both figures are for diffusion class D, which is the predominant one at the site of Krümmel.
It can be seen, that the extension of the cloud transverse to the wind direction is rather small. Since aerosols in the air are only measured at four locations in the surrounding of the nuclear power plant of Krümmel it cannot necessarily be expected, that all short term emissions can be registrated by aerosol measurements.

2.2. Rainout

Modelling of rainout in the German AVV follows the idea that the washed out activity in every volume is proportional to the activity in that volume and to the washout coefficient. The concentration of the activity is calculated from the fallout coefficient. The washout coefficient is calculated by a potence function with exponent κ, base rain intensity and a linear washout coefficient that is proportional to the rain intensity. The necessary two factors were determined by experiments in a manner assumed to be representative for German plant locations.

According to the short term fallout factor a short term washout factor is defined, which is multiplied by the emitted activity to yield the resulting washout soil contamination.
In fig. 3 and 4, isolines of the short term washoutfactor are shown for emission heights of 50 m and 150 m, both for a windvelocity of 1 m/s, a rain intensity of 1 mm/h and diffusion class D.
Clearly to be seen is that the band of impact is even narrower than it is for fallout. Rain measurements are also performed only at four locations in the surrounding of Krümmel, so that it cannot be expected that every emission can be detected by this means.

2.3. γ-Submersion

The dose due to γ-rays of the radioactive cloud is determined by integrating the dose-rate that is induced by every γ-energy of all nuclides over space and time. Within the AVV the effort is reduced in the way that for all nuclides dose rate factors are given, summing the effect of all its γ-energies. According to the dispersion factors for fallout and rainout a γ-Submersion factor can be defined, which is multiplied by the released activity and the
nuclide specific dose rate factor to yield the dose of external γ-Submersion.
For remote monitoring of emissions of nuclear power plants γ-Submersion is of vital importance. Because γ-rays are far reaching and because a high sensitivity of the detectors is achievable, the net of measurements has not to be as closemeshed as for aerosol measurements.
In fig 5 and 6, again for emission heights of 50 m and 150 m respectively, γ-Submersion factors are shown for diffusion class D and 1 m/s windvelocity. Because of the high calculation effort, only distances of up to 4000 m in the direction of the wind and 2000 m transverse to it are shown. Additionally the locations of the ten nearest stations of the remote monitoring system are marked by dark points, as is the situation

when the wind is blowing in the direction of the cases of leukaemia. Stations 1 to 4 are located on the site of the plant, station 3 is nearby the chimney.

In case of unfavorable direction of wind, the stations outside the site of the nuclear power plant Krümmel will show only a small fraction of what is registered at the stations on site. Unfortunately this is just the direction of the leukaemia cases, although opposite to the main direction of wind.

2.4. Discussion

Following [3] and [10] the environment monitoring program of the nuclear power plant Krümmel consists of the following measurements as are relevant for airborne emissions:

- Air and rain measurements are performed at four locations. Besides looking for γ-activity, the air is analysed for beta activity and I-131 every 14 days and for Sr-90 every 3 months, rain is only analysed for γ-activity.
- Soil is sampled at the same locations where air and rain samples are taken, and at two further locations 2 times per year. All samples are analysed for γ-activity and Sr-90.
- Plant stuff is taken at 6 places two times a year. The analysis is like for soil.
- The γ-dose rate is continuously measured at 35 stations of the remote monitoring system, but only ten of them are within a radius of 3 km. The distribution of the stations is inhomogeneous with more weight to Schleswig-Holstein.
- The yearly integrated γ-dose is determined at 80 locations in the surrounding of the nuclear power plant, of which 19 are on site.
- Milk is analysed for every 3 months at two locations for γ-activity and Sr-90, and during summer these samples and 3 further ones are analysed for I-131 additionally.

With respect to the leukaemia cases in the south west of the nuclear power plant of Krümmel, it is concluded, that because of the short half life of some nuclides like Sr-89 and because of the small number of measuring points for air, rain, soil, plants and milk, possible emissions are to be detected mainly by the online γ-measuring systems with special emphasis on those that are on the site of the plant.

3. Emission Scenarios

3.1. Boundary conditions

The emission scenarios that were to be developed here should not be in contradiction to actual foundings of the environment monitoring programs. As was discussed in the previous chapter, the limiting factor is given by the continuous γ-monitoring on site.

There are 4 stations on site, 3 of them are at distances between 180 m and 240 m from the chimney roughly at the north, east and west side. One is 30 m away from the chimney in the south. Affecting dispersion in the direction of where the leukaemia cases are living, this one has to be focused on. The usual dose rate that is registered there during plant operation is between 0.2 and 0.3 μSv/h. Out of operation this value is reduced to 0.1 to 0.15 μSv/h. The reason for this is said by the licensing authority to be radiation from the turbine, but because these measurements do not correspond with other measurements it was also taken as proof for emissions of the nuclear power plant [5]. The calculations presented here are based on the assumption, that including calibration and registration uncertainties a dose rate of 0.05 μSv/h might not be classified as unusual.

3.2. Nuclide vector

The calculated health consequences of assumed emissions are very sensitive to the composition of the nuclide mixture. Taking the γ-dose rate on site as boundary condition for calculating maximum possible emissions, the ratio of emitted aerosols to noble gases is the dominant parameter. Sr-89 for example is the daughter of Kr-89, and in contrast to its mother does not emit any relevant γ-ray. So considering these two isotopes only, an arbitrary amount of Sr-89 could be released in case of aerosol emission, but only a limited amount is possible in case of noble gas emission.

During the licensing procedure of the nuclear power plant Krümmel the TÜV had investigated several accident conditions [9]. The nuclide mixtures that are developed there are taken as a data base for the present calculations.

Table 1 for some scenarios of Ref. 0 shows the characteristic data of the ratio of noble gas to aerosol emission and the ratio of the long living Kr-85 to the short living Kr-90 which is a measure for the overall delay time between production and release.

The nuclide mixture of the accident 'break in line of the gas processing system' has a domination of noble gas emission and during further calculations came out to yield the most serious results with respect to noble gas induced Sr-89 contamination when an additional delay of 1 minute was introduced. Thus the presentation of further results is restricted to this case. Complete results can be found in Ref. [6].

For the resulting nuclide mixture a mean dose rate factor for γ-radiation was calculated. The high energy γ-radiation of N-16 which still played a significant role in the original dataset of Ref [9] became of no importance by the additional delay time of 1 minute. The iodine isotopes are not significant in these scenarios, as are the aerosols.

Thus, only the noble gases Xe-139, Xe-138, Xe-137, Kr-90 and Kr-89 have to be considered. All others have very short half lives, so that they are present in a minor manner, or such long half lives, that they do not decay within the dispersion way of 3000 m.

Xe-139 decays whithin a few hours to the stable La-139 with intermediate nuclides Cs-139 ($T_{1/2}$=9.3 m) and Ba-139 ($T_{1/2}$=82.7 m). Xe-138 decays over Cs-138 ($T_{1/2}$=2.9 m) and Ba-138 ($T_{1/2}$=32.2 m) to La-138 which is also stable. Neither of the intermediate nuclides contributes to the dose of the read bone-marrow.

Finally the noble gases Xe-137, Kr-90 and Kr-89 are left over which decay to Cs-137, Sr-90 and Sr-89 respectively. Because of the long half life of Sr-90 and Cs-137 only small amounts of activity of these nuclides will be produced. The most relevant nuclide for inducing leukaemia within these scenarios is Sr-89.

4. Calculated Results

4.1. KFÜ-Undetected Emission

To stay within the limit of 0.05 µSv/h at the remote monitoring station nearest to the chimney, a release rate of $5.8 \cdot 10^{11}$ Bq/h of the above specified nuclide mixture is possible. Part of this are 20 % of Kr-89 7.5 % of Kr-90 and 26 % of Cs-137.

This comes up to be equivalent to release rates of $5.1 \cdot 10^{06}$ Bq/h of Sr-89, $1.6 \cdot 10^{03}$ Bq/h of Sr 90 and $3.6 \cdot 10^{04}$ Bq/h of Cs-137, and under unfavorable conditions might lead to dose rates of 0.04 µSv/h for children and to 0.014 µSv/h for adults by ingestion. Dose rate is meant in the sense that every hour of release will lead to this dose. Inhalation dose is roughly one order of magnitude lower.

Integrating this dose to a time span of 1 year, or since we do not perform long term analysis we say better 8760 hours of re-

lease, the result ist 0.35 mSv and 0.13 mSv for children and adults respectively.
The resulting soil contamination is calculated to 0.05 Bq/kg Sr-90 and 1.1 Bq/kg Cs-137 assuming that only the first 5 centimeters of soil are affected. Values for Sr-89 cannot be given without detailed data on emissions because of the short half life of this nuclide.

4.2. Unusual results of the environment monitoring

In Ref [10] unusual results of the environment monitoring are compiled and cross checked wether there is some correlation between different values. One of the most suspicious findings were rather high amounts of Cs-137 in the rain at one monitoring station in the northwest of the power plant whith the peculiarity that with the exception of small amounts of Cs-134 no other nuclides were found. In contradiction to the contention of the plant operation company that if Krümmel had emitted anything, other isotopes would have to be found there also, the result of the present scenario calculations is, that such a nuclide vector can indeed be emitted by this nuclear reactor.
In Table 2 results of the attempt are shown to recalculate the emissions that would have to be assumed within the above discussed scenario if the nuclear power plant is taken as the source.
The amount of Cs-137 that has to be released by some emitter to produce such values in the rain if other sources e.g. resuspension from the Chernobyl impact is excluded is independent of these scenarios and is only due to the formalism of the German AVV. The values for Sr-89 which are a factor of 140 higher than for Cs-137 are produced by the nuclide vector of the above models, and is hypothetically.

4.3. Discussion

The presented scenario for emission of noble gases meets the requirement of not being in contradiction to results of the environment monitoring with respect to amount and composition of the nuclide mixture as far as most noble gas daughter products, aerosols, and iodines are concerned.
It has to be mentioned that other findings of the environment monitoring, as there are Cer-isotopes and some corrosion products cannot be explained by this model.
The leukaemia cases could only be explained within the presented scenarios if the γ monitoring system located on site of the nuclear power plant of Krümmel fails. Nuclide mixtures that might be able to do without this are not covered by classical release models.
The findings of Cs-137 with untypical ratio to Cs-134 and no other nuclides fits very well in the presented scenarios assuming that the radioactivity comes from the nuclear reactor, but again, γ monitoring must have been failed.
Independently of the presented scenarios the finding of such amounts of Cs-137 in rain water if it was emitted by a plant would mean that radioactivity was released orders of magnitude higher than claimed.

5. Summary

An attempt has been made to explain the occurrence of leukaemia in the nearby of the nuclear power plant of Krümmel in Germany by scenarios based on emission of mainly noble gases.
Without a basic failure of the γ-ray monitoring system this seems not to be possible within the classical ideas of release of radioactivity.
Nevertheless there are results of the environmental monitoring that need clarification very urgently, as there are high amounts of Cs-137 in the rain near the

nuclear power plant, corrosion products and daughters of very short living noble gases in soil samples.

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Fig 1: Isolines of the fallout disperison coefficient for a height of 50 m

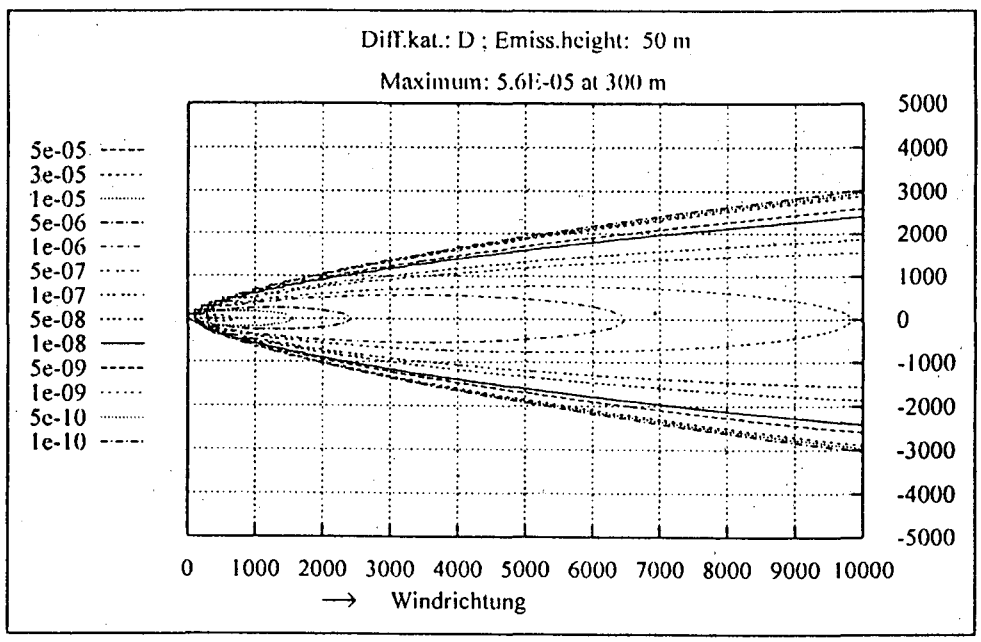

Fig. 2: Isolines of the fallout disperison coefficient for a height of 150 m

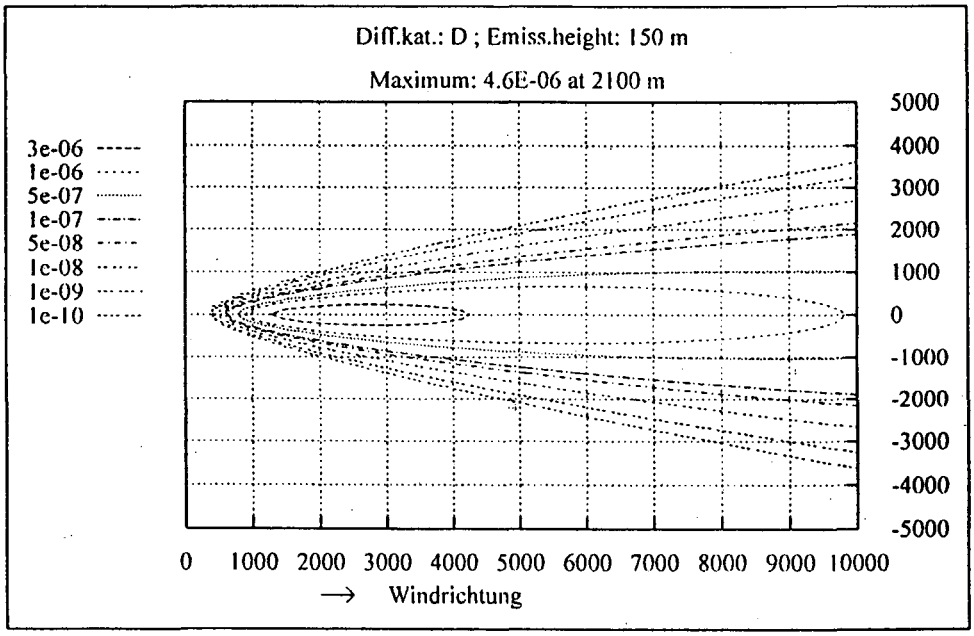

Fig 3: Isoline of the short term washout coefficient for a height of 50 m

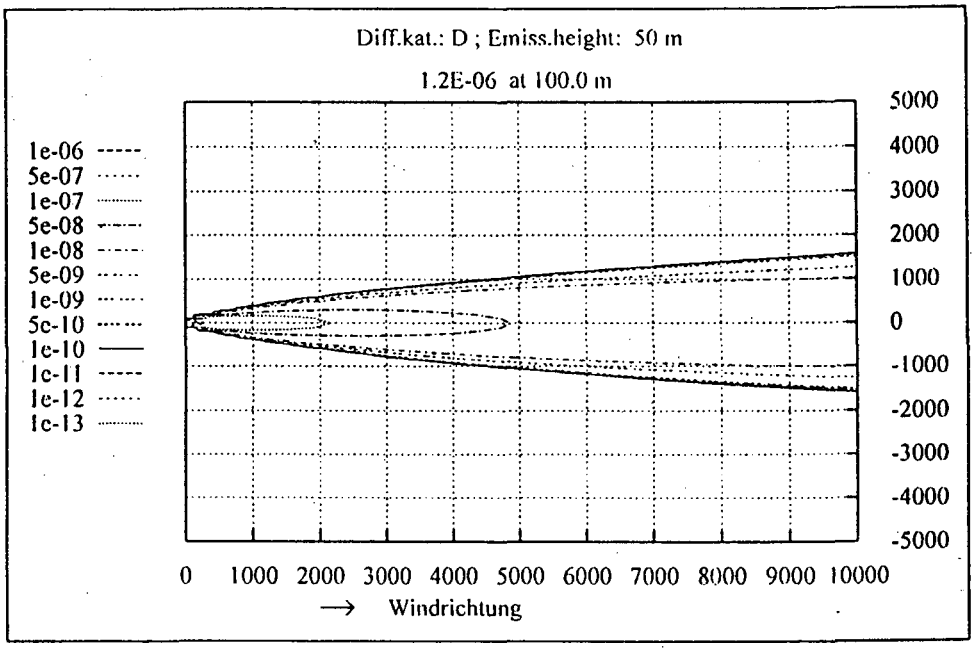

Fig 4: Isoline of the short term washout coefficient for a height of 150 m

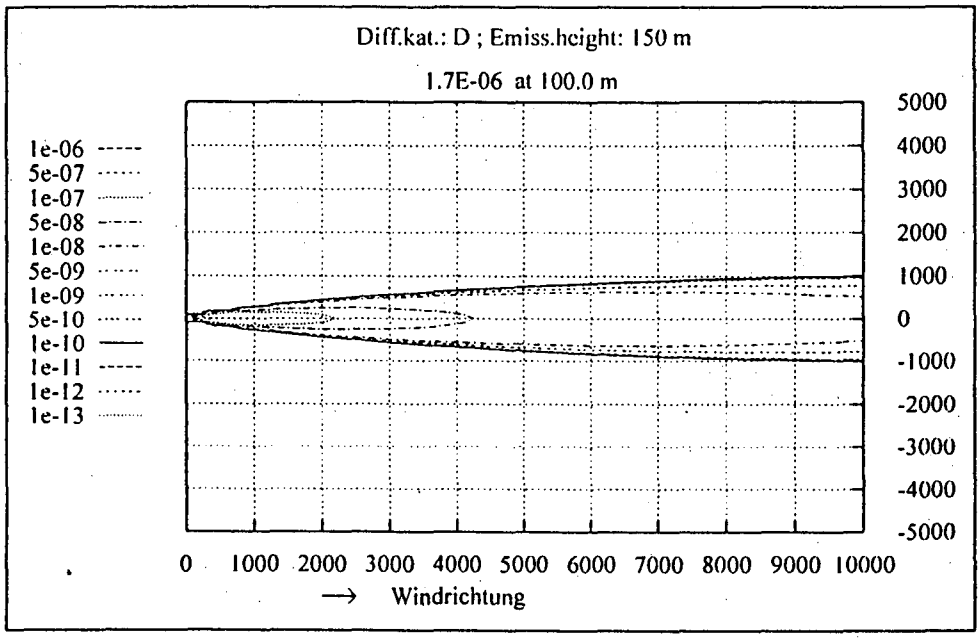

Fig 5: Isolines of the γ-submersion factor for a height of 50 m

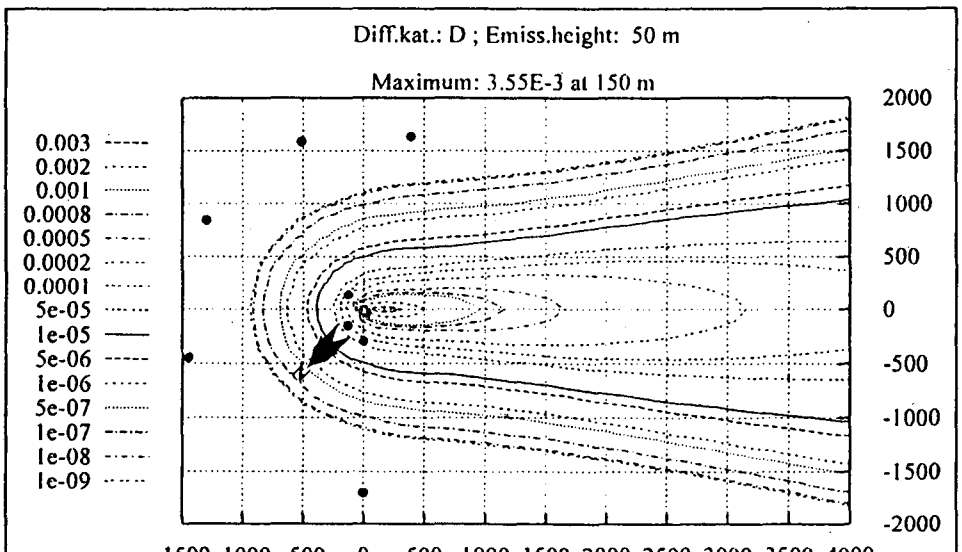

Fig 6: Isolines of the γ-submersion factor for a height of 150 m

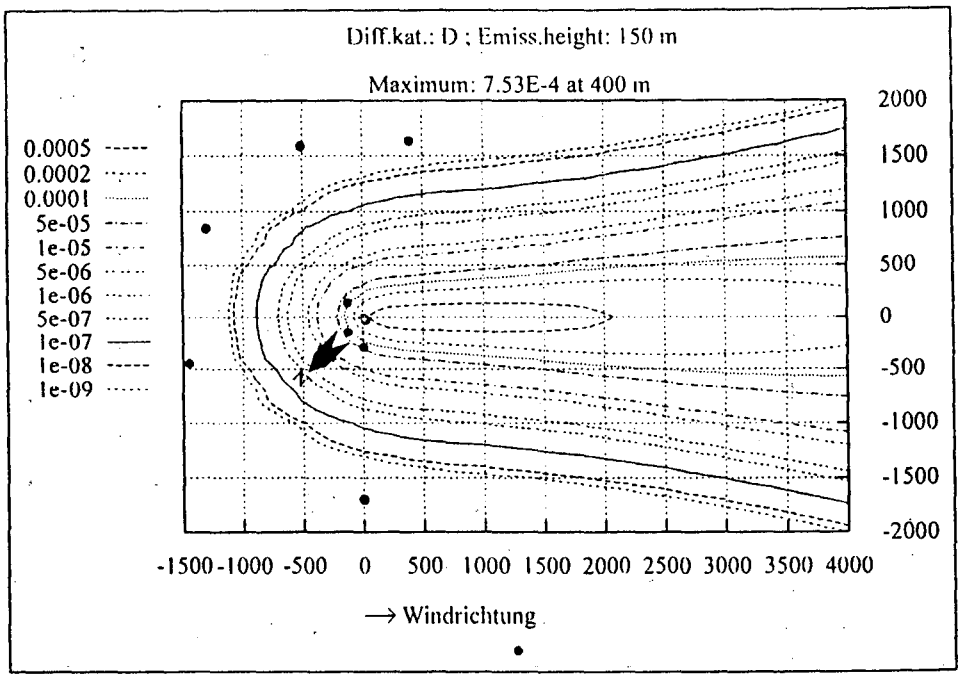

Table 1:Characteristic data of some emission scenarios [9]

	Xe-137/Cs-137 (Cs-137-equiv)	Kr-89/Sr-89 (Sr-89-equiv)	Kr-85/Kr-90 ($\cdot 10^{-06}$)
break steam line	0.04	14.3	5.6
leak steam line	0.03	13.1	5.7
leak feedwater line	0.09	14.06	627
break dec heat rem sys	$9 \cdot 10^{-06}$	0.12	17
leak dry well	0.16	178.2	14
break gas process sys	315.5	121868	8.0

Table 2:Calculations on Cs-137 measurements in rain

			3/89	89	87
	Cs-137	Bq/l	0.035		
		Bq/m^2	4.2	12.4	83.6
	rain	mm	120	276	350
period short term	Cs-137	Bq	$2.1 \cdot 10^{+07}$	$6.2 \cdot 10^{+07}$	$4.2 \cdot 10^{+08}$
	Sr-89	Bq	$2.9 \cdot 10^{+09}$	$8.7 \cdot 10^{+09}$	$5.8 \cdot 10^{+10}$
period of rain	Cs-137	Bq/h	$1.7 \cdot 10^{+05}$	$2.3 \cdot 10^{+05}$	$1.2 \cdot 10^{+06}$
	Sr-89	Bq/h	$2.5 \cdot 10^{+07}$	$3.1 \cdot 10^{+07}$	$1.7 \cdot 10^{+08}$
period long term	Cs-137	Bq	$3.8 \cdot 10^{+08}$	$2.0 \cdot 10^{+09}$	$1.1 \cdot 10^{+10}$
	Sr-89	Bq	$5.3 \cdot 10^{+10}$	$2.8 \cdot 10^{+11}$	$1.5 \cdot 10^{+12}$
	Xe-137	Bq	$1.6 \cdot 10^{+15}$	$8.2 \cdot 10^{+15}$	$4.5 \cdot 10^{+16}$
	Kr-89	Bq	$1.3 \cdot 10^{+15}$	$6.4 \cdot 10^{+15}$	$3.5 \cdot 10^{+16}$
KFÜ-on site	γ-dose rate	µSv/h	0.5	0.7	3.5
children	Ingestion dose	µSv	147	435	2900