Environmental Health Perspectives Supplements
Vol. 101 (Suppl. 3): 113–115 (1993)

Radium-226-Contaminated Drinking Water: Hypothesis on an Exposure Pathway in a Population with Elevated Childhood Leukemia

by Wolfgang Hoffmann,[1] A. Kranefeld,[1] and Inge Schmitz-Feuerhake[1]

A recent epidemiological survey on childhood malignant disease in the region of Ellweiler, Rheinland-Pfalz, Germany, revealed a significantly increased incidence of childhood leukemia, but observed incidences of lymphoma and solid tumors were normal. Established risk factors such as individual exposure to chemicals as well as hereditary genetic disorders were ruled out in interviews with the patients or their families. The general population in the region, however, is subjected to considerable doses of ionizing radiation due to high levels of external γ radiation and high activities of indoor radon. Radiation-specific chromosome aberrations were found in one of two healthy siblings and one father of leukemia patients as well as in any of three probands living in houses with high indoor radon activities. Radon and natural γ radiation, however, cannot explain the geographical pattern of the cases. Four out of seven cases were observed in two particular villages near a uranium processing plant. The drinking water of these villages partly came from a small river that was contaminated with radium-226 washed out from the dumps of the uranium plant. Only sparse measurements of ^{226}Ra are available, but derived red bone marrow doses for children in the two villages obtained from a simple radio-ecological model show the significance of the drinking water pathway. Prenatal ^{226}Ra exposure of fetuses due to placental transfer and accumulation may have led to significant doses and may explain the excess cases of childhood leukemia in the region even in quantitative terms.

Introduction

In the late 1980s, the population of Ellweiler in the southwestern part of Germany suspected an elevation of childhood cancer cases in their region. Ellweiler is a small village in the Federal state of Rheinland-Pfalz near its border with the Federal state Saarland.

Ellweiler first appeared in the literature after high indoor Radon levels had been measured (*1*). The radon is emanating from the subsoil, which contains high concentrations of natural uranium ore. Uranium has been mined in the region since the early 1950s. A uranium processing plant was built in 1961 about 1.5 km from the village. In the plant, uranium is extracted from the ore by physical and chemical means. The resulting uranium concentrate ("yellow cake") is used in the production of nuclear fuel. The residue is pumped outside in pipes and deposited in the open air on an area of about 140,000 m^2, forming big dumps. The remnants still carry nearly all the radium originally present in the natural ore.

The total activity of the dumps of the plant has been estimated at 55 TBq (1500 Ci), with an average of 24 Bq/g (2). A small brook named Steinaubach enters the compound coming from the north and flows between the dumps and empties into the river Nahe.

Epidemiology

We compiled all cases of childhood malignant diseases in the region for a period of 20 years, from 1970 to 1989. The investigation area was defined by a circle of 20-km radius around the uranium processing plant. For geographical analysis, the region was later subdivided in circular areas with radii of 5, 10, 15, and 20 km. A significant increase in childhood leukemia incidence was found in the inner circle of 5 km around the plant, whereas the incidences of solid tumors did not differ from the German average [(*3*) Table 1].

Excess cases were not randomly distributed in the inner zone. Four out of seven cases (57%) in the inner zone (0–5 km) were observed in two particular villages, Hoppstädten-Weiersbach and Gimbweiler. About 3000 people are living in the two villages, thus representing only 23% of the population in the inner zone.

[1]Department of Medical Physics, FB I, University of Bremen, Bremen, Germany.

Address reprint requests to W. Hoffmann, Department of Medical Physics, FB I, University of Bremen, Achterstr., D-2800 Bremen, Germany.

Table 1. Observed versus expected leukemia cases.[a]

Area, km	< 15 years			< 20 years		
	Observed	Expected	$p\ (H_o)$	Observed	Expected	$p\ (H_o)$
0–5	5	1.77	3.42%*	7	2.27	0.88%*
5–10	4	3.63	49.11%	6	4.61	31.60%
10–15	8	6.92	38.94%		(Data incomplete)	
15–20	14	15.15	65.13%		(Data incomplete)	

[a]Control values are from cancer registry data of the Federal state of Saarland.
*Statistically significant.

Exposure Pathways

Elevated external γ irradiation did not show any significant differences within the inner zone or between the four zones (*4*). Radon activities, however, showed marked geographical differences. The highest indoor radon-222 activities were measured in Ellweiler, with an indoor average of 185 Bq/m^3 and an outdoor average of 35 Bq/m^3 (G. Keller, personal communication). In Ellweiler, however, no childhood leukemia was observed in all the years of the investigation.

Radon activities in Hoppstädten-Weiersbach and Gimbweiler were found to be within the range of the German average of about 50 Bq/m^3 indoor and 14 Bq/m^3 outdoor activity (*5,6*). Hence, there was evidence for an additional exposure pathway. Radiation-specific chromosome aberrations observed in two out of three healthy relatives of leukemia patients in the two villages were in the same range as in three healthy persons from Ellweiler exposed to high indoor radon activities. A 12-year-old girl from Hoppstädten-Weiersbach showed two centric ring chromosomes in one cell (*7*). These results support another radioactive exposure pathway.

Drinking Water Pathway

Elevated leukemia rates could possibly reflect the ^{226}Ra contamination the drinking water. Up to 1987, the drinking water for Hoppstädten-Weiersbach and, to a lesser extent, for Gimbweiler (where drinking water was supplemented to some 80% by local spring water), was mainly purchased from the U.S. Army. The army runs a waterworks near the small village of Altmaiersmühle to supply several military facilities and barracks in the region. The waterworks takes raw water from the river Nahe. About 2.5 km upstream from the waterworks, the Steinaubach empties into the Nahe.

Radium-226 from the Steinaubach on its way through the dumps would consequently reach the waterworks and could appear in the local drinking water supply of the villages studied, where most of the leukemia cases were observed. Unfortunately, there are no data on ^{226}Ra activity in the raw water of the American waterworks. Monitoring the Steinaubach, however, revealed contamination with α-emitting nuclides. Total α activity in the brook was always higher downstream from the uranium processing plant than upstream from the plant (*8*). Using these measurements, a simple radio-ecological model was calculated to estimate the resulting doses to the red marrow and bone surface for the exposed people due to ingestion of ^{226}Ra-contaminated drinking water.

Table 2. Age-specific daily fluid intake in milliliters per day (*10*).

Age group, years				
0–1	2–5	6–10	11–15	15
600	100	1300	1500	1500

Table 3. Age-specific annual ingestion of ^{226}Ra in Bq per year.

Village	Age group, years					
	0–1	2–5	6–10	11–15	15	20
Hoppstädten-Weiersbach	79	131	171	197	1965	2950
Gimbweiler	16	27	36	41	405	610

Radio-ecological Model: Data and Assumptions

The contribution of the Steinaubach to the raw water of the U.S. Army waterworks is about 12% (the remaining water is from the Nahe). The highest activity was reported on September 9, 1983. The total activity in the Steinaubach downstream from the plant was 3.01 Bq/L. The conservative assumption would be that this refers to pure ^{226}Ra. This activity would then appear in the raw water of the waterworks after being diluted by 88% water from the Nahe. Furthermore, the Nahe is considered to contain the German average drinking water activity of about 4 mBq/L ^{226}Ra activity (*9*). The raw water would then contain about 360 mBq/L of ^{226}Ra. This water is used to supply Hoppstädten-Weiersbach and, after being diluted by 80% spring water, it supplies Gimbweiler. Contamination of the spring with 4 mBq/L ^{226}Ra is assumed. This would mean a contamination of some 75 mBq/L ^{226}Ra in the drinking water for Gimbweiler.

Assumptions on age-specific daily fluid intake are shown in Table 2. Table 3 shows the resulting annual ingestion of ^{226}Ra of the respective age groups. Using age-specific doses for the ingestion of ^{226}Ra (Table 4), the resulting doses to red marrow and bone surface can be derived (Tables 5, 6).

Discussion

This simple and preliminary radio-ecological model is outlined to encourage discussion of the hypothesis on an environmental exposure pathway that has not been consid-

Table 4. Age-specific doses for red marrow and bone surface for ingestion of ^{226}Ra.

	Dose-Sv/Bq × 10^{-6}	
Age, years	Red marrow	Bone surface
1	5.6	46.0
5	2.6	25.0
10	1.4	14.0
15	0.82	7.7
20	0.6	6.8

Table 5. Age-specific annual individual exposure doses and accumulated doses (red bone marrow, μSv).

	Age group, years					
Village	0–1	2–5	6–10	11–15	15	20
Hoppstädten-Weiersbach	442	342	239	162	3812	4403
Gimbweiler	92	71	50	34	794	917

Table 6. Age-specific annual individual exposure doses and accumulated doses (bone surface, μSv).

	Age group, years					
Village	0–1	2–5	6–10	11–15	15	20
Hoppstädten-Weiersbach	3627	3285	2391	1518	36312	43014
Gimbweiler	756	684	498	316	7565	8961

ered in the Ellweiler survey up to now. Many parameters are still unknown, including such important information as the actual radium-226 contamination of the waterworks in the past years. This work does show, however, that the drinking water pathway could be relevant for the population and gives an initial idea about what might have been the order of magnitude of population exposure due to chronic ingestion of ^{226}Ra-contaminated drinking water. The significance of this pathway is emphasized by the following considerations: *a*) The pathway is selectively exposing the two villages with the highest observed leukemia rates in the survey. *b*) In two healthy relatives of leukemia patients living in Hoppstädten-Weiersbach, radiation specific chromosome aberrations were observed. In one case we found two centric ring chromosomes in one cell, indicating high-LET radiationlike α rays (*7*). *c*) Radium-226 may be very effective in inducing leukemia due to its physiological similiarity to calcium. It is accumulated in the skeletal system and thus stored in close vicinity to the target cells. During prenatal life, when susceptibility to radiation is highest, the fetus accumulates radium from the mother's diet. Fetal content has been measured to be some 6.7 times higher than that of the mother's food (*11*). Additional exposure would result from postnatal breastfeeding. Radium is mobilized from the mother's skeletal system and excreted with the milk.

In 1987, a German study on radium-226 and radon-222 contamination of drinking water was published (*9*). Measurements of Hoppstädten-Weiersbach have been performed only in 1979 and 1980. Results of 13 samples for radium-226 range from 2.96 to 22.2, with an average of 14.9 mBq/L. This value is much lower than the value used in the model. The results, however, do not disprove the theory. The few measurements show the high range of the contamination, which may have differed by orders of magnitude due to operating conditions of the plant, rains, winds and other factors. On the other hand, the results do show that significant contamination is likely: the ^{226}Ra activity of the drinking water of Hoppstädten-Weiersbach was far above the German average of 4.13 mBq/L (*9*).

Fortunately, the population is now not at risk: since 1987 Hoppstädten-Weiersbach and Gimbweiler have been connected to the regional drinking water network, which does not contain any water from the Steinaubach. However, another population at risk has been neglected: the U.S. Army personnel and their families who are still supplied with drinking water from the waterworks. No assessment of the impact of ^{226}Ra ingestion on their health has been made to date. We believe this situation in the region of Ellweiler requires urgent and thorough physical and radio- ecological evaluation.

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