Evaluation of Nuclear Reactor Releases by Environmental Radioactivity in a German Region of Elevated Leukaemia in Children and Adults

Michael Schmidt,Portsmouth,
Inge Schmitz-Feuerhake, Bremen,
Heiko Ziggel, Portsmouth

Introduction

The leukaemia cluster in the proximity of the 1300 MWel German boiling water reactor Krümmel (KKK) was detected by a local physician and showed an increase of 2800 % for children between 1989-1992 in the village of Elbmarsch, another case occured in 1995 [6]. An incidence study conducted for the period 1984-93 showed an increase of leukaemia also in adults [5].

Because the supervising ministry had attested undisturbed operation of the plant and no conspicuous radioactivity was found in the surroundings at that time, we started an independent investigation about contaminations in the past, since the start of operation in 1983, and of long-lived relics of short-lived nuclides.

Tritium in trees

Any release of parts of the volatile inventory of a nuclear reactor will include emissions of tritium. Once this nuclide has entered into the biosphere it will be taken up by the vegetation. In a boiling water reactor tritium is mainly produced by ternary fission, i.e. splitting of the uranium nucleus into 3 fragments.

The maximum permitted emission of tritium by KKK via airborne effluents is 7.4 10^{10} Bq y^{-1}. Using this value and the accepted methods of dispersion calculation [2] we derived, assuming a maximum long-time dispersion factor of 10^{-7} s m^{-2} that the additional tritium contribution due to the operation of KKK should be less than 1 Bq/kg in trees in the vicinity of the plant.

According to König [9] the tritium concentration in spruce and pine needles and the foliage of trees remains below the maximum measured tritium concentration in the water vapour content of the air and thus below its concentration in surface water. For this reason and in view of the continuous decline of the tritium concentration in surface water and organic matter in all parts of Germany since the 60´s, it can be excluded that the operation of KKK under the permitted conditions would result in a significant elevation of tritium in trees of Elbmarsch of more than 5 Bq/kg. The results of measurements in 1992 and 1993 are shown in table 1, they give evidence of unauthorized airborne releases by KKK.

These findings are confirmed by investigations of drinking water reservoirs located in the vicinity of KKK. Regular checks of radioactivity which are performed in the fountain of the village of Geesthacht which is situated opposite the riverside of Elbmarsch showed an elevation to 4 Bq/kg in the 3rd quarter of 1984 [8]. Because no connection to surface water exists only an airborne origin of the tritium contamination can be assumed caused by mixing of water with the surrounding air. In 1991, a tritium concentration of 3 Bq/kg was measured in a deep fountain of the waterworks of Elbmarsch, which is also not connected to surface water. A second fountain of this installation only showed a concentration below

0.3 Bq/kg, as would be expected under normal conditions [3].

Autoradiography in trees

In the years 1992 and 1993 two independent investigators exposed β-sensitive and X-ray films by slices of Elbmarsch trees and found blackenings [1,10]. The slices were plane and had been dried and polished, the films had been fixed upon them without pressure. In 1995 the measurements were repeated with the same samples and there were no blackenings anymore.

In context with later findings we interprete the formerly detected β-radiations as originated from nuclides of medium half-live as predominantly from Ce 144 (184 d) and maybe Ru 106 (368 d), see below. These nuclides are not routinely controlled in the environment.

Cs-isotopes in rain water

We have recently detected that the supposed leakages of the reactor are most clearly shown in rain water [7], which is routinely controlled for long-lived gamma emitters. The Cs 137 values of the 3 measuring stations around the nuclear plant are shown in Fig. 1. The station „Grünhof" which is situated in a distance of 2.2 km in the main wind direction of KKK shows a decreasing but permanent elevation of the Cs-isotopes 137 and 134 since 1986. These isotopes were elevated compared to the other locations of the monitoring program and the Cs 137-concentration was at least 10 times higher than at other measuring points in Northern Germany since the 3rd quarter of 1986 until 1993.

We have therefore calculated for the deposition of Cs on soil by the registered rain falls, taken the mean velocity of winds measured by the operators of the facility [7], and estimated the source term in Bq per year after an approaching formula [2]. The results are shown in Table 2. They are compared to the annual activity limit for β-aerosoles and the emissions which are named by the owners of the facility which show a difference of 3-4 orders of magnitudes. The differences are even higher if one considers the fact that Cs-isotopes will not exclusively escape from the plant but be accompagnied by other aerosols. According to the owners the emissions of Cs were below 10^5 Bq per year.

In our opinion it is undoubtly shown by these measurements that there were repeated releases of nuclides in unpermitted quantities. The owners have stated repeatedly that they have no explanation for this but deny an origin by KKK because of their emission control. There is, however, no other potential source of fission products of similar strength in that region. Therefore, if the emission control via the stack is valid there must be some kind of bypass.

The elevated emissions are confirmed by the results of dry nuclide estimations in the β-aerosols above ground. In several years during the operation of KKK there were concentrations above the detectable limit, which were mostly found in the other wind directions. In these, also remarkable concentrations of Sr 90 were measured in 1984 and 1988 which are also not explainable by other sources.

Gamma Dose in the surroundings

The monitoring programme of KKK includes measurements of the annual external gamma dose by 80 TLD dosimeters distributed in the surroundings. We analysed the results by regarding two zones: 2.5-5 km (without measurements inside the area of the facility) and 5-15 km, and calculating the mean (Fig.2). It can be shown that there was a systematic elevation of gamma exposure in the internal zone since the beginning of operation in 1983 until 1990.

The difference corresponds to a mean additional exposure of 0.09 mSv per year which is rather low compared to the expected doses for leukaemia induction [11], but does not mean that the doses have always been below the permissible limit of 0.3 mSv per year, because mean values are compared. Single measurements show also higher deviations than 0.3 mSv from the whole mean value for gamma submersion. The influence of the reactor operation is clearly demonstrated. If one divides the zones in direction sections it is seen that the elevation stems from the dosimeters in the section of Elbmarsch. The result of this analysis is remarkable because the expert report predicted a maximum dose for all exposures by the reactor of 0.04 mSv per year [14].

Cs 137 and Sr 90 in soils and green plants

While the owners of the facility and the supervising ministry had stated that there is no indication of an elevated activity release of KKK, the immission data show repeated elevations of the nuclides Cs 137 and Sr 90 which are not explainable by Chernobyl emissions. The highest contamination by Sr 90 was registered in August 1987 in grass of a location 10 km of distance to KKK, containing 30 Bq/kg (dry mass) which is more than 10fold of the normal concentration in Germany at that time. The highest concentration of Cs 137 was measured to be 103 Bq/kg in grass in a 2 km distant location in 1989 which corresponds to an about 100-fold increase against the normal value in the vegetation.

Short-lived isotopes in the environment

The environmental monitoring programme for KKK does not include nuclide specific and continuous measurements of pure ß-emitters in the controlled media, except Sr 90 in the air, as well as any measurements of short-lived gamma and ß-emitters. Nevertheless have there been occasional findings of the nuclides Ce 141, Ce 144, Ru 103, Ru 106 and Np 239 as well as radioactive corrosion products. The nuclides Ce 144, Ru 103, Ru 106 and Np 239 are only expected after damages of the fuel rods and in cases of cooling water loss [13]. They are not detectable in the reactor water and the steam in the predicted normal operation.

Location of the leakages

TLD and glass dosimeters positioned on the roof of the turbine hall of KKK, in a distance of about 60 m from the high pressure turbine, show extremely high doses all over the years up to 210 mSv per year. Furthermore are the signals of the two types of detectors significantly different. We therefore suppose an origin partly by ß-radiation which would mean an exposure outside the hall. The high doses at that great distance are not explainable by N 16 in the circulation. We therefore suppose leakages from the turbine hall which are not adaequately controlled in our opinion.

Dose considerations

We have formerly derived [11] that a rather conventional approach to the necessary dose in Elbmarsch which could explain the appearance of the leukaemias would lead to 26 mSv for the bone marrow of children.

From the observed proportion of the registered nuclides Cs 137:Cs 134:Sr 90 which shows an extreme deviation from the predicted scenarios in normal and accident operation [13,14] we concluded that the progeny of noble gases from the reactor contributes to the emissions. The main exposure of the population - proved by biological dosimetry but not to quantify by this method [4] - would therefore have been caused by bone and perhaps gonad seeking short-lived ß-aerosols like Sr 89 (50.5 d; no γ) and Y 91 (58.5 d; 30 % γ). As the low contribution of gamma submersion has to be considered we have no final evaluation of the possible doses up to now.

The leukaemia families in Elbmarsch are known to be agrarian self-suppliers, except one, and the supermarkets sell products of the region which is fruitful and produces vegetables. The observed concentration of 103 Bq/kg of Cs 137 in vegetation would lead to a bone marrow dose rate in children of 8 mSv per kg of ingested greens (dry mass), assuming a possible relation of 140:1 for the nuclides Sr 89 and Cs 137 after scenario considerations of [12] and the conventional modelling of radioactive pathways [2]. In our opinion, the leukaemia in Elbmarsch is therefore a completely explainable phenomenon.

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Table 1:Tritium in trees

Tree	Location	Annual Ring Zone	Tritium Concentration [Bq/kg]	Laboratory
Chestnut	Elbmarsch, riverside opposite KKK	ca. 1984 - 92	14.9 ± 0.3	1*
Chestnut	Elbmarsch, riverside opposite KKK	ca. 1986 - 92	33.0 ± 1.0	2**
Apple	Elbmarsch,riverside opposite KKK	ca. 1984 - 92	33.9 ± 0.3	1
Spruce	Grünhof, 1,6 km east of KKK	ca. 1984 - 92	5.5 ± 0.5	2
Chestnut	Bremen	ca. 1986 - 92	< 4.0	2

*) Institute for Radiochemistry, Technical University of Munich
**) Central Laboratory for Isotope Technology, Federal Research Centre for Nutrition, Karlsruhe

Table 2:Isotopes in rain at station N II (Grünhof) and estimated source term for KKK releases
Detection limit 5,0 Bq/m³ = 5 mBq/l
1981-1985 < Detection limit

Year	Rainfall mm	Washout Cs 134 + Cs 137 Bq/m²*)	Mean velocity of winds m/s	Estimated emission Cs 134 + Cs 137 Bq	above limit for β-Aerosols >8d factor	Release β-Aerosols >8d as declared by KKK Bq **)
1986 2. Halbj.	216	69,0	7,6	47 E+10	32	13,9 E + 06
1987	350	97,3	8,0	43 E+10	29	45,9 E + 06
1988	340	37,9	7,8	17 E+10	11	20,3 E + 06
1989	276	15,2	7,7	8 E+10	5,5	14,3 E + 06
1990	374	16,5	8,2	7 E+10	4,7	14,1 E + 06
1991	328	10,1	7,6	44 E+09	2,9	93,3 E + 06
1992	344	7,0	7,8	30 E+09	2,0	
1993	400***)	15,1	8,9	64 E+09	4,3	

*) Difference to washout at station N I or N III (lowest value)
**) Theoretical maximum (for results below the detection limit the limit value was used).
***) 1st-3rd quarter, in the 4th quarter the reactor did not run.

Fig.1: Cs 137 in rain water measured at three locations in the vicinity of the nuclear power plant Krümmel

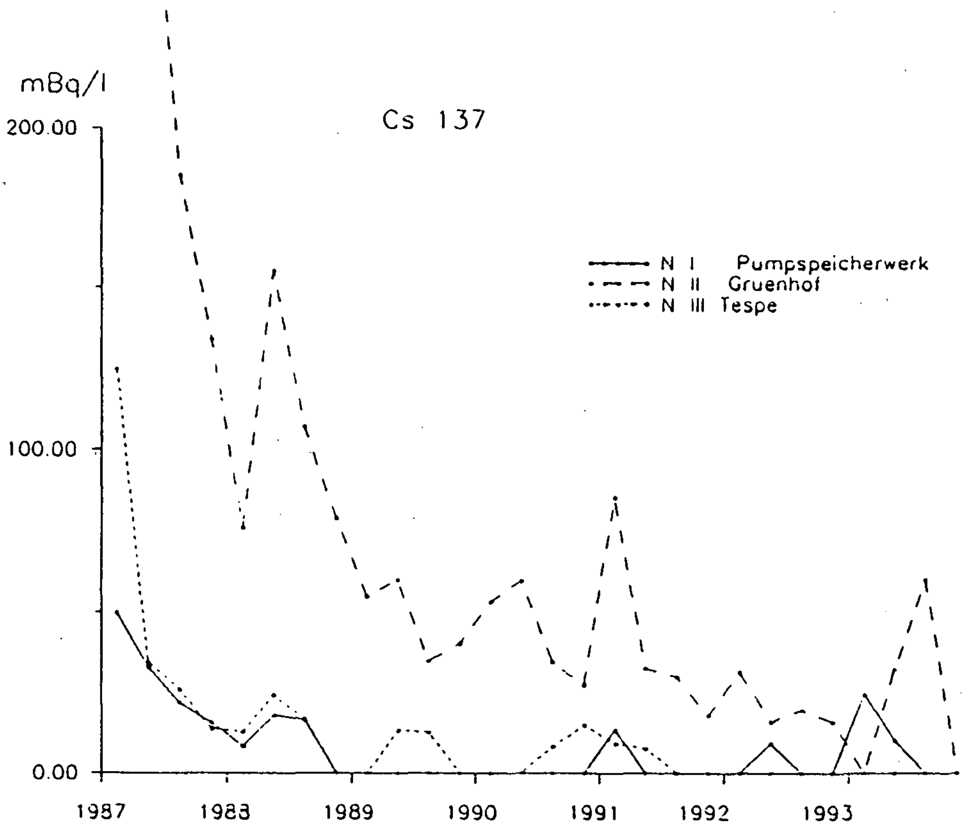

Fig.2: Mean annual gamma dose measured by TLD dosimeters in 2 radial zones around the nuclear power plant Krümmel
drawn line: 2.5-5 km
broken line: 5-15 km