Paper

TRANSURANIC ISOTOPES AND ^{90}Sr IN ATTIC DUST IN THE VICINITY OF TWO NUCLEAR ESTABLISHMENTS IN NORTHERN GERMANY

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Abstract—**Attic dust was chosen as the test medium in order to search for traces of man-made bone seeking alpha and beta emitters. The samples were taken from 5 houses in the community of Elbmarsch situated at the river Elbe, adjacent to the Krümmel nuclear power plant and the nuclear research center of Geesthacht. Five houses in other regions of northern Germany were taken as a control. ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm were measured by alpha spectrometry after chemical separation. Additionally, ^{241}Pu was measured by liquid scintillation spectrometry, and the fission product ^{90}Sr was measured in a separate investigation. All nuclides except ^{244}Cm showed activities above the detection limit in the Elbmarsch samples and an elevated mean concentration compared to the control. It can be concluded from the activity ratio $^{241}Am/^{239,240}Pu$ that the Elbmarsch contamination cannot be accounted for by the background levels of transuranic nuclides resulting from weapons fallout. The derived release of alpha emitters is assumed to have contributed to the induction of a leukemia cluster in children, which was observed in Elbmarsch between 1990 and 1996.**

Health Phys. 84(5):599–607; 2003

Key words: environmental impact; ^{241}Am; inhalation; leukemia

INTRODUCTION

THE COMMUNITY of Elbmarsch consists of a chain of several small villages located at the southern bank of the river Elbe, about 35 km southeast of Hamburg. In 1990, a leukemia cluster appeared in children living there (Hoffmann et al. 1997). Most cases up to now occurred in the village Tespe, lying opposite to the nuclear research center Gesellschaft für Kernenergieverwertung in Schiffbau und Schiffahrt (GKSS), established in 1958 (Fig. 1). West of GKSS, at a distance of about 1.5 km, a nuclear power plant named Kernkraftwerk Krümmel (KKK) began full operation in 1984. It is equipped by a 1,300 MWel boiling-water reactor.

The primary initial project of the GKSS was research and development on the harnessing of nuclear power for commercial ships. In the eighties, the center was engaged in several research programs on nuclear reactor safety and the development of components for nuclear power plants. It was equipped with two nuclear research reactors of 5 and 15 MW capacities.

In 1990, about 1,500 children under the age 15 y lived in the Elbmarsch community. According to the Childhood Cancer Registry for West Germany, one leukemia case would have been expected there in 15 y. However, five cases appeared between 1990 to 1991. Four additional cases occurred by 1996 in the 5-km area surrounding KKK. In the period 1990 to 1996, the incidence of childhood leukemia in that region was elevated by a factor of 5.6 (Schmitz-Feuerhake et al. 1997). All affected children were less than 11 y old at time of the diagnosis, and five were below 5 y of age.

Because of the temporal correspondence between the start-up of KKK and the appearance of the leukemia cases—regarding known latencies for radiation-induced malignancy—the suspicion arose that non-permitted emissions of the plant might be responsible for the induction of the diseases. The supervising ministry, however, claimed undisturbed operation for KKK, the two research reactors, and the experimental facilities of GKSS and denied any relevant environmental contamination.

Analyses of the environmental monitoring data were also carried out by members of an expert committee, officially appointed to identify possible causes of the observed health effect. They registered several increases of fission and activation products between 1984 and 1990 (Schmitz-Feuerhake et al. 1996; Schmidt et al. 1998). Biological dosimetry for 21 adults living in Elbmarsch showed a significant elevation of dicentric chromosomes (Schmitz-Feuerhake et al. 1997).

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(*Manuscript received* 8 *March* 2002; *revised manuscript received* 19 *September* 2002, *accepted* 10 *December* 2002)

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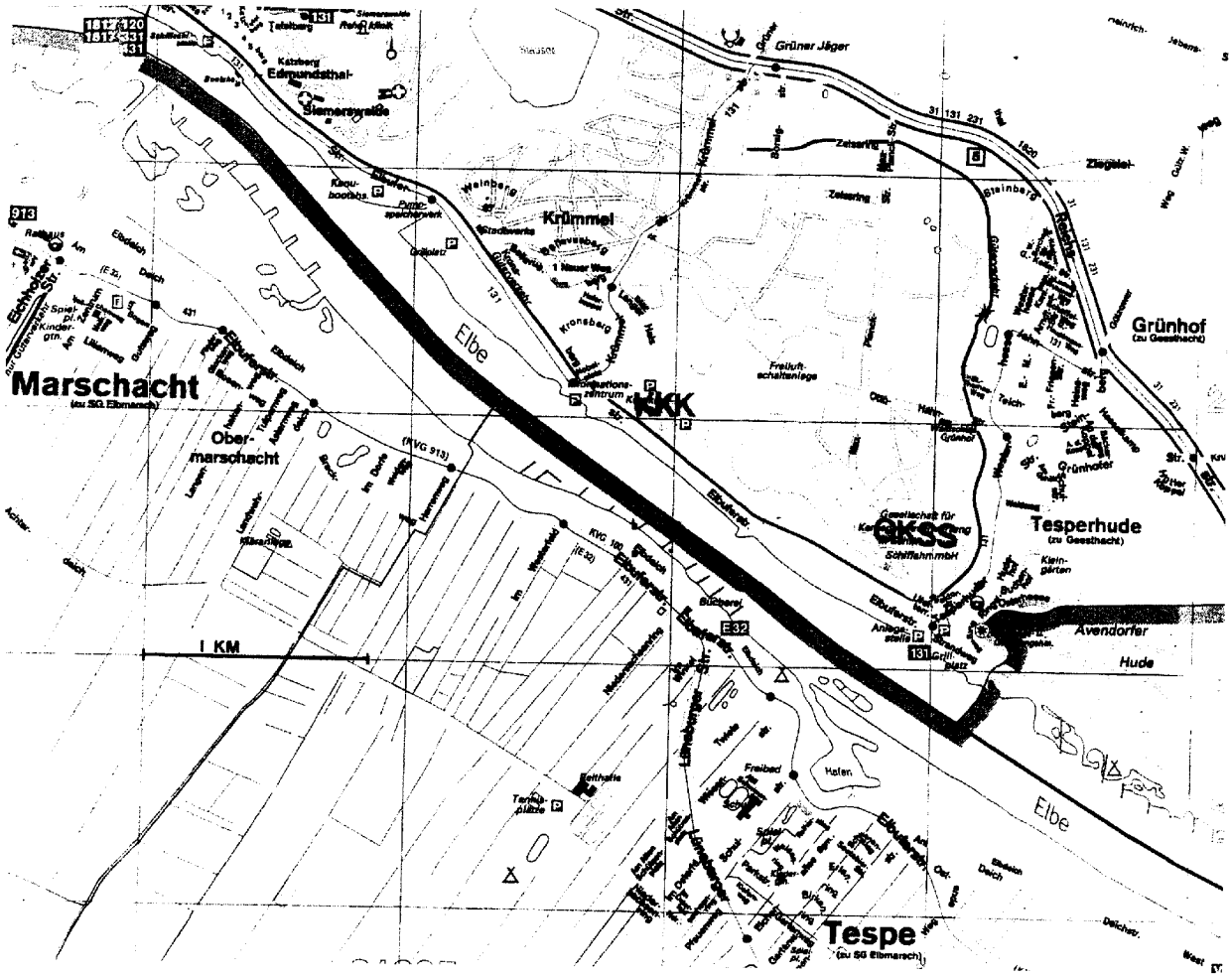

Fig. 1. Map of the investigated Elbmarsch region.

The KKK and GKSS environmental monitoring programs do not call for continuous measurements of pure beta- and alpha-emitting nuclides in the controlled media except for ^{90}Sr in the air. Therefore, it was decided to look for transuranic nuclides delivered in the preceding period, i.e., at least since the test operation phase of KKK in 1983, and also for ^{90}Sr as a possibly relevant bone-seeking nuclide responsible for the induction of leukemia.

MATERIALS AND METHODS

Attic dust in older houses, where the roof had not been restored, was thought to be a suitable means to detect contaminations from the past in a rather concentrated form. In 1997 and 1998, inhabitants of such houses were instructed to collect attic dust in a fresh paper bag from their vacuum cleaner. Twelve samples from different locations in Elbmarsch were obtained, which had masses between 60 and 400 g. In these samples a screening measurement was undertaken by gamma spectrometry. Seven of the samples showed ^{241}Am above or near the detection limit.

Five of the Elbmarsch samples were selected for further analyses. Three were from Marschacht and two from Tespe, which are the villages located nearest to the nuclear establishments (Fig. 1).

Five samples from different places in northern Germany were taken as a control. They are described in Table 1.

The material was passed through a sieve of 1-mm hole diameter in order to eliminate coarse particles. Thirty to fifty grams of each sample, either in their original form or after incineration to ash, were sent to the

Table 1. Locations for control samples of attic dust.

Location	Installation of attic	Distance to Elbmarsch (km)	Date of sampling	Sample mass (g)
Adendorf	circa 1992	14	August 1998	75
Bremen	1957	~110	Sept. 1998	100
Grossefehn near Aurich	≤1988	~250	Sept. 1998	107
Lüneburg	1963	20	July 1998	116
Scharnebeck near Lüneburg	unknown	14	July 1998	103

investigating laboratory for the transuranic analyses. Another portion of the material was saved for future measurements of ^{90}Sr.

The procedure for plutonium nuclides followed the method applied by LaRosa and his co-workers (1992) at the IAEA Laboratories Seibersdorf for the analyses of the Chernobyl Project samples. Only small modifications, described elsewhere (Mietelski and Wąs 1995), were introduced. The analysis was combined with an Am/Cm radiochemical procedure (Holm and Ballestra 1989; Mietelski and Wąs 1997). Determination of ^{90}Sr activity was done separately. Both procedures started with incineration at 600°C for ≥8 h. Then each sample was homogenized, weighed, and its respective dry mass calculated.

Pu, Am/Cm procedure

After transferring the ashes to PTFE beakers, known amounts of ^{236}Pu and ^{148}Gd tracers were added. The samples were dissolved by means of sequential application of hot concentrate acids including HF, HNO_3, HCl with added H_3BO_3 and again HNO_3. The solution was diluted to a HNO_3 concentration of 1 M and filtered after cooling. The oxidation adjustment procedure (LaRosa et al. 1992) was performed using hydrazine and $NaNO_2$. After that, the solution was diluted to a HNO_3 concentration of 8 M and cooled down. Thorium and plutonium were separated from all the other elements by anion-exchange chromatography using DOWEX 1×8, 200 mesh, Cl^- form (Fluka Ltd., SAF Germany, D-89231 Neu-Ulm, Germany). Thorium was stripped off the column with 50 mL of 10 M HCl, and plutonium was then stripped off with 25 mL of 0.1 M HCl–0.1 M HF solution.

Thin alpha spectrometric sources were prepared directly from solutions using the NdF_3 co-precipitation method (Sill 1987; LaRosa et al. 1992). Americium, curium, and all rare-earth elements including the gadolinium tracer were expected to pass the columns without any retention, so they should remain in the 8M HNO_3 solution. These solutions were evaporated to about 20 mL, diluted to 200 mL, 30 mL of saturated oxalic acid solution was added, and the pH was adjusted to 3. Under these conditions, americium, curium, and the rare-earths co-precipitate with oxalates (Holm and Ballestra 1989). The precipitated oxalates were centrifuged, dried, and then decomposed at 600°C. The residues were dissolved in hot 14 M HNO_3, and a small amount of iron chloride was added. Then the solution was diluted with de-ionized water and adjusted to pH 9 using ammonia. This was done to separate the rare-earths, americium, and curium, which co-precipitate with iron hydroxides from any bi-valent element ions, which stay in the solution (Holm and Ballestra 1989). The precipitated iron hydroxide was centrifuged, dissolved in hot *aqua regia*, converted to a HNO_3 concentration of 8 M and the anion-exchange procedure repeated in order to purify the solution from any remaining traces of thorium.

After passing through the column, the solution was evaporated to 3 mL, and 40 mL of CH_3OH was added. This solution was passed through further DOWEX-1 columns pre-conditioned with a similar solution. Americium, curium, and rare-earths were expected to be in a complex configuration in the column (Holm and Ballestra 1989). They were finally removed together from the column using 50 mL of 1 M HCl in CH_3OH. The solution was evaporated to dryness, and the residue was re-dissolved in 12 M HCl and then diluted to a HCl concentration of 1 M. NdF_3 co-precipitation sources were prepared (Sill 1987).

Alpha spectrometric measurements were done using an alpha-spectrometer Silena AlphaQuattro (Silena International Spa., Via Firenze, 3-20063 Cernusco sul Naviglio (Mi), Italy) with Canberra PIPS silicon detectors. The alpha-sources were measured in a detector to source distance of about 1.5 mm. The obtained spectra were analyzed using ALF software (Mietelski and Wąs 1995).

After the alpha spectrometric measurements, the plutonium sources were dissolved in small amounts of hot 14 M HNO_3 with traces of H_3BO_3 added, evaporated to dryness, re-dissolved in 2.5 mL of 1 M HNO_3, mixed with liquid scintillation cocktail Wallac HiSafe3, and measured for ^{241}Pu using a Wallac 1414-003 Guardian liquid scintillation spectrometer (PerkinElmer Life Sciences—Europe, Imperiastraat 8, B-1930 Zaventem, Belgium). The total alpha emitting plutonium activity present in the source, already known from the alpha spectrometric measurement, was used to monitor the chemical recovery. The tritium protocol together with the built-up library data were applied for all quenching and efficiency problems. Details of the applied method of ^{241}Pu activity calculation were described elsewhere (Mietelski et al. 1999).

^{90}Sr measurements

The applied radiochemical procedure for ^{90}Sr was a modification of our previous method for bone matrix

samples (Mietelski et al. 2001). Samples of about 5 g ash weight each were mineralized by a subsequent treatment with hot concentrated HF, HNO_3, HCl (with H_3BO_3 added), and again HNO_3. Then they were diluted to 1 M HNO_3. The solutions were filtered after cooling. Oxalic acid and ammonia were added, and the oxalates were precipitated at pH 6 from the hot solution. The centrifuged oxalates were decomposed in concentrated hot HNO_3. The solution was diluted to 3 M HNO_3 and deposited on a Sr-Resin (Eichrom Ltd., Eichrom Technologies, Inc., 8205 Cass Ave., Suite 106, Darien, IL 60561, USA) column (10 cm high, 0.4 cm inner diameter). Strontium was stripped off with water and the solution evaporated to 2 mL and mixed with liquid scintillation cocktail Wallac HiSafe3. The samples were then left for 2 wk to achieve secular equilibrium between ^{90}Sr and its progeny, ^{90}Y. ^{90}Sr activity was measured in a Wallac 1414-003 Guardian liquid scintillation spectrometer using only the high energy part of the beta spectrum to reduce the interfering influence of traces of ^{210}Pb in the spectra. The yield of the radiochemical procedure was calculated from the stable strontium concentration, which was estimated after mineralization of the sample and before mixing the final solution with scintillation cocktail. These measurements were done using flame emission spectrometry (Perkin-Elmer 5100 ZL spectrometer, PerkinElmer Life Sciences Inc., 549 Albany Street, Boston, MA 02118-2512, USA). For each of these measurements 10% of the appropriate solution was taken out.

RESULTS

The results of the attic dust measurements are shown in Table 2, except for ^{244}Cm. All results for this nuclide lay below the detection limits in the range of 0.01–0.2 Bq kg $^{-1}$ of ash.

The mean concentrations for all measured nuclides, except ^{244}Cm, were higher in the Elbmarsch houses than in the controls (Table 2, last line). ^{90}Sr and plutonium nuclides were 2–3 times higher than controls. An extreme difference resulted for the nuclide ^{241}Am, which showed a 19-fold higher concentration. This is due to the findings in the Marschacht 3, Marschacht 4, and Tespe 3 samples. (The sample for "Grossefehn" in the ^{90}Sr series was lost.) The attic dust concentrations for the alpha-emitting transuranium nuclides are shown in Fig. 2. The ^{90}Sr concentration was correlated to that of ^{241}Am for all measuring points, which was not the case for ^{90}Sr against $^{239,240}Pu$ (Fig. 3).

DISCUSSION AND CONCLUSION

In Germany, the contents of transuranic nuclides in the normal background contamination are dominated by the fallout of the atomic bomb tests until 1965. The Chernobyl accident contributed only a marginal elevation. Measurements in the region of Munich in soil and grass resulted in the following contributions to the former deposition: ^{238}Pu by 1%, $^{239,240}Pu$ by 0.1%, and ^{241}Pu by 0.4% (Bunzl and Kracke 1990; SSK 1989). The transuranic contamination in northern Germany is even lower.

Therefore, whether the Elbmarsch samples show significant deviations from normal background can be

Table 2. Results of the measurements in attic dust, values in Bq/kg of ash.
The values are given with the 1 S.D. statistical counting error.

Sample	^{90}Sr	^{238}Pu	$^{239,240}Pu$	^{241}Pu	^{241}Am
ELBMARSCH (E)					
Marschacht 2	7.2 ± 0.4	0.072 ± 0.013	1.34 ± 0.11	4.5 ± 0.1	0.88 ± 0.10
Marschacht 3	53.6 ± 2.0	0.189 ± 0.015	5.86 ± 0.31	15.3 ± 0.2	10.7 ± 1.2
Marschacht 4	11.5 ± 0.5	0.046 ± 0.004	0.93 ± 0.05	2.5 ± 0.1	2.1 ± 0.6
Tespe 2	8.0 ± 0.6	0.042 ± 0.007	0.79 ± 0.06	3.6 ± 0.1	0.34 ± 0.10
Tespe 3	25.3 ± 0.5	0.065 ± 0.007	1.32 ± 0.07	2.5 ± 0.1	3.8 ± 0.5
Mean	21.1	0.083	2.05	5.7	3.6
1 S.D. range	11.4–30.8	0.053–0.113	0.98–3.12	3.0–8.4	1.5–5.7
CONTROLS (C)					
Adendorf	5.7 ± 0.5	0.041 ± 0.007	0.77 ± 0.06	2.8 ± 0.1	0.10 ± 0.06
Bremen	13.1 ± 1.2	0.076 ± 0.010	1.42 ± 0.09	2.6 ± 0.1	0.65 ± 0.12
Grossefehn	—	0.038 ± 0.005	1.05 ± 0.06	2.3 ± 0.1	0.16 ± 0.13
Lüneburg	6.3 ± 0.7	0.003 ± 0.003	0.14 ± 0.01	<0.1	<0.03
Scharnebeck	3.8 ± 0.9	0.023 ± 0.006	0.28 ± 0.03	3.2 ± 0.1	<0.01
Mean[a]	7.2	0.036	0.73	2.2	0.19
1 S.D. range	4.9–9.5	0.023–0.049	0.46–1.00	1.6–2.8	0.06–0.32
Ratio (E/C)	2.9	2.3	2.8	2.6	19

[a] The values below the detection limit were considered by taking half of the detection limit.

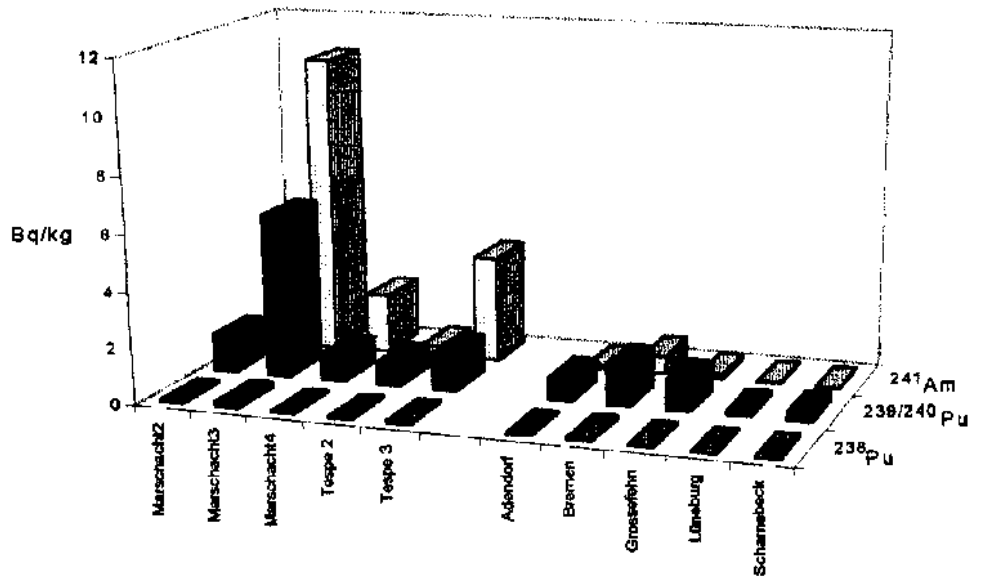

Fig. 2. Alpha-emitting transuranium nuclides in attic dust; left side: Elbmarsch; right side: controls.

determined by comparison with the isotopic ratios in atomic bomb fallout (Table 3).

The activity ratio $^{238}Pu/^{239,240}Pu$ for weapons fallout is reported to be 3.5% (Eisenbud 1987). According to Hardy et al. (1973), the SNAP accident in 1964 (a satellite was equipped with a System for Nuclear Auxiliary Power containing 1 kg of ^{238}Pu) with a release of ^{238}Pu led to a ratio of 3.6% in the northern hemisphere (Munich 3.1%; Roskilde/Norway 3.6%). Measurements in German soils resulted in ratios of about 3% (Bunzl and Kracke 1987) and 3.7 ± 0.33% (Bunzl and Kracke 1988). Compared to these measurements, the corresponding ratios in Elbmarsch (Table 3, second column) and control samples do not show any significant deviation.

The activity ratio $^{241}Pu/^{239,240}Pu$ (Table 3, third column) also does not indicate a deviation from normal background. At the time of the atomic tests it was about 15:1 (Hakanen et al. 1984) and is expected to be about 2:1 for weapons fallout in 1999.

Considerable alterations, however, are shown in the relations including ^{241}Am. ^{241}Am, as the progeny of ^{241}Pu (14.4 y), increased until 1999 up to an activity of 0.3–0.4 in relation to $^{239,240}Pu$ (Krey et al. 1976; Bunzl and Kracke 1987; Ryan et al. 1993). As a consequence of the elevated ^{241}Am activity (Table 2) the Elbmarsch samples (Marschacht 3, Marschacht 4, and Tespe 3) show a completely deviant increase of the ratio $^{241}Am/^{239,240}Pu$. This is true also for the ratio $^{241}Am/^{241}Pu$, which is far above expected values in these samples (about 0.15–0.2 for weapons fallout in 1999). It seems rather unlikely that any decay mechanism might have affected the Pu/Am ratio since the attic dust was not exposed to any direct weathering processes.

The Elbmarsch contamination by transuranic nuclides therefore cannot be accounted for by the background levels resulting from weapons or Chernobyl fallout. It is, however, not similar to the inventory of a normal operating light water reactor. This can be inferred from the measured proportion of ^{238}Pu, which lies in a range of 0.03–0.08 related to $^{239,240}Pu$. Such low ratios only exist in the initial operating phase of the reactor, where the build-up of ^{241}Pu and ^{241}Am is low (Wilson et al. 1988). The peculiarity of the Elbmarsch results must be seen, however, in the high amount of ^{241}Am and the relatively high ratios $^{241}Am/^{241}Pu$ and $^{241}Am/^{239,240}Pu$, which are not compatible with normal fuel burn-up.

There was only a slight deposition of ^{90}Sr in Germany from the Chernobyl accident. For northern Germany, a contribution of 5% to the former fallout was reported (SSK 1987). Fig. 3 shows that the ^{90}Sr concentrations in attic dust samples of Elbmarsch do not correspond to those of $^{239,240}Pu$ but to those of ^{241}Am. The $^{239,240}Pu$ deposition in Elbmarsch must be interpreted after our measurements as a combination of radionuclides from weapons fallout and from some additional source, while the ^{241}Am concentration seems to be mainly generated by the additional release. The corresponding appearance of the fission product ^{90}Sr might indicate that it was released together with ^{241}Am. The underlying scenario, however, cannot be derived by these data.

The leukemia incidence in Elbmarsch peaked in the years 1990–1991. According to the findings in Hiroshima and Nagasaki where the incidence in young children peaked about 5 y after the exposure (Shimizu et

Table 3. Ratio of transuranic nuclides in attic dust in comparison to expected values by nuclear weapons fallout in 1999 (values in parentheses: 2 S.D. confidence interval).

Sample	$^{238}Pu/^{239,240}Pu$	$^{241}Pu/^{239,240}Pu$	$^{241}Am/^{239,240}Pu$	$^{241}Am/^{241}Pu$
Expected for weapons fallout	0.03–0.04	~2	0.3–0.4	0.15–0.2
ELBMARSCH (E)				
Marschacht 2	0.054	3.4	0.66	0.20
	(0.032–0.075)	(2.8–3.9)	(0.47–0.84)	(0.15–0.24)
Marschacht 3	0.032	2.6	1.8	0.70
	(0.026–0.038)	(2.3–2.9)	(1.4–2.3)	(0.54–0.86)
Marschacht 4	0.049	2.7	2.3	0.84
	(0.039–0.060)	(2.3–3.0)	(0.9–3.6)	(0.36–1.32)
Tespe 2	0.053	4.6	0.43	0.09
	(0.034–0.073)	(3.8–5.3)	(0.17–0.69)	(0.04–0.15)
Tespe 3	0.049	1.9	2.9	1.5
	(0.037–0.061)	(1.6–2.1)	(2.1–3.7)	(1.1–1.9)
Mean E	0.040	2.8	1.8	0.67
CONTROLS (C)				
Adendorf	0.053	3.6	0.13	0.04
	(0.020–0.073)	(3.0–4.3)	(0.03–0.29)	(0.01–0.08)
Bremen	0.054	1.8	0.46	0.25
	(0.038–0.069)	(1.6–2.1)	(0.28–0.64)	(0.16–0.34)
Grossefehn	0.036	2.2	0.15	0.07
	(0.026–0.047)	(1.9–2.5)	(0–0.40)	(0.04–0.18)
Lüneburg	0.021	<0.7	<0.2	n.c.[a]
	(0–0.06)			
Scharnebeck	0.082	11.4	<0.04	<0.003
	(0.096–0.128)	(8.9–14)		
Mean C	0.049	3.0	0.26	n.c.

[a] n.c. = not calculable (means that a quotient was not calculable), see Table 2.

exposure as would be expected from incorporated radionuclides with long effective half-lives. Quantitative dose estimations according to the yield of dicentric chromosomes are not possible under such conditions. A further important result of the study was given by the distribution of the dicentric chromosomes among the cells, which did not follow a Poisson pattern but showed a significant overdispersion. In the low dose region, this is an indicator for high linear energy transfer radiation. Therefore, the study warranted the search for alpha emitters.

12 September 1986, appears as a possible date of an underlying event. As was reported by local newspapers, persons in protective clothing were observed on the grounds of the nuclear power plant working with radiation monitors. The emission monitoring of the plant registered a 2- to 4-fold elevation of gamma-emitting aerosols in the downwind pathway between 7.30 h and 11.30 h, but no source for elevated releases was detected in the plant.

The authorities for reactor safety explained the event as an unusual accumulation of natural radon because of calm and inversion weather conditions and reported an outdoor aerosol concentration of about 500 Bq m^{-3}. They stated that the nuclear power plant had drawn in radioactivity via the inlet air fan. The inlet is, however, 44 m above ground level, which corresponds to at least 14 diffusion lengths for radon (Porstendörfer 1993). Hence, the interpretation of the authorities cannot be accepted. Furthermore, the environmental monitoring systems of the nuclear establishments indicated contaminations on that day that excluded any relation to the natural emitters. A routine monitoring of soil in Marschacht (a village of Elbmarsch, Fig. 1) showed an increase of the beta-surface activity of about 2,400 Bq m^{-2} (KKK 1986). Measurements by gamma spectroscopy of sediments of the Elbe at locations upriver (relative to the plants) showed a variety of long and short-lived fission products in samples drawn on 15 September 1986 (GKSS 1983–1991). The existence of these fission products cannot be explained on the basis of the Chernobyl accident, since the corresponding measurements taken before 12 September 1986 and after the date of the Chernobyl accident showed no elevation in the concentration of fission products in sediment.

It is unknown, however, if these fission products were accompanied by transuranic nuclides. The profile of these nuclides in attic dust support the assumption that experiments with special nuclear fuels were carried out at the GKSS site. MOX fuel, for instance, would elevate ^{241}Am, because it contains ^{241}Pu from reprocessing. A delivery of nuclear fuel—possibly because of a critical

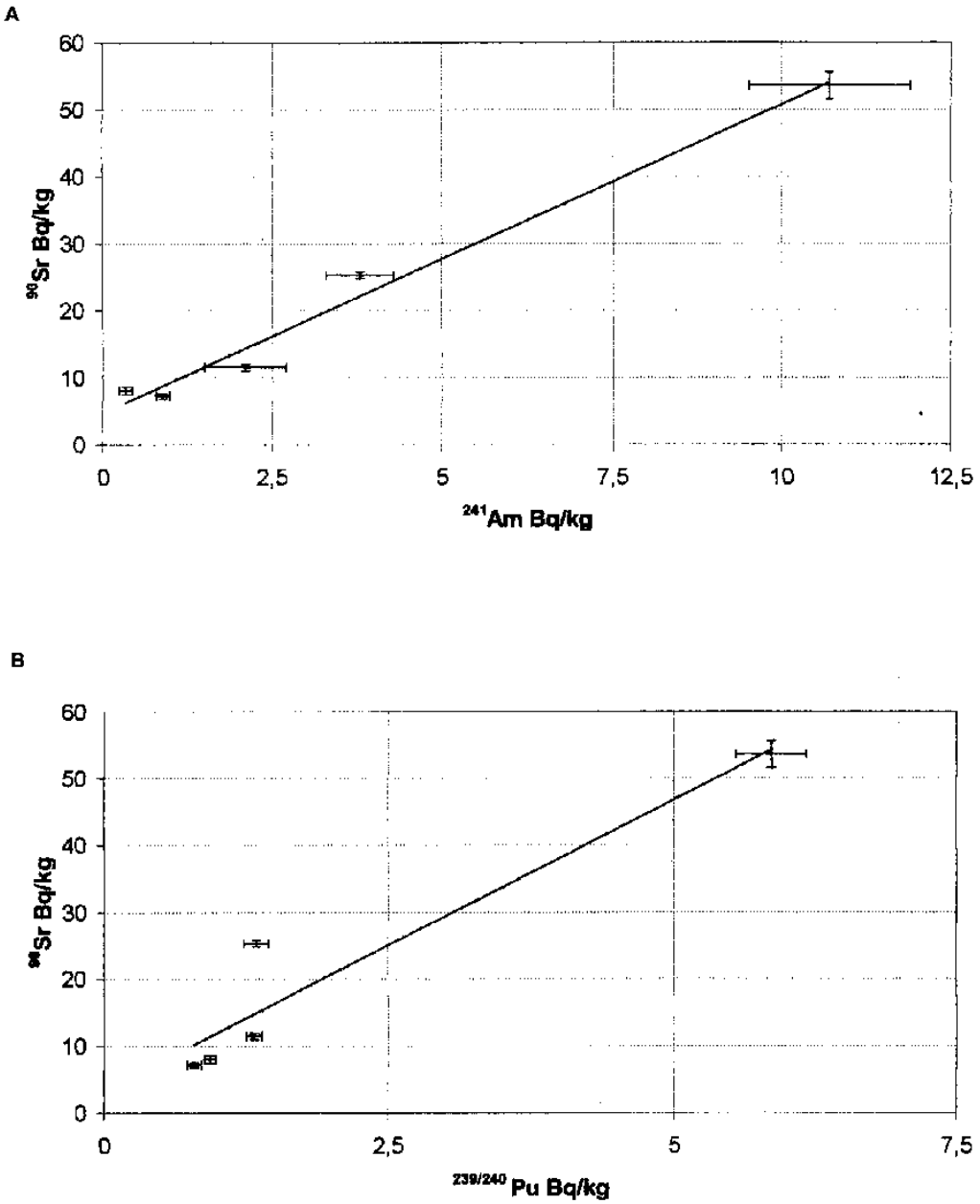

Fig. 3. a) Dependency between ^{90}Sr and ^{241}Am in Elbmarsch samples; b) Dependency between ^{90}Sr and $^{239/240}$Pu in Elbmarsch samples.

al. 1990), a putative causal radiation event in Elbmarsch could be assumed to have occurred about 1986.

An investigation on chromosome aberrations in inhabitants of Elbmarsch—among them 7 parents of leukemia children—had been done during the years 1991–1993. It showed a near to 4-fold significant elevation of dicentric chromosomes in peripheral blood lymphocytes (Schmitz-Feuerhake et al. 1997). As the aberrations are unstable, these findings several years after the purported radiation event indicate a kind of continuous

event—is confirmed by the fact that enriched uranium was found in 3 of the Elbmarsch samples of the attic dust. A review of the measurements of soil radioactivity by several institutions pertaining to the Elbmarsch problem resulted in repeated findings of enriched uranium (Schmitz-Feuerhake[‡]). The screening measurements in attic dust also showed elevated concentrations of the fission product ^{137}Cs, which correspond to the reported findings for ^{90}Sr. Furthermore, activation products like ^{22}Na, ^{57}Co, and ^{60}Co were detected in the Elbmarsch houses.

Further studies on the environmental contaminations must be undertaken in order to identify the source of contamination and its relation to leukemia-induction among children of Elbmarsch between 1990–1996. A case-control study of the whole population living in that part of Germany was sponsored by the corresponding Federal States in order to identify the possible causes for the leukemias.

Acknowledgments—The measurements were financed by the Citizen's Initiative against Leukemia in Elbmarsch who also organized the sampling of attic dust.

Pawel Zagrodzki of INP and Collegium Medicum UJ (Kraków) is warmly acknowledged for the determination of stable strontium concentrations.

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