

Introduction to terrestrial gamma ray background natural & artificial nuclides present in environment

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Natural radioactivity (about 60 isotopes)

1. $T_{1/2} > 500$ mln lat (^{238}U , ^{235}U , ^{232}Th , ^{40}K , ^{147}Sm , ^{87}Rb , ^{138}La , ^{176}Lu)

Natural nuclear reactor: Oklo (Gabon)!

2. Daughters of above (^{234}U , ^{226}Ra , ^{228}Ra , ^{234}Th , ^{230}Th , ^{222}Rn , ^{220}Rn , ^{210}Po , ^{210}Pb)

{Series U, Th, Ac, (Np)}

1+2 – common name: „terrestrial radionuclides”

3. Cosmogenic (^3H , ^7Be , ^{10}Be , ^{14}C , ^{22}Na , ^{24}Na , ^{26}Al , ^{35}S , ^{36}Cl , ^{129}I , ...)

Series U

[illegible]

Are activities in series equal?

- secular equilibrium

Is parent in equilibrium with daughter?

- <NO>: (relatively low total activities)
- <YES>: whole serie equilibrated (high total activities)
- <Partial> fragments of serie equilibrated
 $^{238}\text{U} + ^{234}\text{Th} + ^{234\text{m}}\text{Pa}$, ^{234}U (so called sub-series)

Spectrum analyses needed

Series U

Z Element	124	125	126	127	128	129	130	131	132	133	134	135	136	137	138	139	140	141	142	143	144	145	146
92 uranium																			²³⁴ U 0.3 Ma, α				²³⁸ U 4.5 Ga, α
91 protactinium																				²³⁴ Pa 1 min β ⁻			
90 thorium																					²³⁴ Th 24 d β ⁻		
89 actinium																							
88 radium																							
87 fransium																							
86 radon																							
85 astatium																							
84 polonium																							
83 bismuth																							
82 lead																							
81 talium																							
80 mercury																							

LEAD SUB-S.

RADIUM SUB-S.

URANIUM SUB-S.

Series Th :

[illegible]

URANIUM (U)- isotopes

- $Z=92$
- $A=234, 235, 238, \dots$
- 0.0058%, 0.71%, 99.28% of mass
- ^{234}U $T_{1/2}=244$ ka, alpha
- ^{235}U $T_{1/2}=700$ Ma alpha, gamma
- ^{238}U $T_{1/2}=4.6$ Ga, alpha

Examples of U activities

For 1 M (238 g) U, (without Ra):

2.9 MBq alpha, + 2x2.9 MBq beta (+gamma)

1 ppm U = 12 Bq/kg (x18 if equilibrated with whole series)

In uranium glasses (green dye):

Up to 1% U = 10000 ppm U = 120 000 Bq/kg

U in environment

In rocks (as 160 minerals):

vulcanic:

basic < 1 ppm (< 12 Bq/kg)

acidic: 2.5-6 ppm (30-72 Bq/kg)

sedimental:

clay and silts 3-4 ppm (36-48 Bq/kg)

sands ~0.5 ppm (~6 Bq/kg)

carbon ~2.5 ppm (~30 Bq/kg)

In sea – (~3 ppb) ~3.3 $\mu\text{Bq}/\text{dm}^3$

Soil – 0.3-11 ppm (4-130 Bq/kg)

Human body– 10-20 ppb, mainly in kidneys (200 mBq)

Thorium (Th) - isotopes

Z= 90

^{232}Th – alpha, 14 Ga (Th ser.)

^{230}Th – alpha, 80 ka (U ser.)

^{228}Th – alpha, 1.9 a (Th ser.)

^{234}Th – beta minus, gamma, 24 d (U ser.)

^{227}Th – alpha, 18 d (Ac ser.)

^{231}Th – beta minus, 26h, (Ac ser.)

Chemical properties similar to Ce or other REE

Thorium (Th):

Abundance (mean for Earth):

2.4 ppm (1 ppm ^{232}Th = 5 Bq/kg)

Minerals

- Euxenite $(\text{Y,Ca,Ce,U,Th})(\text{Nb,Ta,Ti})_2\text{O}_6$
- Monazite $(\text{Ce,La,Th}).\text{PO}_4$
- Sartorite PbAs_2S_4
- Torianite ThO_2
- Xenotime YPO_4

Radium (Ra):

$Z = 88$

Isotopes:

^{226}Ra – alpha, gamma, 1622a (U ser.)

^{228}Ra – beta minus, 6.7 a (Th ser.)

^{224}Ra – alpha, 3.64 d (Th ser.)

^{223}Ra – alpha, 11.4 d (Ac ser.)

Second group metal (like Ca, Sr)

In general not in equilibrium with parent if it is U nor Th

Radon (Rn):

Z= 86

- Isotopes:
- ^{222}Rn – alpha, 3.84 d (U ser.)
- ^{220}Rn – alpha, 55 s (Th ser.)
- ^{219}Rn – alpha, 4s (Ac ser.)

Noble gas

Polonium (Po):

Z=84

Isotopes:

- ^{218}Po - alpha, 3.05 min (U ser.)
- ^{216}Po – alpha, 0.16 s (Th ser.)
- ^{215}Po – alpha, 1.7 ms (Ac ser.)
- ^{214}Po – alpha, 26.8 m (U ser.)
- ^{212}Po – alpha, 0.3 μs (Th ser.)
- ^{211}Po – alpha, 0.52 s (Ac ser.)
- ^{210}Po - alpha , **138 d (U ser.)**

Metal, similar to selenium

Bismuth (Bi)

Z=83

Izotopy:

- ^{214}Bi – beta minus, alfa 19.7 min, (U szer.)
- ^{210}Bi – beta minus, 5 d (U szer.)
- ^{212}Bi – beta minus, alfa, 60.5 m (Th szer.)
- ^{211}Bi – alfa, beta minus, 2.16 min (Ac szer.)
- ^{209}Bi – stabilny, nieradiogeniczny

Ciężki, toksyczny metal z grupy
azotanowców – gromadzi się w kościach

Lead (Pb):

Z=82

Isotopes:

- ^{214}Pb – beta minus, gamma, 26.8 min (U szer.)
- ^{210}Pb – beta minus, gamma, 22 a (U ser.)
- ^{206}Pb – stable (U ser.)
- ^{212}Pb – beta minus, gamma, 10.6 h (Th ser.)
- ^{208}Pb – stable (Th ser.)
- ^{211}Pb – beta minus, gamma, 36 min (Ac ser.)
- ^{207}Pb – stable (Ac ser.)

Toxic heavy metal

Potassium (K) & rubidium (Rb):

^{40}K (1.26 Ga), beta minus 89%, EC 11%
Abundance 0.0117% (32 Bq/g potassium)

^{87}Rb (48 Ga), beta minus,
Abundance 27%, ~1 kBq/g Rb

First group metal, K –important macro-nutrient, Rb toxic, in traces.

REE

^{138}La (112 Ga), beta minus+gamma

^{147}Sm (105 Ga), alpha

+ ^{144}Nd , ^{142}Ce , ^{148}Sm , ^{146}Sm , ^{152}Gd , ^{156}Dy ,

....

Cosmogenic radionuclides (main)

IZOTOP	$T_{1/2}$	Radiation E [MeV]	AKTYWNOŚĆ [Bq/m ³]
³ H	12 a	β^- , (0.0186)	$3.5 \cdot 10^{-4}$
⁷ Be	54 d	EC, γ (0.477)	0.003
¹⁰ Be	1.6 Ma	β^- (0.555)	$1.2 \cdot 10^{-9}$
¹⁴ C	5.7 ka	β^- (0.156)	0.04
²² Na	2.6 a	β^+ (0.545), γ (1.275)	$3 \cdot 10^{-7}$
²⁴ Na	15 h	β^- (1.4), γ (1.37, 2.75)	-
²⁶ Al	7.2 Ma	β^+ (1.2), γ (1809)	-
³² P	14 d	β^- (1.71)	$7 \cdot 10^{-5}$
³³ P	24 d	β^- (0.246)	$4 \cdot 10^{-5}$
³⁵ S	88 d	β^- (0.167)	$4 \cdot 10^{-5}$
³⁸ S	2.84 h	β^- (1.1), γ (1.88)	-
³⁶ Cl	0.3 Ma	β^- (0.714)	$2.5 \cdot 10^{-10}$
³⁸ Cl	37 min	β^- (4.91), γ (1.60, 2.17)	-
³⁹ Cl	55 min	β^- (1.91), γ (0.25, 1.27)	-
...			

dla beta - E max.

Production rate in atmosphere

Depends on

- Altitude – about 2/3 on stratosphere
- Geomagnetic latitude

IZOTOP	Produkcija (cm ² s) ⁻¹ Troposfera	Whole atmosphere
³ H	8.4·10 ⁻²	2.5·10 ⁻¹
⁷ Be	2.7·10 ⁻²	8.1·10 ⁻²
¹⁰ Be	1.5·10 ⁻²	4.6·10 ⁻²
¹⁴ C	1.1	2.5
²² Na	2.4·10 ⁻⁵	8.6·10 ⁻⁵
²⁶ Al	3.8·10 ⁻⁵	1.4·10 ⁻⁴
³² P	2.7·10 ⁻⁴	8.1·10 ⁻⁴
³³ P	2.2·10 ⁻⁴	6.8·10 ⁻⁴
³⁵ S	4.9·10 ⁻⁴	1.4·10 ⁻³
³⁶ Cl	4.0·10 ⁻⁴	1.1·10 ⁻³

Production rate *in situ*

„*in situ*” (on Earth’s crust {rocks, waters, soil} surface) –

Cosmogenic Isotopes are
Produced In Situ by the
Interaction of Cosmic Rays with
a Variety of Target Atoms



Variation of production rate

- Sun activity

Revers mode (antiproportional)

- Size of Earth's Magnetosphere

Revers mode (antiproportional)

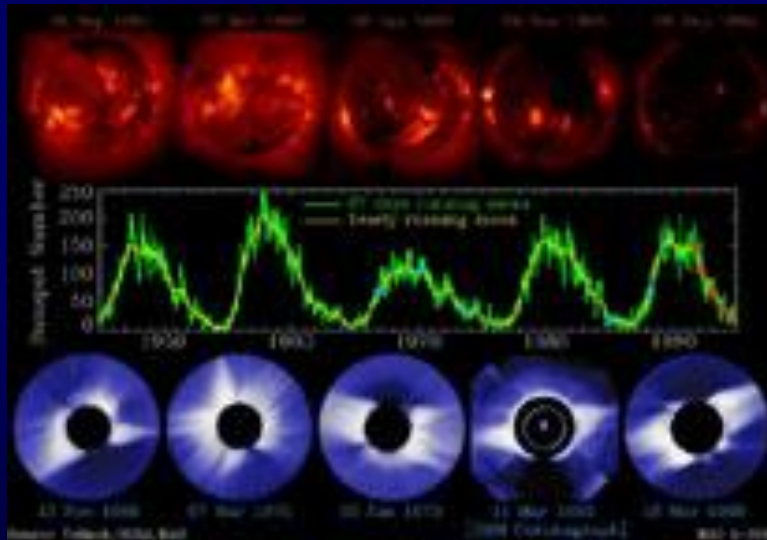
- Cosmic rays flux

Proportional

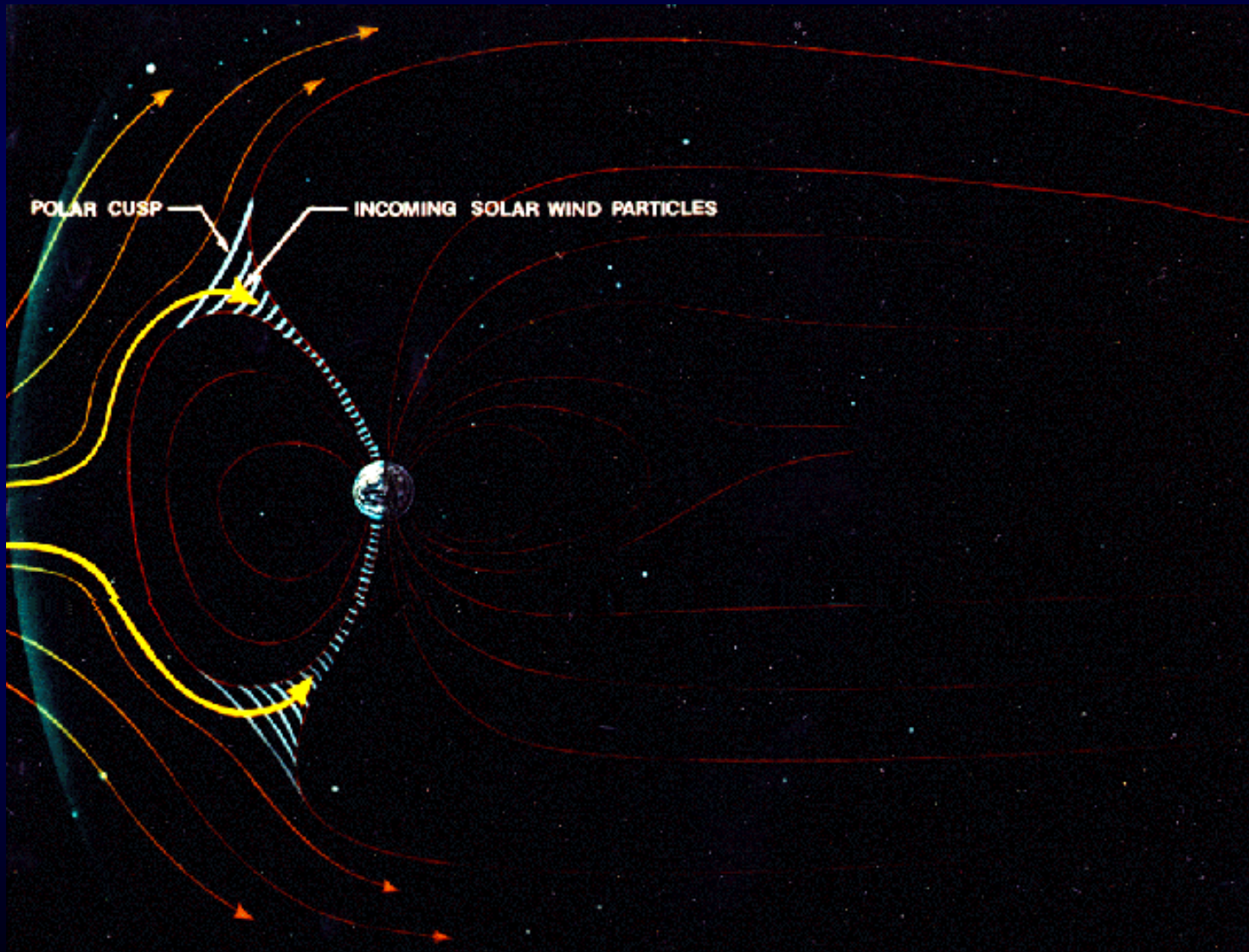
Sun activity

A set of phenomena (sunspots, solar wind flux, ...)

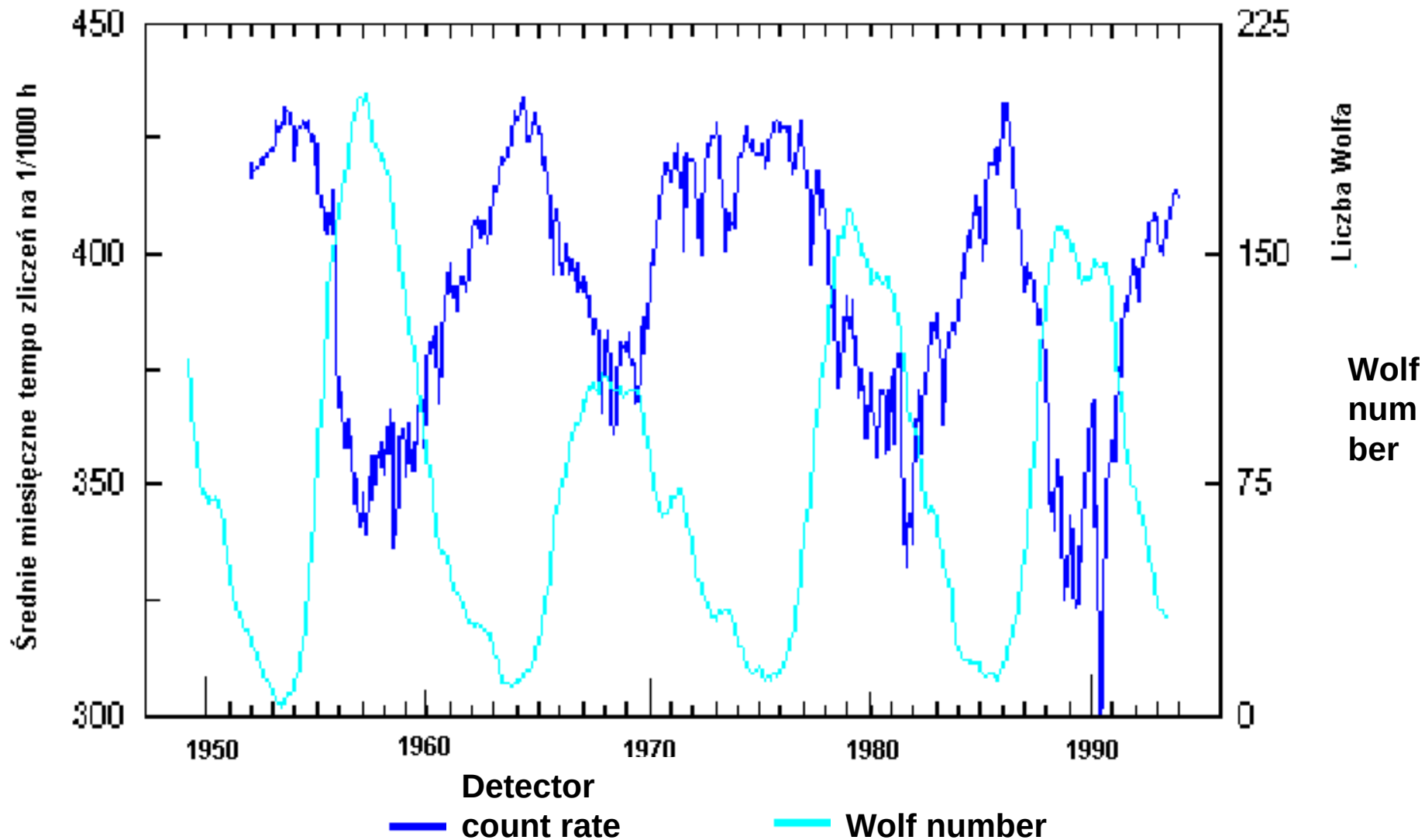
Main cycle - 11 years period



Earth magnetosphere and solar wind



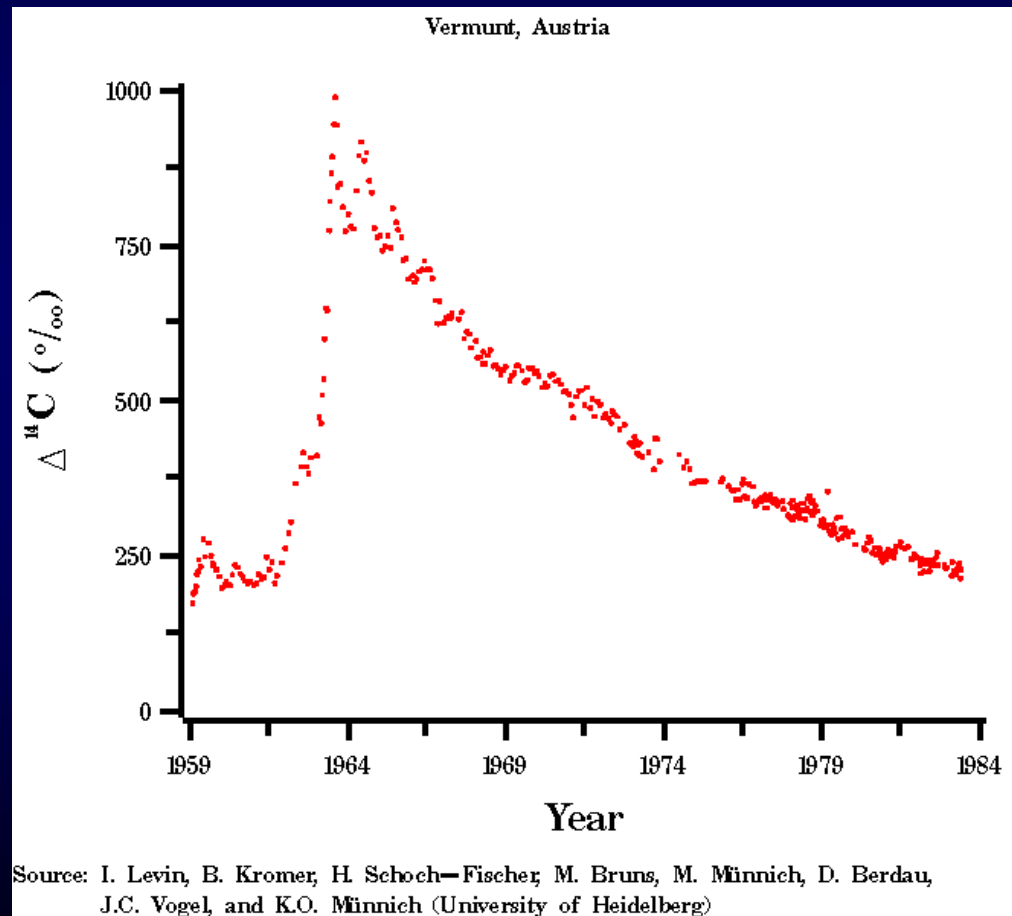
Cosmic radiation & Sun activity



Man made cosmogenic radionuclides: ^3H , ^{14}C & ^{85}Kr (year 2000)

Nuclide	$T_{1/2}$ [years]	Natural PBq	Global fallout PBq	Nuclear Energy PBq	Fuel Reprocess. PBq
^3H	12.33	1275	22 000	150	150
^{14}C	5730	12 750	213	-	-
^{85}Kr	10.76	0.09	~17	10	4800

Excess of C14 due to nuclear explosion



Typical environmental activities

Isotopes	Material	Activity [Bq]
^{14}C	Man, body	~2000
^{40}K	Man, body	~3000
^{40}K	Milk , 1 L	40
^{40}K	Soil, 1 kg	100-1000
^{238}U	Soil, 1 kg	30-100
^{226}Ra	Soil, 1 kg	30-100
^7Be	Air, m ³	~10 ⁻³
^{222}Rn	Air, m ³	Od <1 do 10 ⁶
^{22}Na	Air, m ³	~10 ⁻⁶

Man-made radionuclides in environment

MAIN

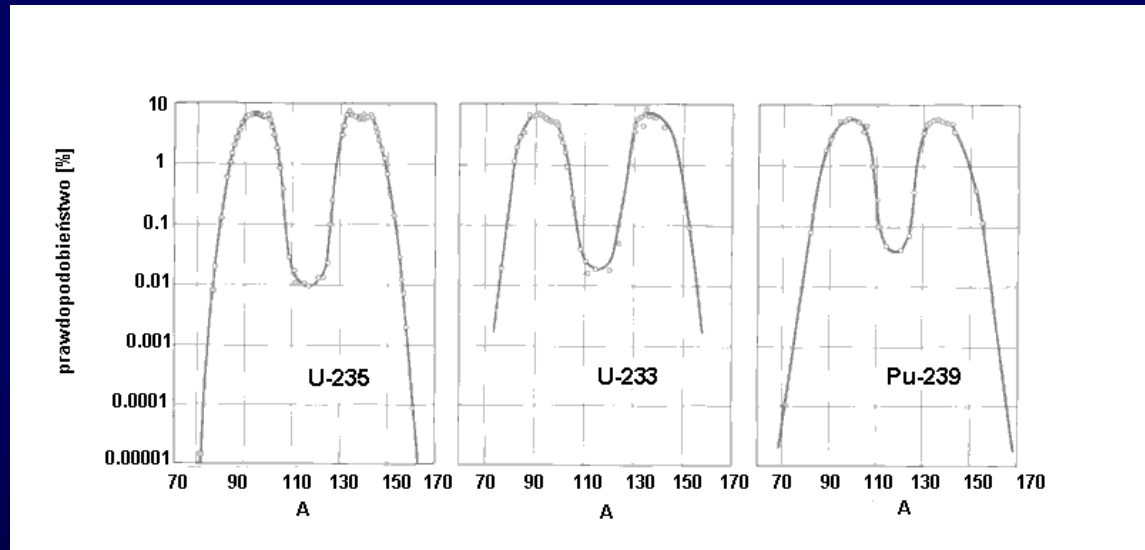
- **Whole cycle of nuclear fuel production and reprocessing – including accidents**
- **Nuclear weapons – production and use**

SECONDARY

- Nuclear medicine
- Industry
- Laboratories

Physical processes in which radionuclides are produced

1. Nuclear fission (NPP, bomb A): >100 isot.: ^{137}Cs , ^{90}Sr ,...



2. Activation (by **neutrons**, protons, other) ^{60}Co , ^{63}Ni , ^{134}Cs ...

3. Nuclear fusion (thermonuclear explosion) ^{14}C , ^3H

Man-made radionulides

long-lived (years):

^{85}Kr , ^{137}Cs , ^{134}Cs , ^{99}Tc , $^{238,239,240,241}\text{Pu}$, ^{241}Am ,
 ^{90}Sr(+many others)

Intermediate: (weeks): ^{103}Ru , ^{95}Nb , ^{95}Zr , ^{89}Sr ,...

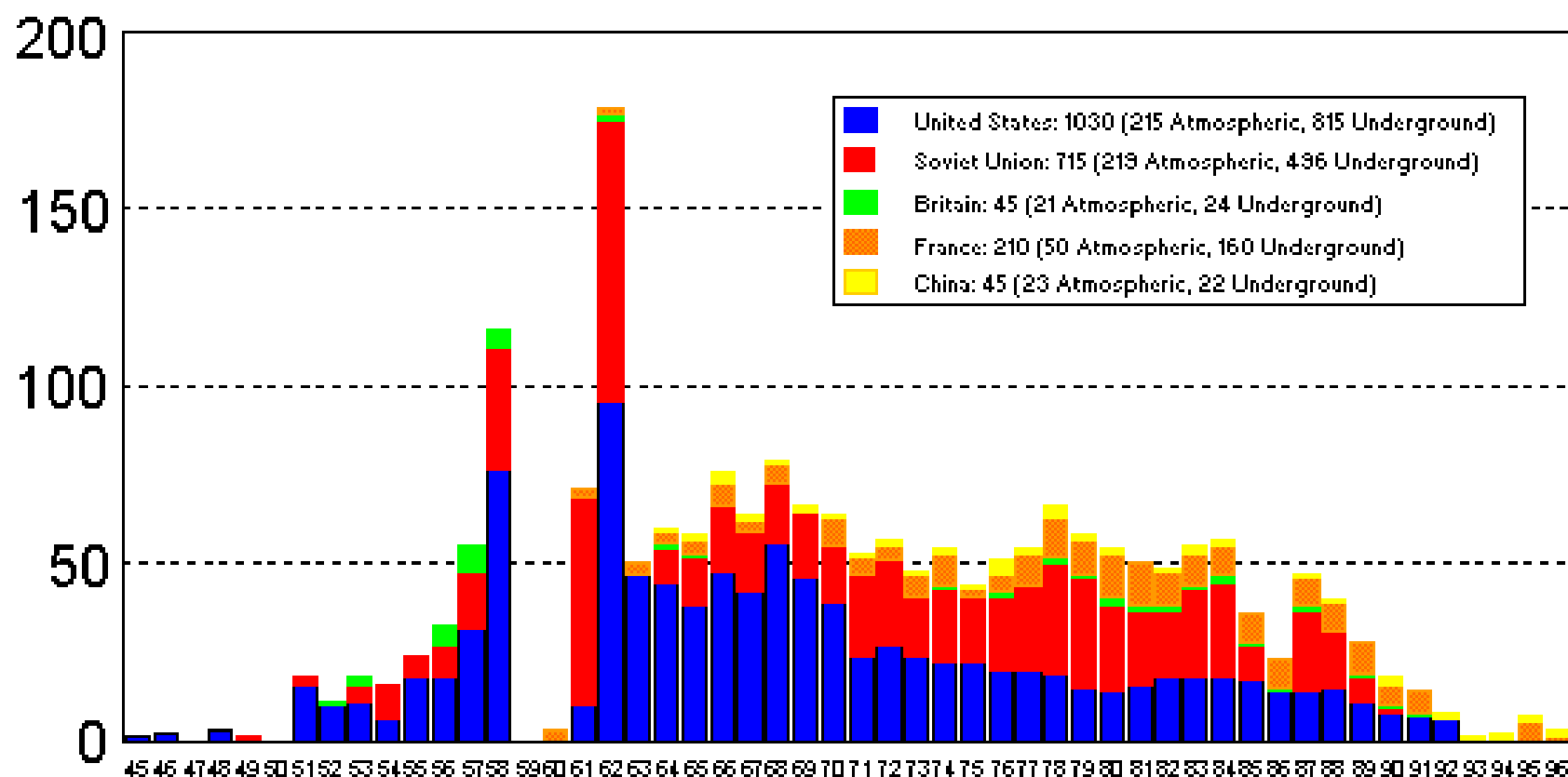
Short lived (days): ^{131}I , ^{132}I ,

Main events (Poland)

- Global fallout (1945...-1957- 1963-...1980)
- Chernobyl (1986)

Global Nuclear Weapons Tests

1945-1996



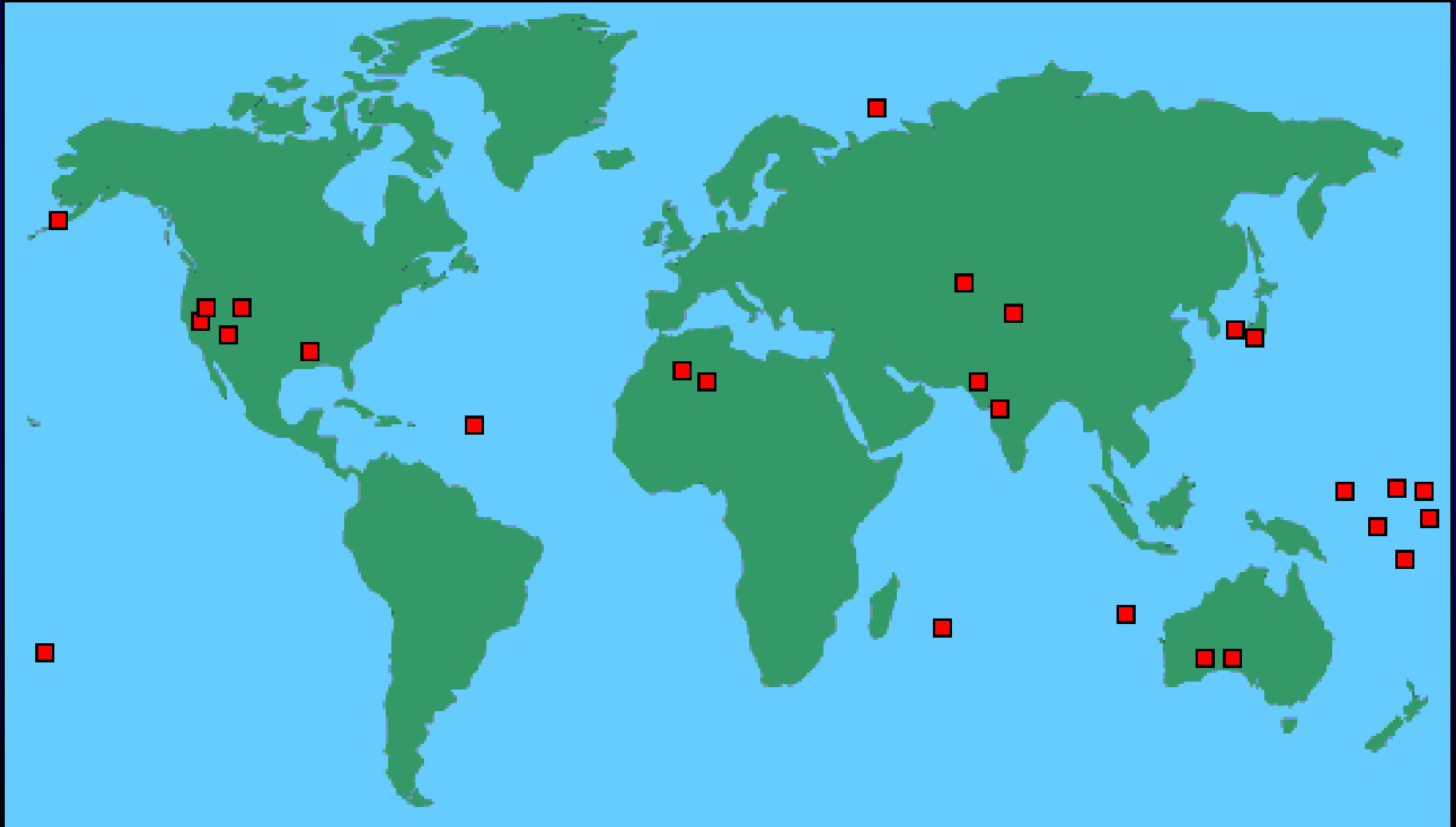
U.S. total does not include the two atomic bombs dropped on Hiroshima and Nagasaki in August 1945.

The U.S. and the Soviet Union conducted 27 and 124 "Peaceful Nuclear Explosions," respectively, which are included in the above totals.

India conducted an underground nuclear test in 1974.

Sources: U.S. Department of Energy; Natural Resources Defense Council, Nuclear Weapons Databook Project

Atmospheric nuclear explosions

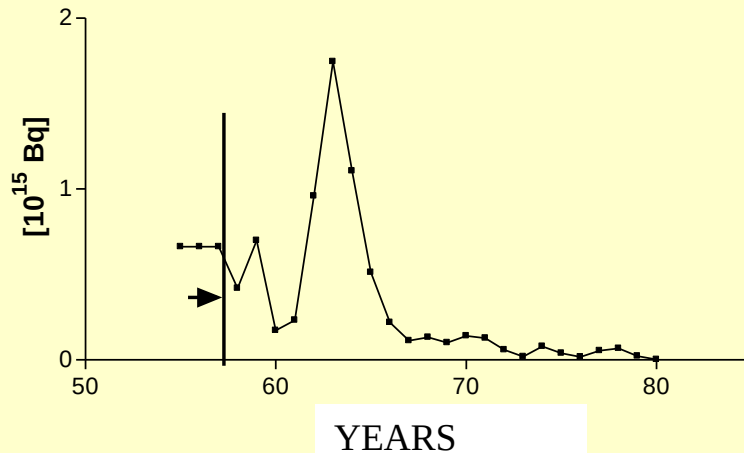


Releases and cummulated deposition - decay corrected

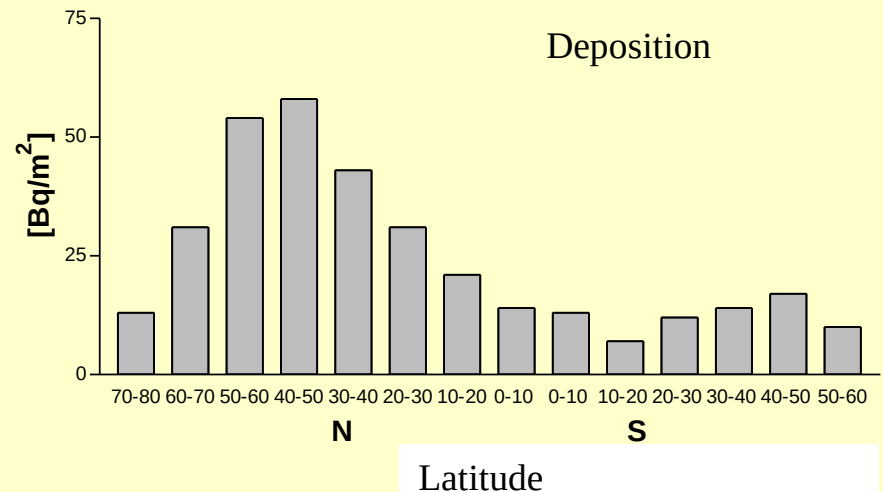
Radionuclid	T _{1/2}	Releases PBq	Deposition till 1963 Bq/m ²	Deposition till 1999 Bq/m ²
⁸⁹ Sr	50.5 d	117000	604	0
⁹⁰ Sr	28.78 a	622	1040	825
⁹⁵ Zr	64 d	148000	1770	0
¹⁰⁶ Ru	373.6 d	12200	4780	0.0004
¹²⁵ Sb	2.76 a	741	602	0.46
¹³¹ I	8.02 d	675000	9.11	0
¹³⁷ Cs	30.07 a	948	1560	1300
¹⁴⁴ Ce	284.9 d	30700	9290	0
²³⁹ Pu	24110 a	6.52	24.5 (NH)	24.5 (NH)
²⁴⁰ Pu	6563 a	4.35	16.3 (NH)	16.3 (NH)
²⁴¹ Pu	14.35a	142	549 (NH)	88 (NH)

Example – global fallout of $^{239+240}\text{Pu}$

Przebieg depozycji Pu (GF)
0-80 N
[UNSCEAR 82]



Depozycja Pu [UNSCEAR 77]
(GF)



SNAP 9A (USA)

(Navigation satellite Transit 5-BN-1)

- 1 kg ^{238}Pu – 0.629 PBq

Launched: 28.9.1963,

Re-entry: 24.04.1964 over Indian Ocean near Madagascar,
burned at ~50 km (stratosphere!)

Cosmos 954 (USSR)

Accidental re-entry and burn of nuclear reactor
(January 1978)

Northwest Territory of Canada

^{131}I – 181 TBq

^{90}Sr – 3 TBq

^{137}Cs - 3 TBq

Global fallout for Central Europe

Deposition (kBq/m²)

Cs-137: 5

Sr-90: 3

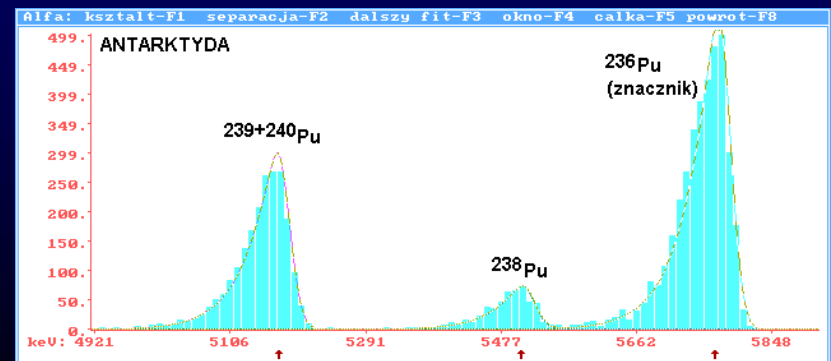
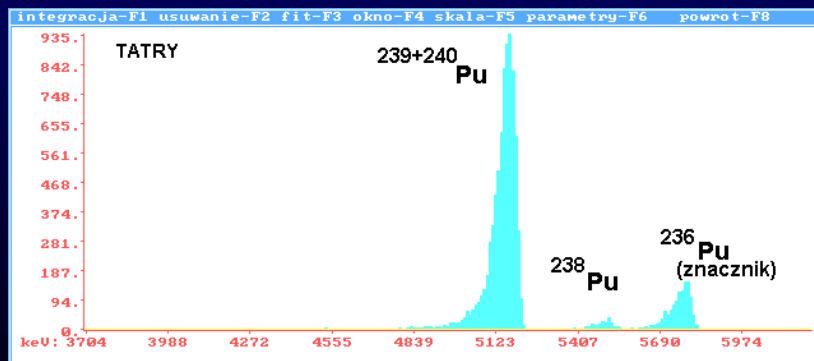
(activities for 1963)

Pu-239 i 240: 0.06

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Satellite SNAP 9A, '64 (1 kg ²³⁸Pu)

Pu-238: 0.002



Chernobyl fallout

POLAND

26 April 1986
~24hour emission moved
over Poland toward Europe
Total 10 day of release
~7 EBq

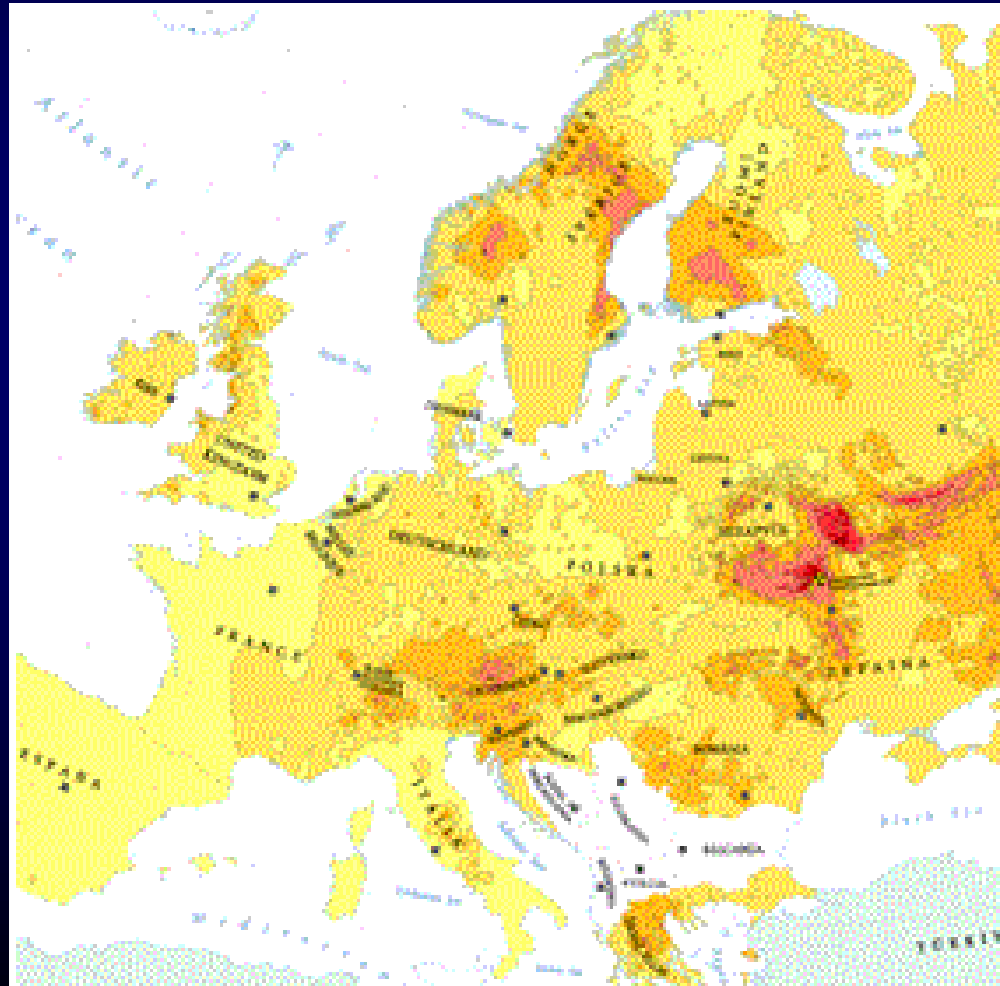
Radioactive plum
(for release in date given in box)



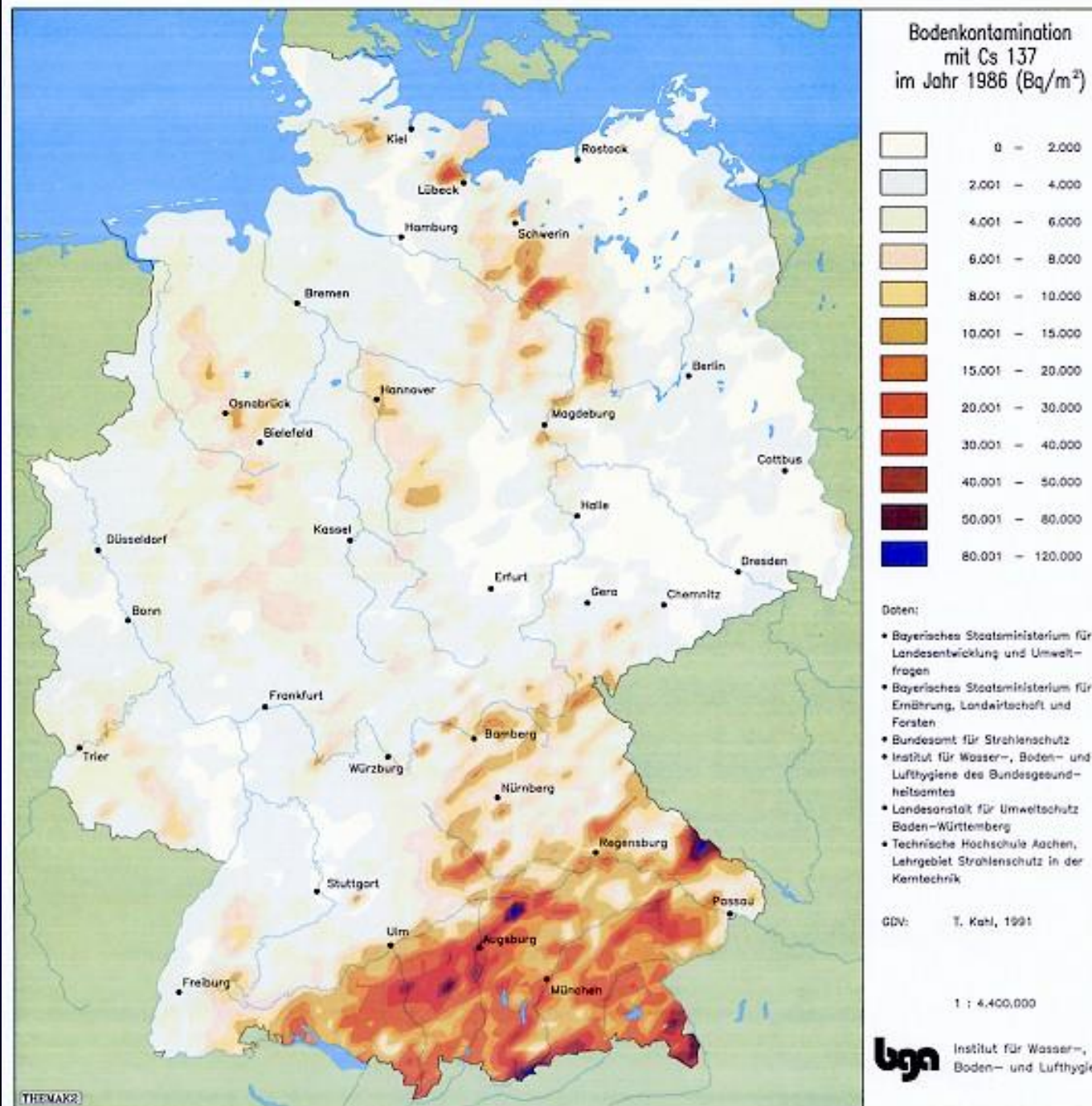
Cs-137 nearby NPP (IAEA 1991)



Cs-137 contamination of Europe



Cs-137 contamination of Germany



Chernobyl fallout

„continuous fraction” ($\sim 1\mu\text{m}$),
(kBq/m²):

Long lived isotopes:

Cs-137 - mean 5 (max. ~ 100)

Sr-90 - $1/50 \times \text{Cs-137}$

$^{239+240}\text{Pu}$ $10^{-6} \times \text{Cs-137}$

+several short or intermediate T_{1/2} (I, Te, Ru, Ba, La, Ce, Sb, Ag...)

For comparison Global Fallout (UNSCEAR data for Polish latitude):

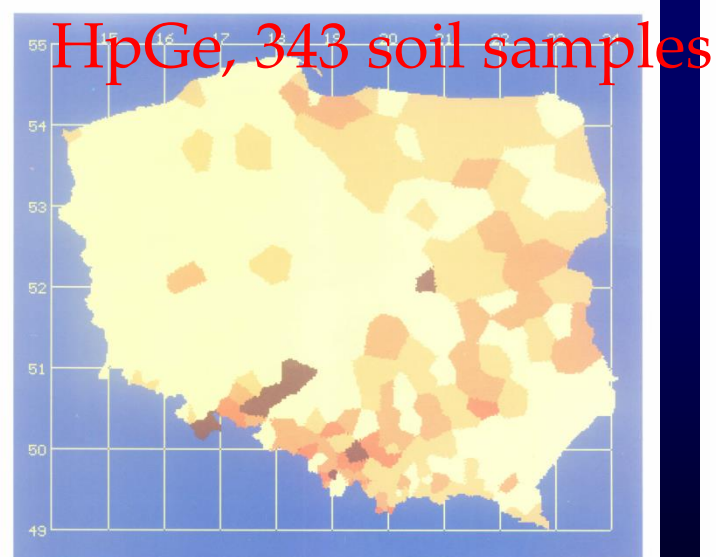
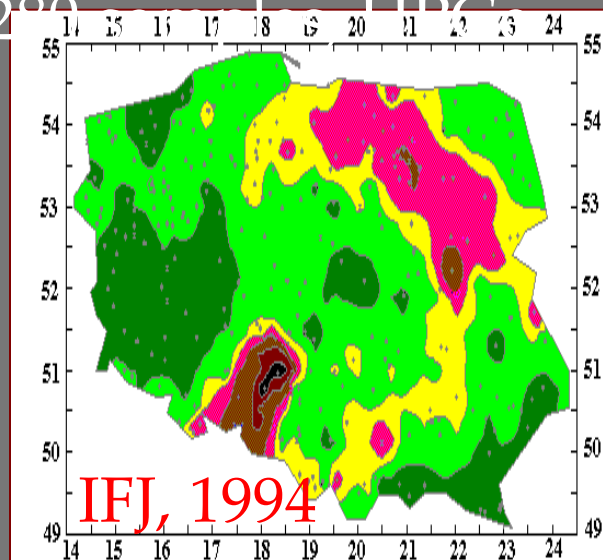
1963, kBq/m ² : Cs-137	5
	Sr-90

MUSHROOM Xerocomus badius

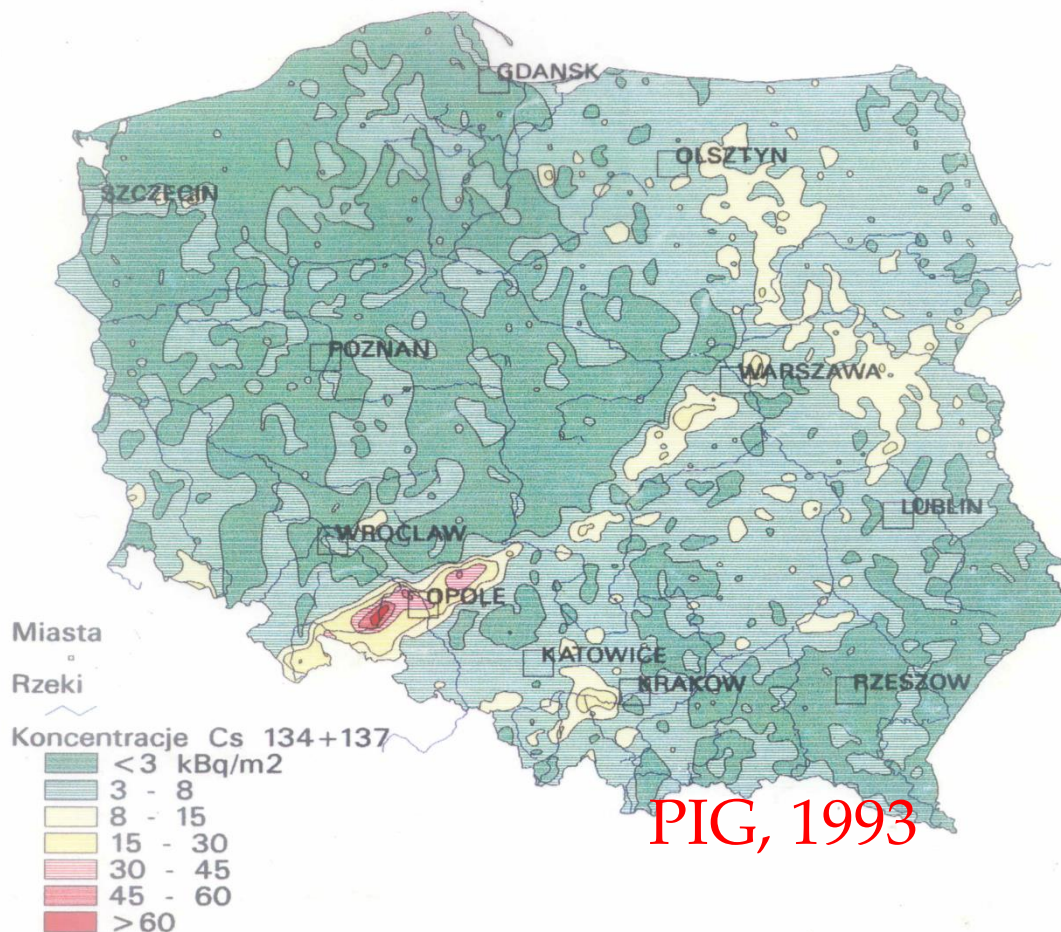
2001 IJC

Chernobyl fallout

Approximate maps for ^{137}Cs in Poland



In situ NaI spect



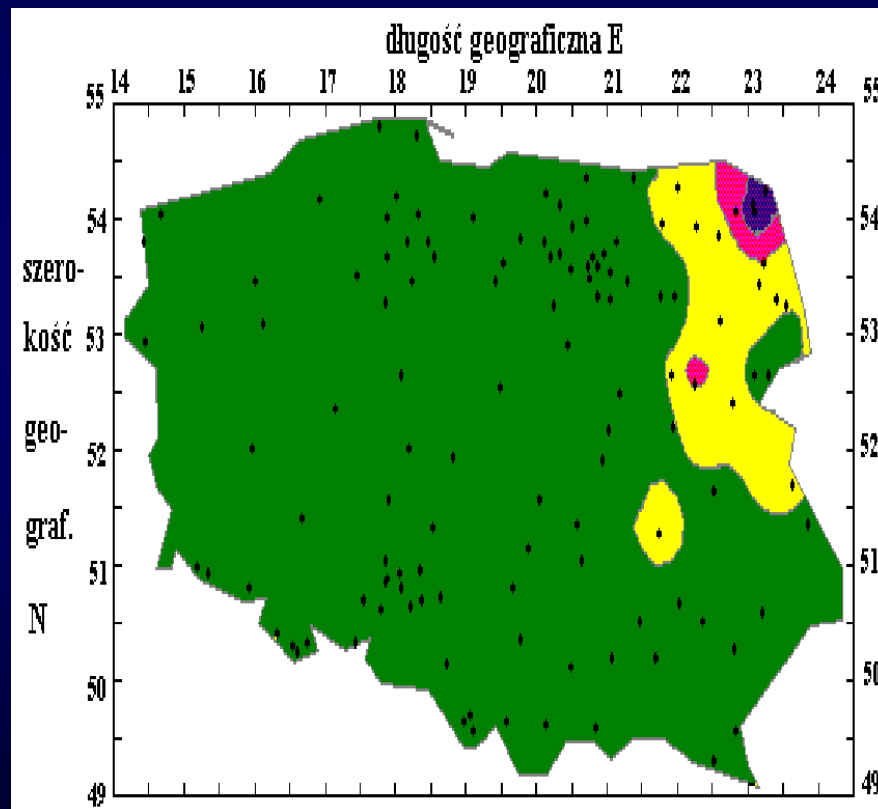
„Hot particles” (in Poland- 5-10 μm)

- „Fuel-like” UO_2 or U_3O_8 matrix
 $^{238,239,240,241}\text{Pu}$, ^{241}Am , $^{243,244}\text{Cm}$, ^{90}Sr , ^{154}Eu ,
($^{141,144}\text{Ce}$, ^{95}Zr , ^{95}Nb , ^{155}Eu , ^{89}Sr , ^{242}Cm ...)
- „condensation” or „monoelemental”) -
condensation of vapours or „white inclusions”? –

Matrix of Ru, Mo, ...Fe, Ni...: ($^{103,106}\text{Ru}$...) ^{63}Ni ,
 ^{60}Co ^{99}Tc ?

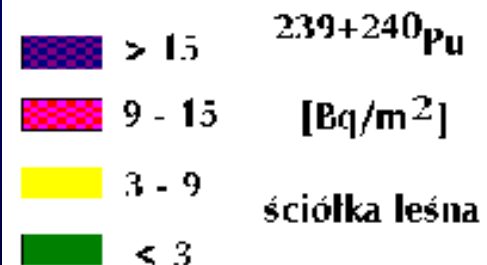
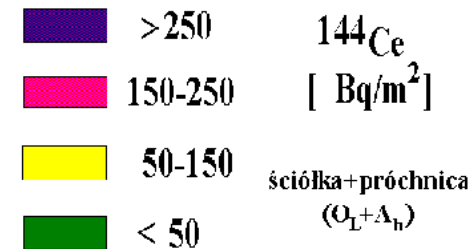
Approximate geographical distribution of HP in Poland

Mietelski, IFJ, 1994



Activity for 1 st Sep. 1991

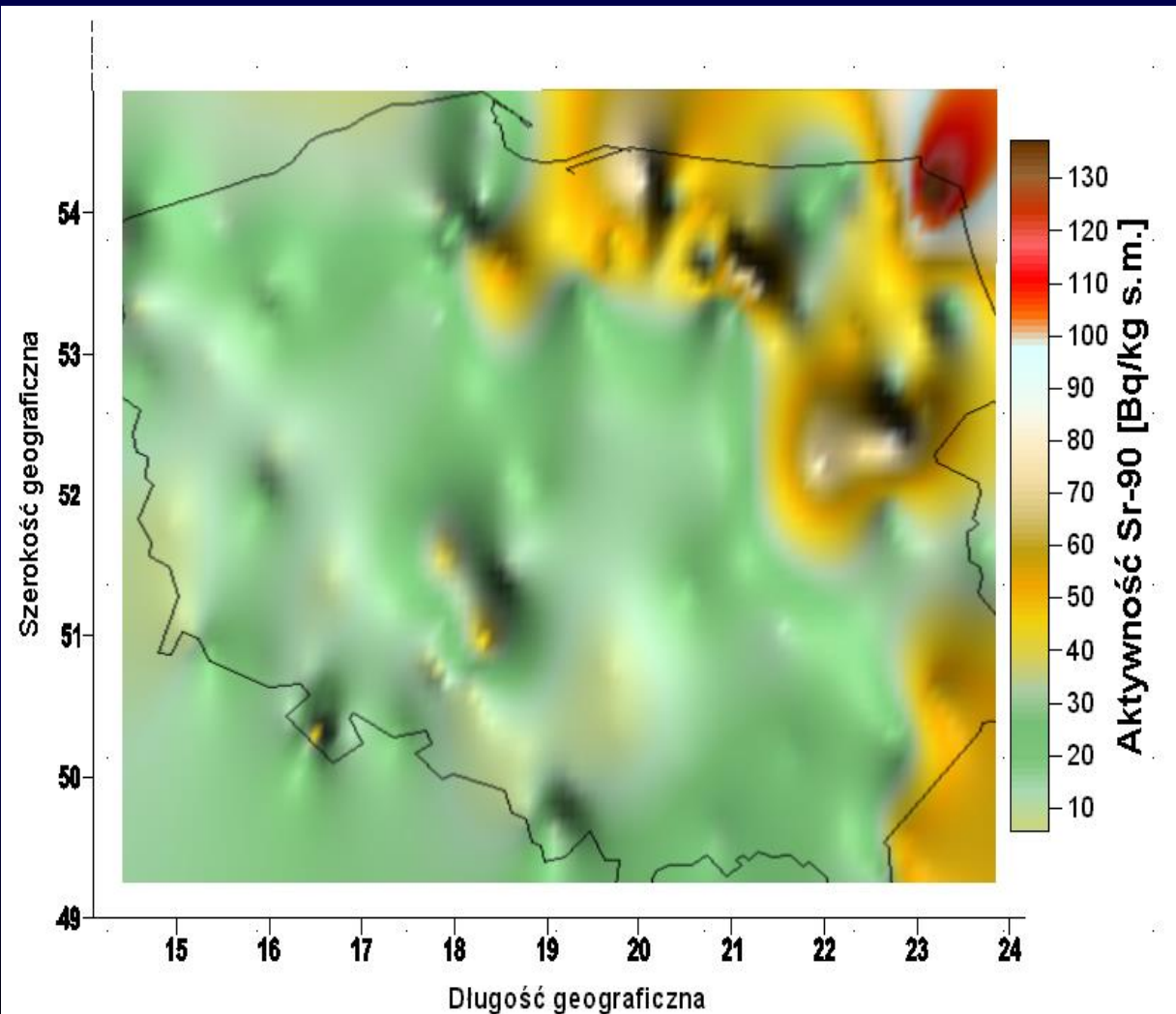
(Ce-144 x 110 for 1986)



^{90}Sr (bilberry leaf)

(P.Gaca PhD, IFJ-UG , 2004)

~ 2 kBq/m² in NE



Conclusions

- Natural and artificial radionuclides are present all over globe
- Large variety can be observed
- Local background should be recognized