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I. Winkelsmann et al.

Bundesamt für Strahlenschutz, Waldowallee 117, 10318 Berlin

In-situ-Spektrometrie zur Messung der Umweltradioaktivität

Zusammenfassung

Die Methode der In-situ-Spektrometrie zur Überwachung der Umweltradioaktivität wird beschrieben. Die meßtechnischen Möglichkeiten und Einsatzbereiche werden erläutert. Die Kalibrierung des In-situ-Spektrometers mit Halbleiterdetektor und die Auswertung der Spektren werden diskutiert. Meßzeiten von 15 - 30 Minuten reichen im allgemeinen aus, um mit Hilfe der In-situ-Spektrometrie die spezifische Aktivität natürlicher Radionuklide im Boden zu bestimmen und Kontaminationen künstlicher auf dem Boden abgelagerter Radionuklide von einigen 10 Bq/m² noch zu messen. Über Messungen der Bodenkontamination im südlichen Deutschland nach dem Reaktorunfall in Tschernobyl wird berichtet. Insgesamt konnten dabei 22 künstliche Radionuklide gemessen werden. In den ersten Tagen nach dem Reaktorunfall lieferten die Radionuklide ¹³²Te/¹³²I und ¹³¹I mit jeweils 100 kBq/m² bzw. 70 kBq/m² den Hauptanteil zur Bodenkontamination.

In situ gamma ray spectrometry for measurements of environmental radioactivity

Abstract

The method of in situ gamma ray spectrometry for measurements of environmental radioactivity is described. The range of application is shown. The calibration of the in-situ gamma ray spectrometer with semiconductor detector and the evaluation of the spectra are described. A measuring time of about 15 - 30 minutes is sufficient to determine the specific activity of natural radionuclides in soil or artificial radionuclides of some ten Bq/m². The results of nuclide specific measurements of soil contamination in Germany after the Chernobyl reactor accident are reported. A total of 22 artificial radionuclides could be detected. In the first days after the Chernobyl accident, the main contamination resulted from the radionuclides ¹³²Te/¹³²I and ¹³¹I with about 100 kBq/m² each and 70 kBq/m², respectively.

1 Introduction

In Germany the operators of nuclear power plants perform, among other evaluations, continuous measurements of the dose rate in the vicinity of their facilities. These measurements are relatively simple to perform using a scintillation dose rate meter. This method, however, does not allow to assess individual radionuclides contributing to the total dose rate. In-situ spectrometry, on the other hand, is a suitable method to gain rapid and detailed information on radiation in the environment along with the resulting dose rate of individual radionuclides. The possibility of carrying out rapid measurements of the environmental radiation by this method also enables to ascertain higher releases of radioactive substances caused by incidents in nuclear power plants (e. g. soil contamination, noble gas emissions). Especially after the Chernobyl accident, it was necessary to obtain a quick overview of the actual soil contamination and the resulting exposure rate.

2 Application

In-situ spectrometry is suited for rapid measurement of the activity of individual radionuclides in the environment and for assessment of the resulting gamma dose rate. The following applications are considered in detail:

- Individual nuclide measurement of specific soil activity (e. g. ascertainment of the natural radionuclide concentration and its contribution to the dose rate).
- Individual nuclide measurement of soil contamination (e. g. measurement of the accumulation of radionuclides over a long period of time, measurements during and after accidental releases, fallout measurements along with the resulting gamma dose rate contribution of individual nuclides).
- Nuclide specific measurement of the activity in the air (assessment of radioactive noble gases and iodine in air during and after accidents).

3 Measuring system

The in-situ spectrometer used by the Federal Office for Radiation Protection consists of a high purity germanium detector with a relative efficiency of 50 % and a peak-to-compton ratio of 58.6 to 1. The detector is connected to a portable multichannel analyzer and a tape recorder. This system already allows a preliminary evaluation of the gamma spectrum during the actual measurement. In general, the detector is mounted 1 m above ground. A measuring time of about 15 - 30 minutes is sufficient to determine the specific activity of natural radionuclides in soil such as ^{40}K , ^{232}Th and ^{238}U . Lower limits of detection for several artificial radionuclides of some ten Bq/m^2 can be achieved.

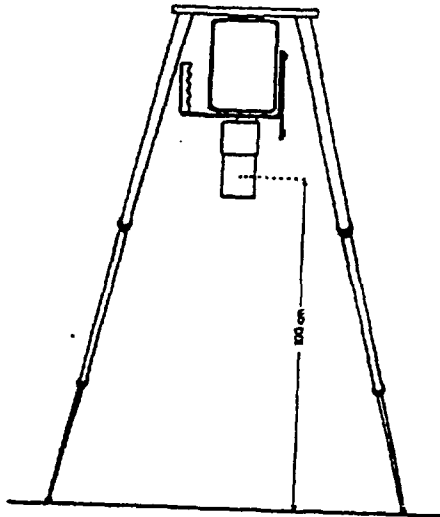


Figure 1: Portable germanium detector on a tripod.

4 Calibration of an in-situ spectrometer

The calibration factor gives the relation between netto peak counting rate and activity concentration, activity or specific activity, depending on the distribution of radioactive substances in a medium. The calibration factor N_f/A combines three terms:

- the detector response R_0 , which only depends on the detector,
- the geometrical factor F_G , depending on nature and distribution of the radionuclides and properties of the irradiated material,
- the correction factor R_G , which depends on the angular detector response $N(\Theta)/N_0$ and on the gamma flux density Φ .

$$\frac{N_f}{A} = \frac{\text{netto peak count rate}}{\gamma\text{-activity}} = \text{calibration factor}$$

$$\frac{N_f}{A} = R_0 * F_G * R_G = \frac{N_0}{\Phi} * \frac{\Phi}{a_G} * \frac{N_f}{N_0}$$

$\frac{N_0}{\Phi} = R_0$; detector response function, gives the energy dependance of the detector efficiency

$$\frac{\Phi}{a_G} = F_G; \frac{\gamma\text{-flux density at the detector}}{\gamma\text{-activity with a given geometry G}} = \text{geometrical factor}$$

$$\frac{N_f}{N_0} = R_G; \text{correction factor, considering the angular response of the detector and the angular distribution of the } \gamma\text{-flux density for various geometries G}$$

4.1 Geometrical factor

The geometrical factor is a measure for the flux of unscattered photons generated by a medium with a definite activity concentration at the site of the detector. The geometrical factor depends strongly on the distribution of radioactive substances in soil or air. Because extended calibration sources cannot be realized in practice, it has to be simulated by mathematical models. This method has been described by Beck et al. The following source geometries have been considered:

- homogeneous activity distribution in the lower half space for the natural radionuclides with an activity concentration a_v [Bq/cm³] and a specific activity a_v/ρ [Bq/g];
- an activity concentration $a_v(z)$ decreasing with increasing depth (as model for the radionuclides deriving from fallout which migrate into the soil according to the function

$$a_v(z) = a_{v0} * \exp(-az)$$

with $1/a$ is the relaxation depth [cm], in which the activity concentration decreases to $1/e$, and with $a_{v0} = a_v(z=0)$: activity concentration for $z = 0$.

- homogeneous activity distribution a_f [Bq/cm²] on the soil surface (e. g. fresh fallout);
- homogeneous activity distribution a_v [Bq/cm³] in the air, and
- line sources in air.

The calculation scheme is summarized in annex 1.

4.2 Detector efficiency

The detector efficiency is tested by measuring the detector response to a unit flux of photons incident normal to the detector window. Usually one uses several point sources with different gamma energies. In order to get a nearly parallel beam of incident photons at the detector window the point source is placed $r = 100$ cm distant from the detector. The detector response is given by

$$R_0 = 4\pi r^2 * \frac{\text{netto pulse rate of the full energy peak}}{\text{emitted quanta of the source}} \quad [\text{cm}^2]$$

4.3 Angular response

The angular response $N(\Theta)/N_0$ is determined by comparing the detector response to photons incident at various zenith angles Θ to the normal with that for photons incident normal to the detector window. All variations in azimuthal response have to be determined, e. g. caused by attenuation through the high voltage power supply filter, preamplifier, crystal mounting or dewar handles. The response is weighted by the expected flux at each angle $\Phi(\Theta)$ in a given field situation (i. e. depth distribution and gamma-ray energy) in order to obtain the final angular calibration factor N_f/N_0 . For a detector without angular dependance on the detection limit, the correction factor reduces to $N_f/N_0 = 1$. This is the case for the detectors used by the Federal Office for Radiation Protection.

5 Evaluation

In order to evaluate the soil contamination from measured spectra, knowledge of the distribution of the radionuclides in the soil is essential. Several soil samples gave evidence that the distribution of the artificial radionuclides could be described by an exponential decrease with increasing depth with the relaxation depth. The relevant parameter for the evaluation is the relaxation depth $1/a$. Generally, it is determined by gamma spectrometric measurements of soil samples in layers. Another method is the comparison of count rates measured in photopeaks of lines at different energies emitted from the same

radionuclide. It indicates a change in the distribution of radionuclides in the soil from data measured with an in-situ spectrometer. The soil contamination calculated from low energy peaks should be less than the corresponding values calculated from high energy peaks with increasing relaxation depth. The gamma ray energies of the radionuclide under consideration should be clearly separated from each other. ^{140}La which shows intensities of 20.7 %, 45.9 % and 95.4 % at energies of 328.8 keV, 487.0 keV and 1596.5 keV could be a suitable radionuclide for such a method.

By knowing the activity distribution of the individual nuclides in the soil, the resulting dose rate can be calculated from the measured spectrum. An extended list of dose rate factors for various fission and activations products, for soil surface contamination as well as for exponential distribution and for 4 different migration depths has been published in /4/.

The activity concentrations of ^{238}U and ^{232}Th cannot be measured directly, but they are determined by the intensities of the gamma lines in the spectra due to the daughter nuclides, assuming the radioactive equilibrium with the corresponding parent. The activity concentration of the natural decay families were determined by evaluating 4 gamma lines, each:

- the gamma line at 352 keV of ^{214}Pb , and lines at 609 keV, 1120 keV and 1765 keV of ^{214}Bi for the U-Ra-series,
- the gamma lines of ^{208}Tl at 583 keV and 2615 keV, and of ^{228}Ac at 911 keV and 965 - 969 keV for the Th-series.

6 Detection limits

As an example, table 1 shows the detection limits attainable by the described in-situ measuring system for some artificial radionuclides and for the specific activity of natural radionuclides, assuming homogeneous distribution on the soil surface and in the soil.

Table 1: Detection limits (confidence level 3σ , rel. detector efficiency 28 %, measuring time 30 min).

Radionuclide	Detection limit
^{60}Co	40 Bq/m ²
^{95}Zr	80 Bq/m ²
^{95}Nb	40 Bq/m ²
^{131}I	40 Bq/m ²
^{137}Cs	40 Bq/m ²
^{40}K	30 Bq/kg
^{232}Th	8 Bq/kg
^{238}U	8 Bq/kg

7 Measurements with an in-situ spectrometer

7.1 Soil contamination

The radioactive cloud from Chernobyl reached the southern part of the Federal Republic of Germany during the early hours of April 30, 1986. Gamma spectrometric measurements with an in-situ spectrometer were immediately executed at the site of the Institute for Radiation Hygiene near Munich in order to determine the soil contamination. Through these measurements, a total soil contamination of about 400 kBq/m² was ascertained for the beginning of May at this site. The results are in good agreement with the measurements of the activity in the rainfall.

During the first days, ¹³²Te/¹³²I and ¹³¹I were the main components of the soil contamination rendering activities of about 100 kBq/m² (50 %) each and 70 kBq/m² (17 %), respectively. The portion due to the radionuclides like ⁹⁹Mo/⁹⁹Tc^m, ¹⁰³Ru, ¹⁰⁶Ru, ¹²⁹Te, and ¹²⁹Te^m as well as ¹³⁴Cs, ¹³⁶Cs, ¹³⁷Cs, and ¹⁴⁰Ba/¹⁴⁰La amounted each to about 1-5 % of the total soil contamination during the first days of May. The soil contamination by ¹³⁷Cs of about 18 kBq/m² at this site is about four times as high as the ¹³⁷Cs contamination due to nuclear weapon testing. In table 2 some results of soil contamination measurements from May to August 1986 are summarized.

Table 2: Results of soil contamination measurements using an in situ gamma ray spectrometer. Site at the Institute for Radiation Hygiene of the Federal Office for Radiation Protection, Neuherberg near Munich

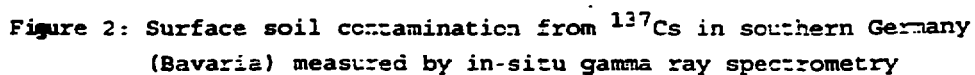
Date 1986 Nuclide	Soil Contamination (kBq/m ²) ^a							
	April 30 ^b before rainfall	May 1 after rainfall	May 14	May 19	June 1	July 3	July 15	August 7
⁹⁵ Zr	-	-	0.31	-	0.30	0.18	0.12	0.13
⁹⁵ Nb	-	0.62	0.51	0.39	0.58	0.45	0.34	0.31
⁹⁹ Mo	-	7.3	-	-	-	-	-	-
^{99m} Tc	0.3	6.6	-	-	-	-	-	-
¹⁰³ Ru	0.75	18	17	15	11	6.4	5.0	3.5
¹⁰⁶ Ru	-	4.2	4.2	4.2	4.2	4.0	4.1	3.8
^{110m} Ag	-	-	0.32	0.34	0.24	0.23	0.26	-
¹²⁵ Sb	-	-	-	-	0.40	0.50	0.41	0.44
¹²⁷ Sb	-	1.5	-	-	-	-	-	-
^{129m} Te	-	18	20	16	12	5.6	4.2	-
¹²⁹ Te	-	12	12	9.4	7.8	3.7	3.4	0.73
¹³¹ I	4.0	68	26	16	5.0	0.29	0.12	-
¹³² Te	3.5	100	6.3	1.9	0.1	-	-	-
¹³² I	4.5	110	6.6	2.0	0.1	-	-	-
¹³³ I	-	1.7	-	-	-	-	-	-
¹³⁴ Cs	0.33	9.4	11	9.7	8.9	8.8	8.5	8.8
¹³⁶ Cs	-	3.3	1.6	0.8	0.6	-	-	-
¹³⁷ Cs	0.66	18	19	17	16	17	16	17
¹⁴⁰ Ba	0.40	11	5.3	3.8	1.7	0.31	-	-
¹⁴⁰ La	0.31	8.8	6.3	4.5	2.2	0.39	0.16	-
¹⁴¹ Ce	-	-	-	0.42	0.35	0.20	0.08	-
¹⁴⁴ Ce	-	-	-	-	0.46	-	-	-
Total	~ 15	~ 400	~ 140	~ 100	~ 70	~ 50	~ 40	~ 40

^a Calculation based on relaxation depth of $1/\alpha = 1$ cm, model of exponential decrease of the activity with depth.

^b Activity in the air already subtracted.

Early in May, this measuring program was extended to cover the whole territory of the Federal Republic of Germany. Due to intensive rainshowers during the passage of the radioactive cloud, the southern part of Bavaria showed the highest contamination. Therefore, the first measurements were carried out in this part of the country. A total of about 100 different sites were chosen for the purpose of gathering data on soil contamination from all parts of Bavaria. Measurements were generally taken on pasture land where the distribution of radionuclides was not disturbed by ploughing. This is of great

importance for the interpretation of the data. Related to May 1st, 1986, the total soil contamination was by a factor 20 higher than the corresponding ^{137}Cs value. For a graphic demonstration of the results from soil contamination measurements, the long-lived ^{137}Cs was selected as the reference nuclide. Figure 2 shows the soil contamination for ^{137}Cs in the southern part of Germany (Bavaria) as determined by in-situ spectrometry measurements. The surface soil contamination in entire territory of Germany due to ^{137}Cs is shown in figure 3 (in-situ spectrometry measurements and soil sample measurements). It varies within a range of 1 kBq/m^2 to 44 kBq/m^2 . The lowest values (generally below 6 kBq/m^2) were found north of the river Danube where the precipitation was less intensive during the passage of the radioactive cloud. South of the river Danube, a significant activity increase in the direction to the Alps is observed, explained by the more intensive precipitation during the time of the highest amount of activity in the air. The highest values were found in southeastern Bavaria.



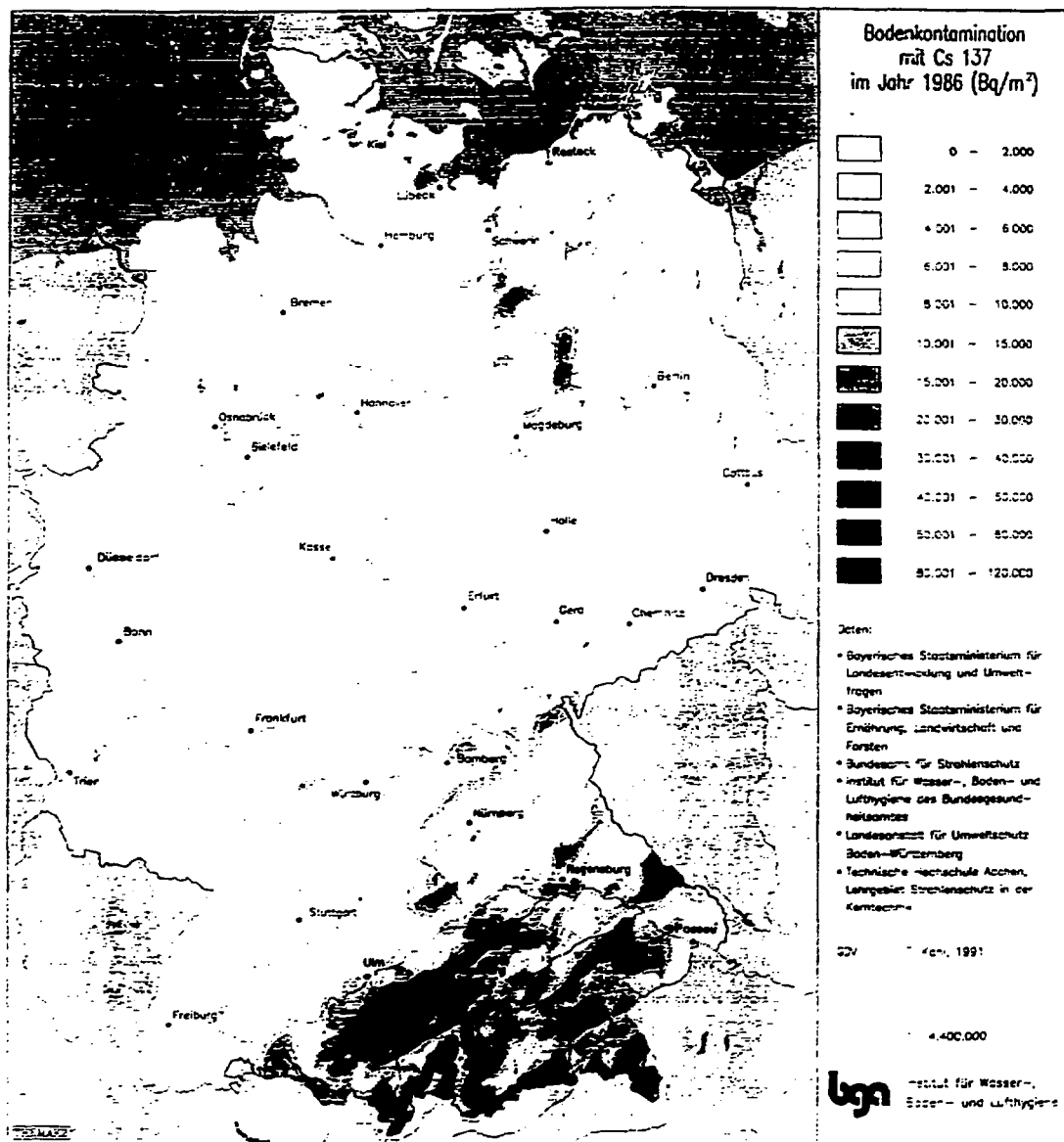


Figure 3: Surface soil contamination from ^{137}Cs in Germany measured by in-situ gamma ray spectrometry (south) and soil sample measurements (north)

Generally, a clearly defined south-to-north decline of radioactive deposition is observed. It is caused by a varying intensity of precipitation in late April and early May 1986, while the activity concentration in air was approximately constant. In general, a clear correlation between soil contamination by ^{137}Cs and the intensity of precipitation by the end of April, as well as during the first days of May, 1986, can be observed.

The contribution to the exposure rate by individual radionuclides was calculated from gamma ray spectra measured with an in-situ spectrometer. For this calculation the depth of migration of radionuclides is of much less importance than it is for determining the soil contamination. Figure 4 shows the relation between soil contamination and exposure rate for individual radionuclides, based on a relaxation depth of 1 cm. This figure clearly shows the main contribution to the exposure rate of about 70 % due to ^{132}I in early May. By May 19, the contribution of this radionuclide to the exposure rate had already decreased to about 7 %, while it became completely insignificant by June 1st. The dominant role played here ^{103}Ru , ^{134}Cs , ^{137}Cs , and ^{140}La . After the decay of the iodine isotopes ^{131}I and ^{132}I in August 1986, about 30 % of the exposure rate came from ^{134}Cs and another 15 % from ^{137}Cs deposited on the soil. The natural sources of cosmic and terrestrial radiation (e. g. ^{40}K , ^{232}Th , and ^{238}U and their daughter products) contributed approximately 30 % and 20 % to the exposure rate with the remaining 5 % due to ^{103}Ru and other radionuclides. Obviously, this relation continuously changes towards the natural radiation sources and ^{137}Cs , since the half-life of two years for ^{134}Cs is comparatively short. In the north of the Federal Republic of Germany, with its relatively low soil contamination by artificial radionuclides, the contribution of cosmic and terrestrial radiation to the exposure rate is still predominant factor.

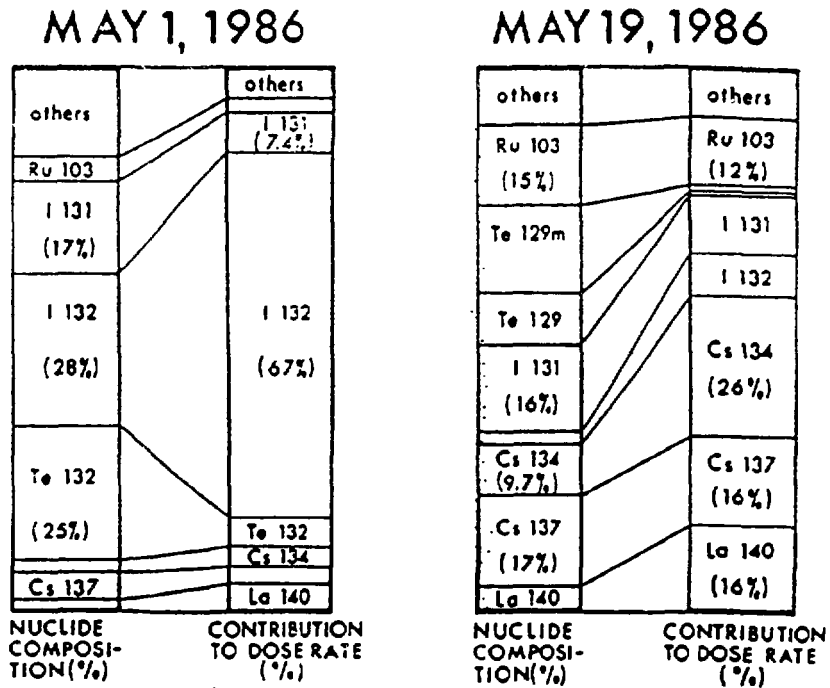


Figure 4: Relation between surface soil contamination and exposure rate of individual radionuclides (relaxation depth 1 cm, exponential decrease of activity with increasing soil depth assumed).

7.2 Noble Gases

With the in-situ spectrometer used for soil contamination measurements the activity concentration of ^{133}Xe in air could be determined on April 30 and May 1, 1986. The results of these measurements are summarized in table 2. The background at the 81 keV peak of ^{133}Xe coming from ^{131}I in air and deposited on soil was taken into account.

Table 3: Activity concentration of ^{133}Xe in air, measured with an in-situ spectrometer.

Date (1986)	Activity concentration (Bq/m ³)*)
April 30 (12.00 p.m.)	160
May 1 (09.30 a.m.)	120
May 1 (12.30 p.m.)	80
May 1 (01.30 p.m.)	50
May 1 (05.00 p.m.)	60
May 1	≤ 40

*) Uncertainty of data about 25 %.

The highest activity concentration of ^{133}Xe in air was 160 Bq/m³ measured on April 30, 1986 (at that time the activity concentration of particulates including iodine was 210 Bq/m³). In the following days the ^{133}Xe activity concentration was less than the detection limit of about 40 Bq/m³ (detector not shielded to reduce the count rate from deposited radionuclides).

Annex 1

Calculation of the geometrical factor

For a point source of activity A and with quantity p_γ of emitted photons of energy E per decay the source strength S is

$$S = A * p_\gamma \text{ [s}^{-1}\text{]}.$$

At the distance r from the source, the photons per second

$$\Phi = \frac{S}{4\pi r^2} \text{ [cm}^{-2}\text{s}^{-1}\text{]}.$$

Flow through one square centimeter of a sphere surface (flux density) with the source in the center. It is assumed that the reduction of photons in the medium between source and detector is neglectable. With media of high density or in high distances, the exponential decrease of the flux density of photons has to be considered:

$$\Phi(\mu r) = \Phi(\mu r_0) * \exp[\mu(r_0 - r)],$$

with $(r_0 - r)$ the distance between detector and source. At the distance r from a point source, the flux density then is

$$\Phi(\mu r) = \frac{S}{4\pi r^2} * \exp(-\mu_M r),$$

with μ_M the mass attenuation coefficient in the medium M.

For the calculation of the flux densities for different geometries, the activity has to be split up in point sources and the flux density can be summarized from all point sources. In praxi, by turning from "discrete point sources" to a continuous distribution of nuclides and replacing the summation by the integration over the total volume, the formula becomes

$$\Phi = \int_V \frac{p_\gamma a_V(r, \theta)}{4\pi r^2} \exp(-\mu_M r) dV.$$

Taking polar coordinates and considering the symmetry around the vertical (according to figure 8), the volume element dV is

$$dV = 2\pi r^2 \sin(\theta) d\theta dr.$$

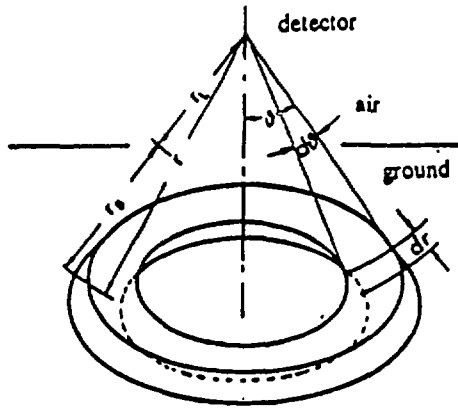


Figure 4: Volume element

The flux of unscattered photons at the site of the detector is

$$\Phi = \int_v \int_{\theta} \frac{p_{\gamma} a_v(r, \theta)}{2} \exp(-\mu_B r_B) \exp(-\mu_L r_L) \sin \theta d\theta dr.$$

The calibration factors calculated with this integral are summarized in table 4.

Table 4: Calibration factors F_G for various geometries.

geometry	activity concentration	calibration factor F_G for various geometries
homogeneous distribution in the soil	$a_v(x,y,z) = a_v [Bq \cdot cm^{-3}]$ $a_m = a_v / \rho_s [Bq \cdot g^{-1}]$	$F_m = \frac{p_T}{2\mu_s / \rho_s} E_2(\mu_L h) \left[\frac{cm^{-2}s^{-1}}{Bq \cdot g^{-1}} \right]$
exponential decrease of the activity concentration in the soil	$a_v(x,y,z) = a_v(z)$ $= a_{v0} \exp(-\alpha z) [Bq \cdot cm^{-3}]$ $a_F = \int_0^\infty a_v(z) dz = a_{v0} / \alpha [Bq \cdot cm^{-2}]$	$F_{v,\alpha} = \frac{p_T}{2\alpha} \left\{ E_1(\mu_L h) - \exp\left(-\frac{\alpha}{\mu_s} \mu_L h\right) E_1\left((1 + \frac{\alpha}{\mu_s}) \mu_L h\right) \right\}$ $F_{Fa} = \alpha F_{v,\alpha} \left[\frac{cm^{-2}s^{-1}}{Bq \cdot cm^{-2}} \right] \text{ or } \left[\frac{cm^{-2}s^{-1}}{Bq \cdot cm^{-2}} \right]$
soil surface contamination	$a_F(x,y,z) = a_F [Bq \cdot cm^{-2}]$	$F_F = \frac{p_T}{2} E_1(\mu_L h) \left[\frac{cm^{-2}s^{-1}}{Bq \cdot cm^{-2}} \right]$
homogeneous distribution in air	$a_{vL}(x,y,z) = a_{vL} [Bq \cdot cm^{-3}]$	$F_{vL} = \frac{p_T}{2\mu_L} \left[\frac{cm^{-2}s^{-1}}{Bq \cdot cm^{-3}} \right]$

with p_T : transition probability of the considered line
 μ_s : mass attenuation coefficient of soil [1/cm]
 μ_L : mass attenuation coefficient of air [1/cm]
 h : detector height above ground
 E_1 : exponential integral function of 1th order
 E_2 : exponential integral function of 2nd order
 ρ_s : soil density [g/cm³]
 $1/\alpha$: relaxation depth [cm]

Annex 2

Assessment of the sample mass considered at measurements with an in-situ spectrometer

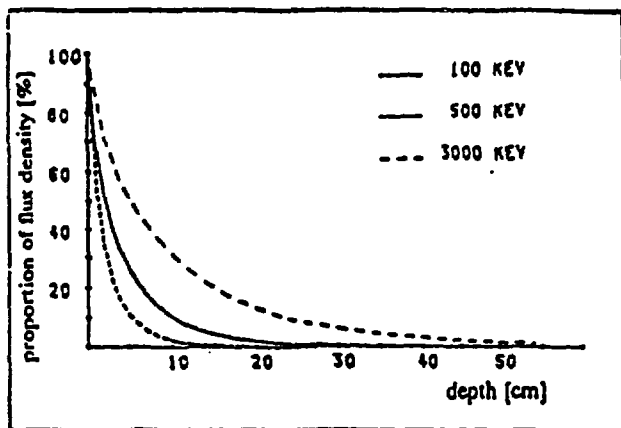


Figure 5: Gamma flux density from depths $> z$.

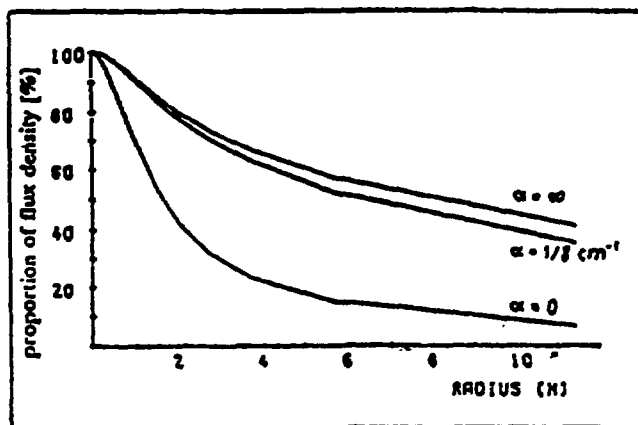


Figure 6: Rate of the gamma flux densities of nuclides from areas "outside of r ".

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