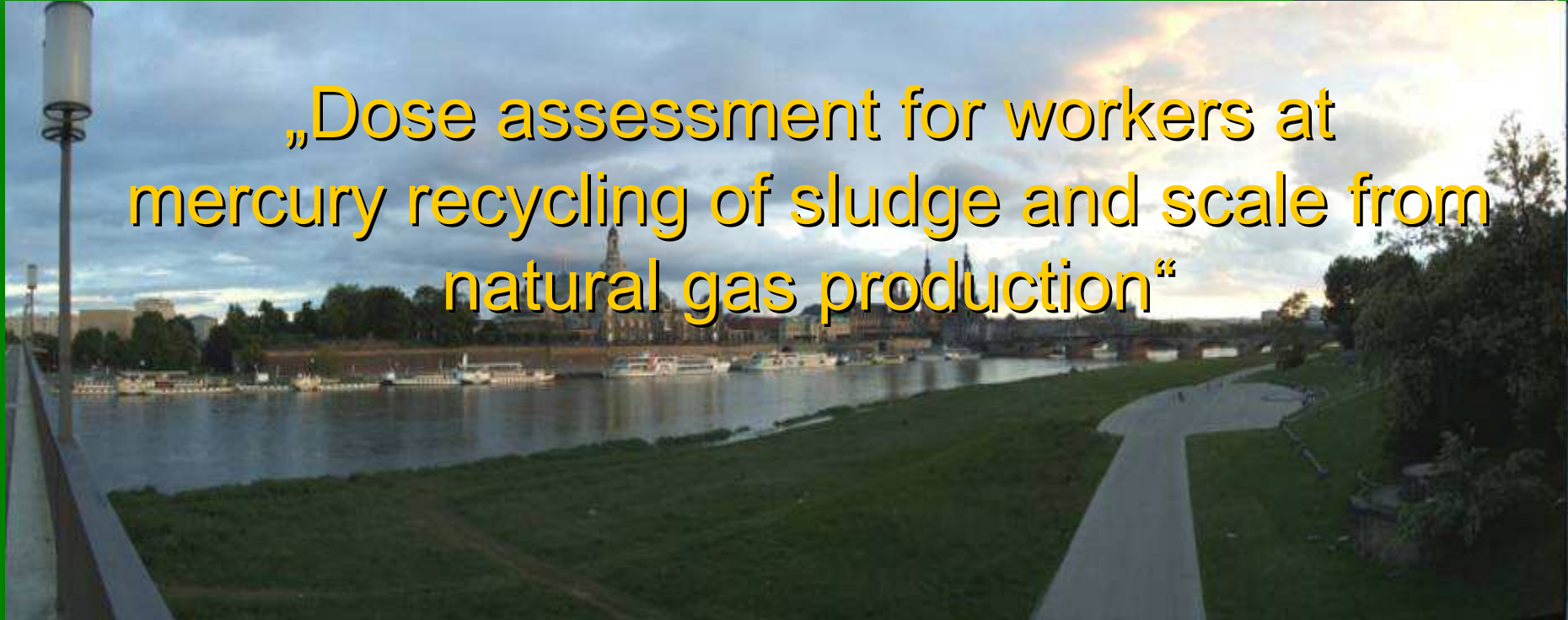


IAF - RADIOÖKOLOGIE GMBH

Labor für Radionuklidanalytik Radioökologische Gutachten Consulting



„Dose assessment for workers at mercury recycling of sludge and scale from natural gas production“



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Dose assessment for workers at mercury recycling of sludge and scale from natural gas production

- **About Scale and Sludge**
- **Radiological measurements and dose assessment at the recycling of mercury and the immobilisation of the residues**
- **Radiological measurements and dose assessment at the disposal of immobilised residues at a landfill**



What is the topic

NORM: Scale and Sludge containing Mercury



Transport



Recycling of Mercury



landfill

radioactive immobilisates

Mercury

Marketing



Typical scale in tubings



Scale thickness up to 5 mm



- Scale mass 4 to 10 kg per 10 m tubing
- 2 to 5 tons in 5 km length of pipeline



• 0 - 8 %mass percent mercury





Mercury drops in a typical sludge



- 8 - 15 %mass percent mercury
- but also up to 30% possible

- The mercury drops contain always Pb-210
- typical values: 5 - 10 Bq/g (/gram mercury)



5 Relevant radionuclides in the residues (sludge and scale)

U-238 decay series: Ra-226, Pb-210, Po-210

Th-232 decay series: Ra-228, Th-228

U-238 < 1 Bq/g

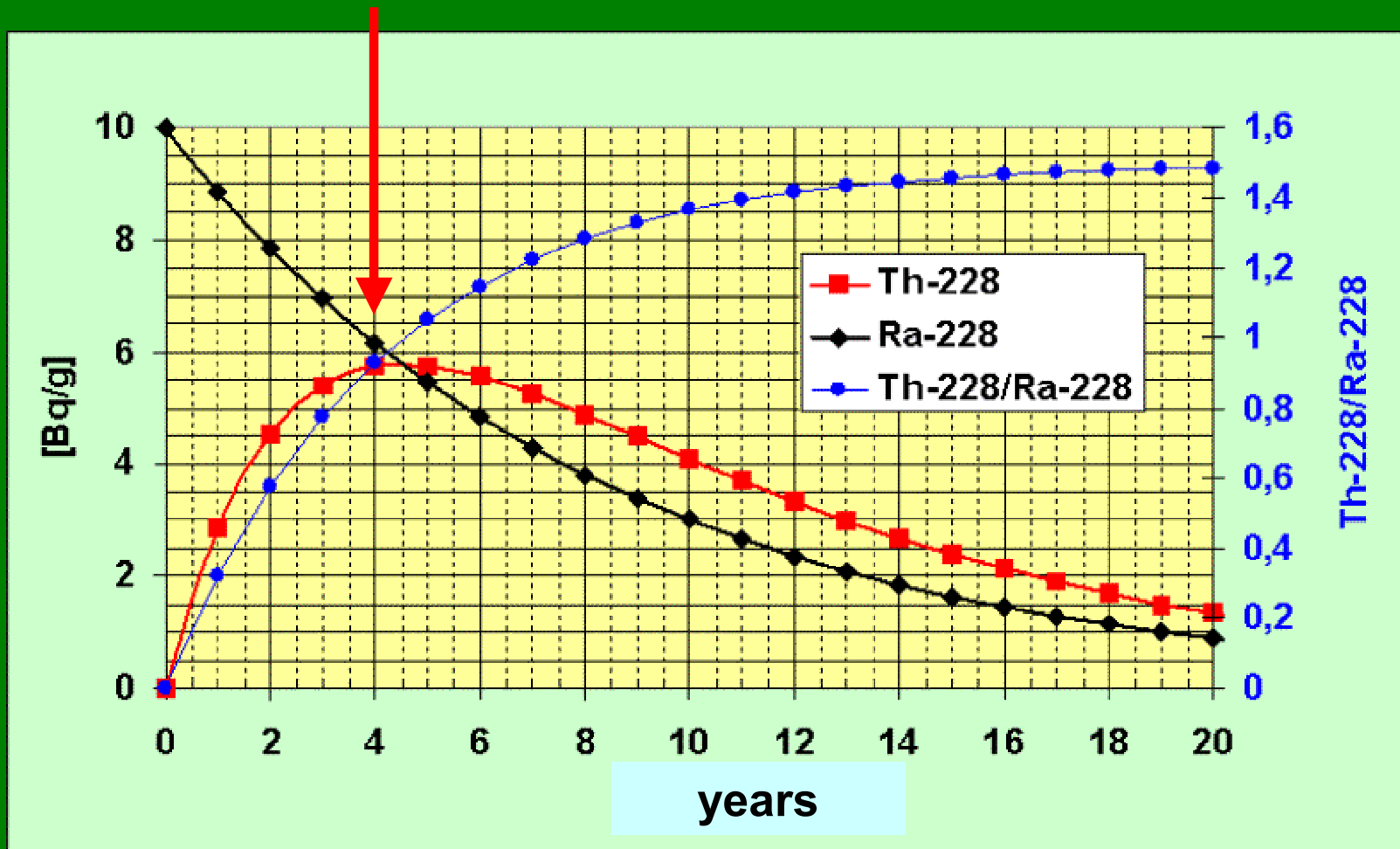
Th-232 < 1 Bq/g

**Practically
of minor importance**

**After 30 years practically no Ra-228 and Th-228
in the residues!**

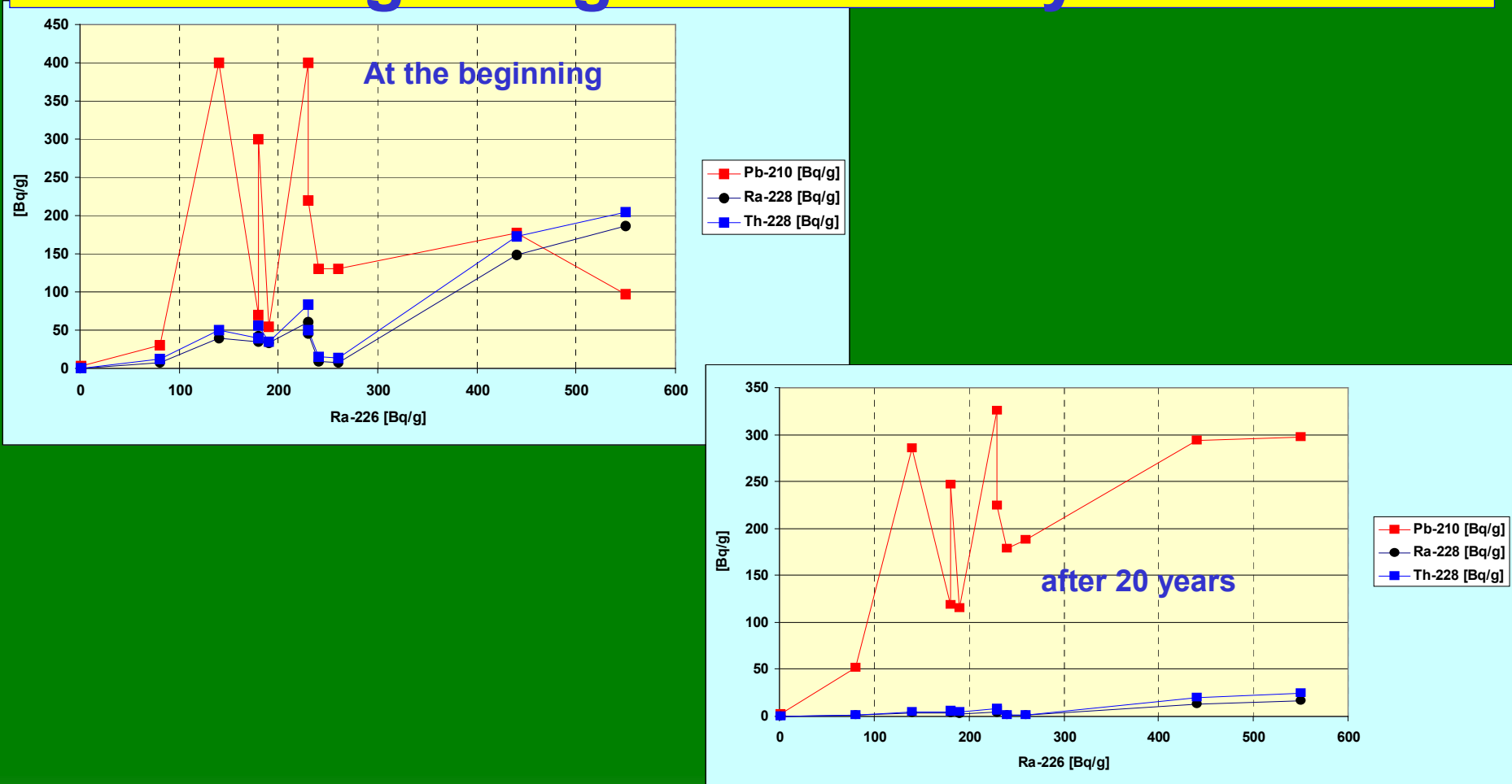


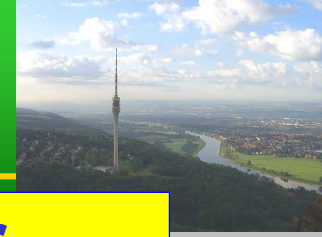
$A(\text{Ra-228}) \approx A(\text{Th-228})$ after 4 years



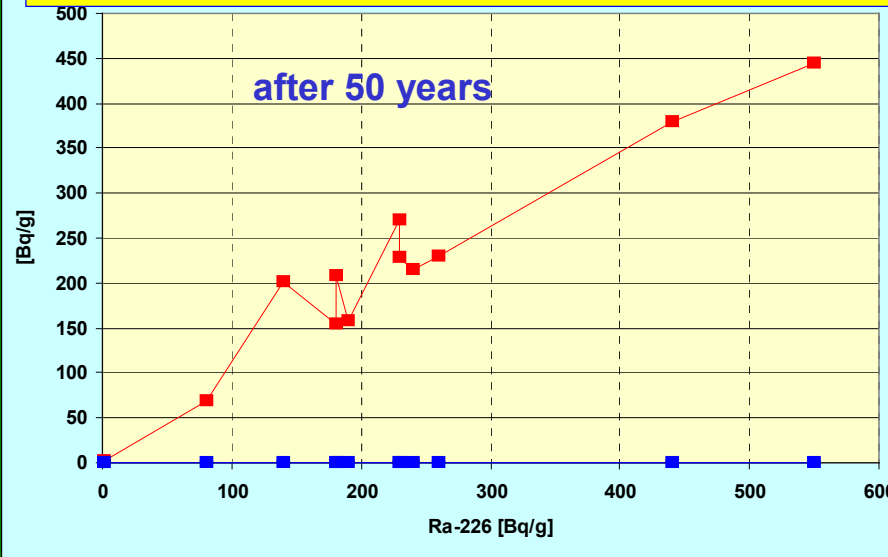


Typical radionuclide compositions at the beginning and after 20 years

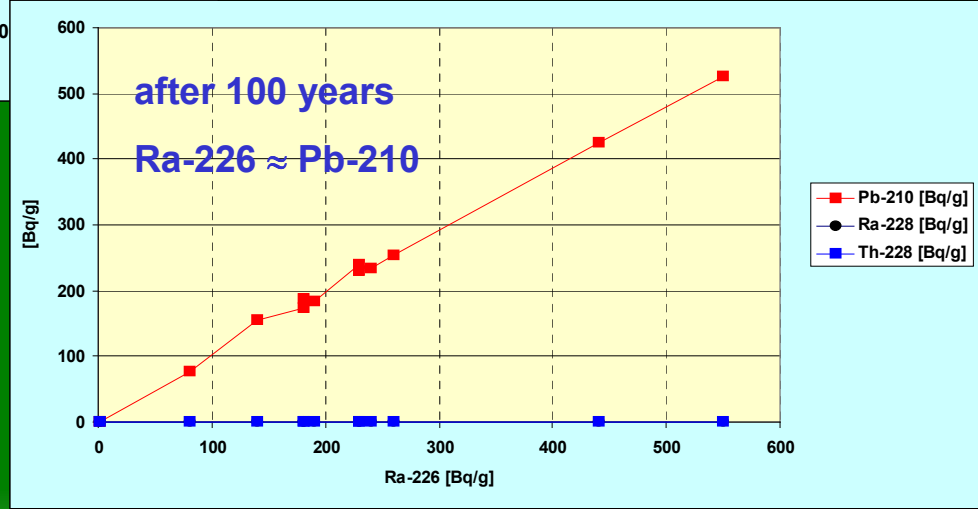




Radionuclide compositions after 50 and 100 years



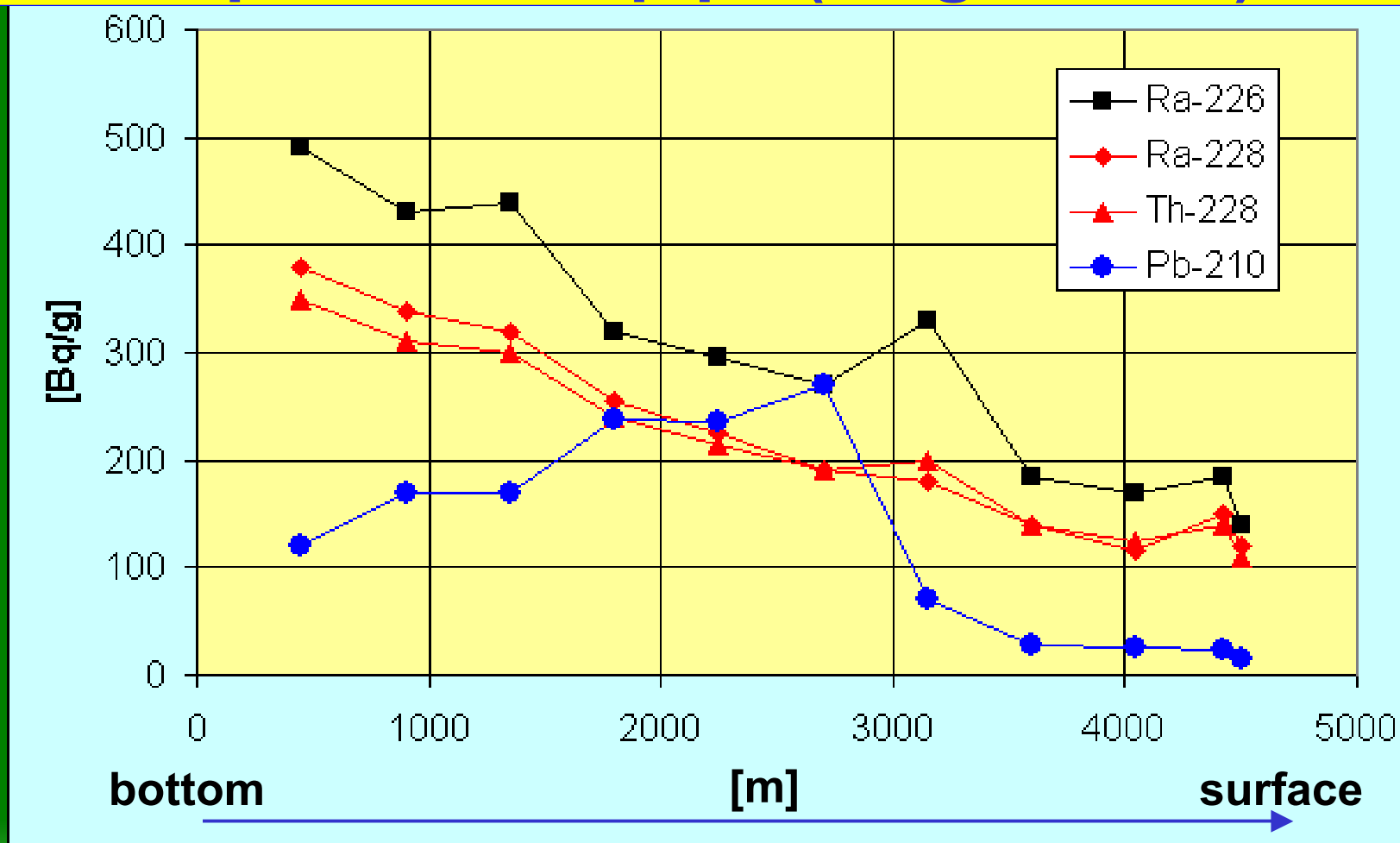
- Pb-210 [Bq/g]
- Ra-228 [Bq/g]
- Th-228 [Bq/g]



- Pb-210 [Bq/g]
- Ra-228 [Bq/g]
- Th-228 [Bq/g]

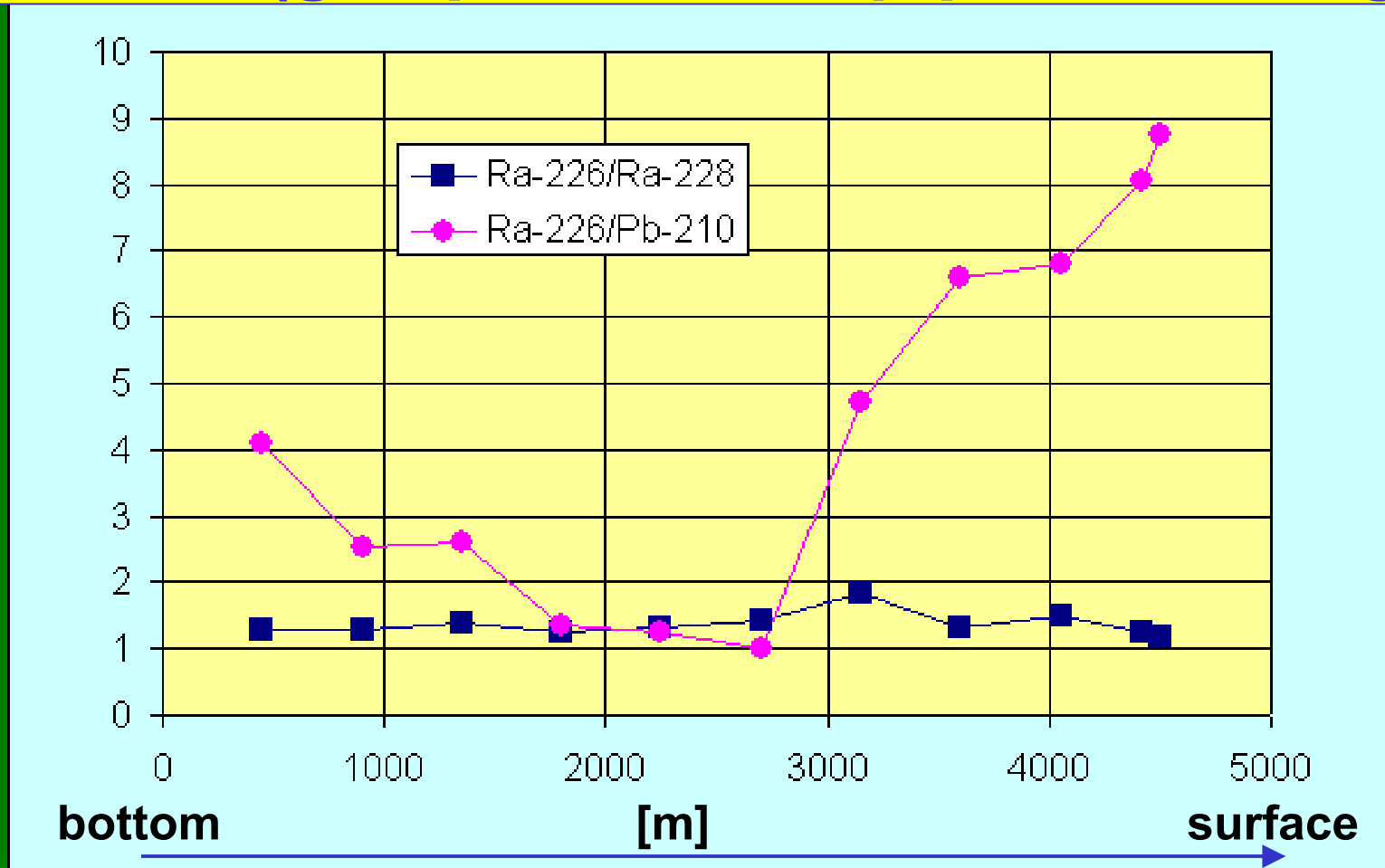


Distribution of Radionuclides in a gas production pipe (length 5 km)



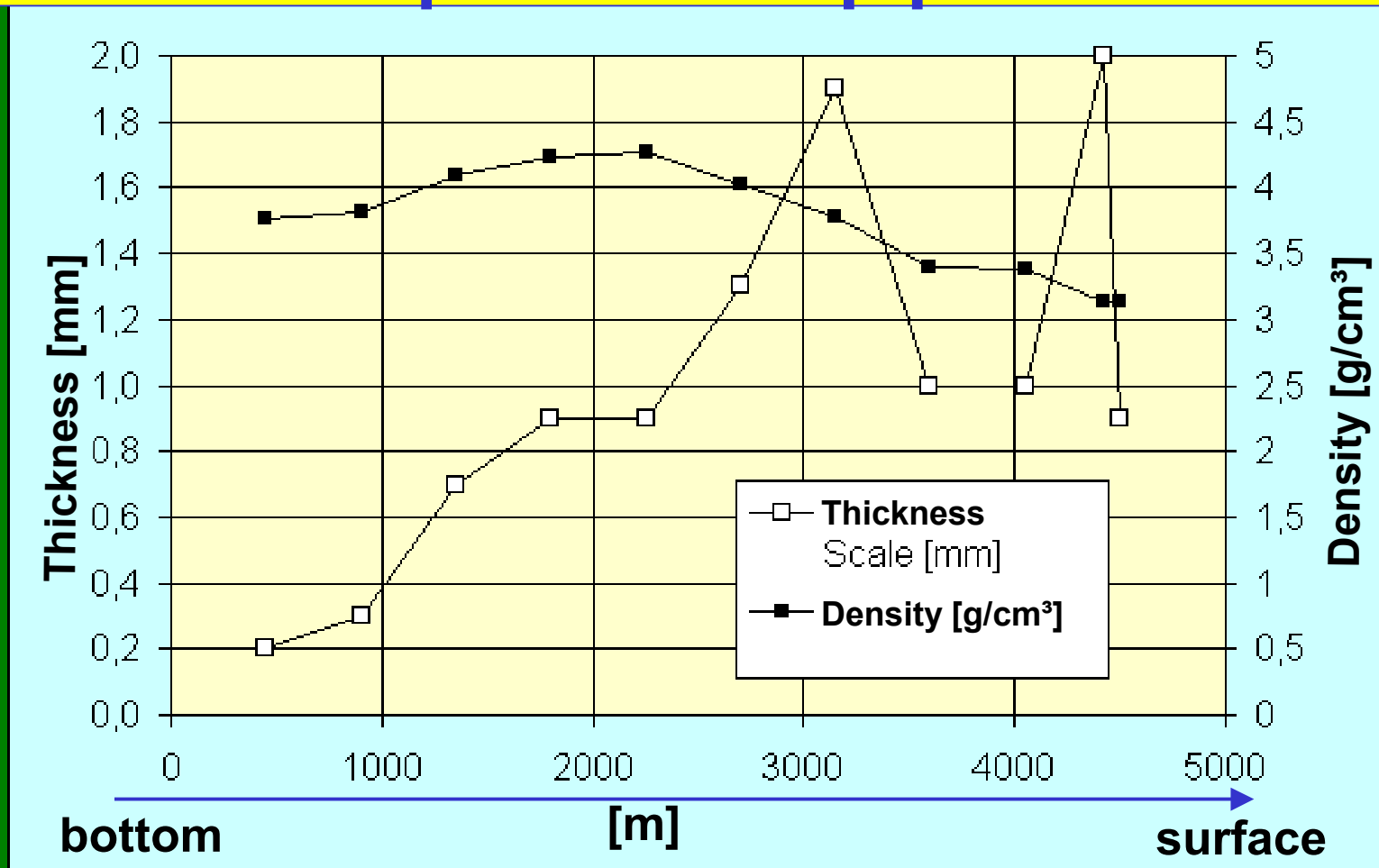


Activity ratios from the bottom to the surface (gas production pipe 5 km length)



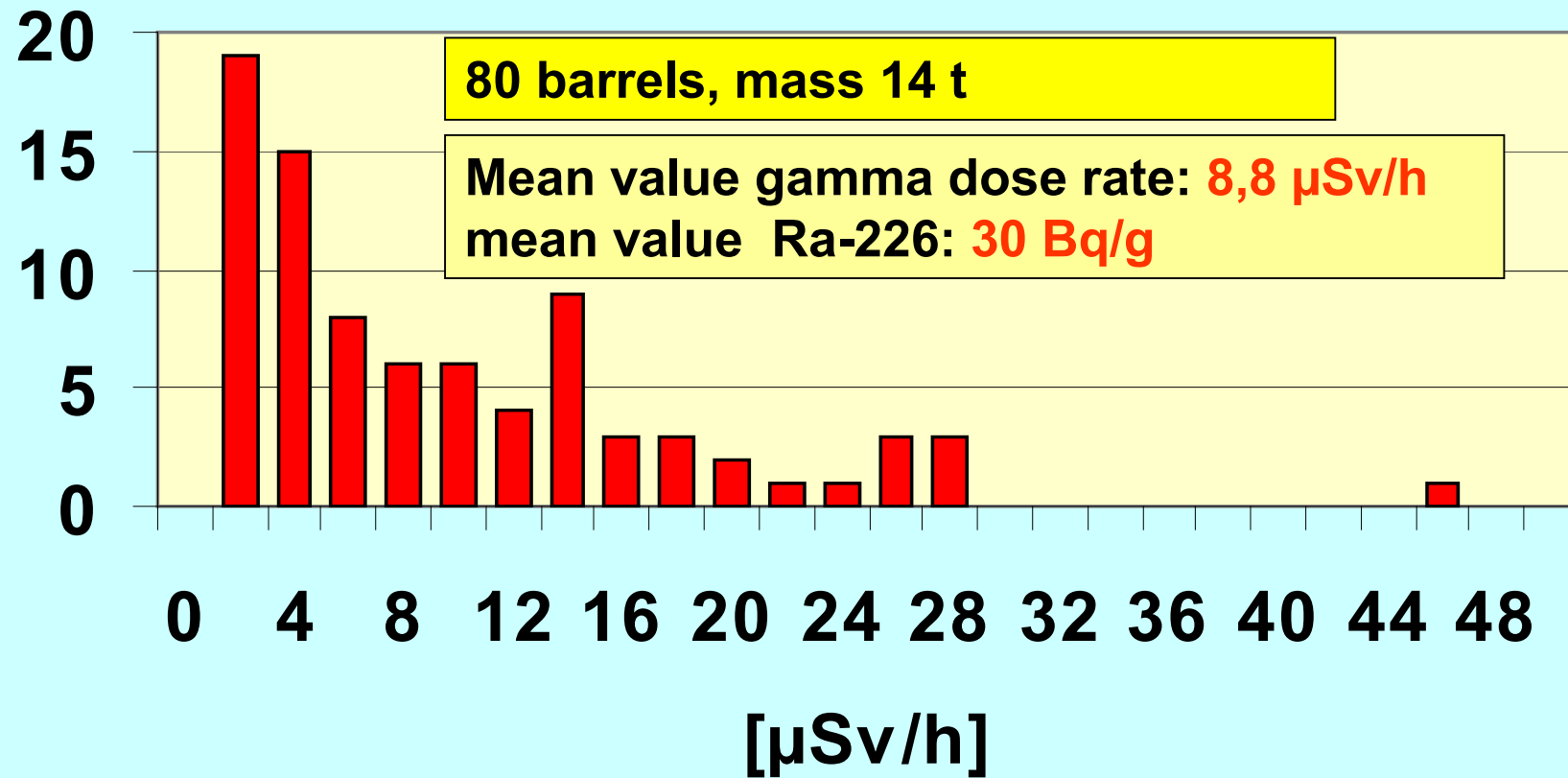


density and thickness of scale in a gas production pipe



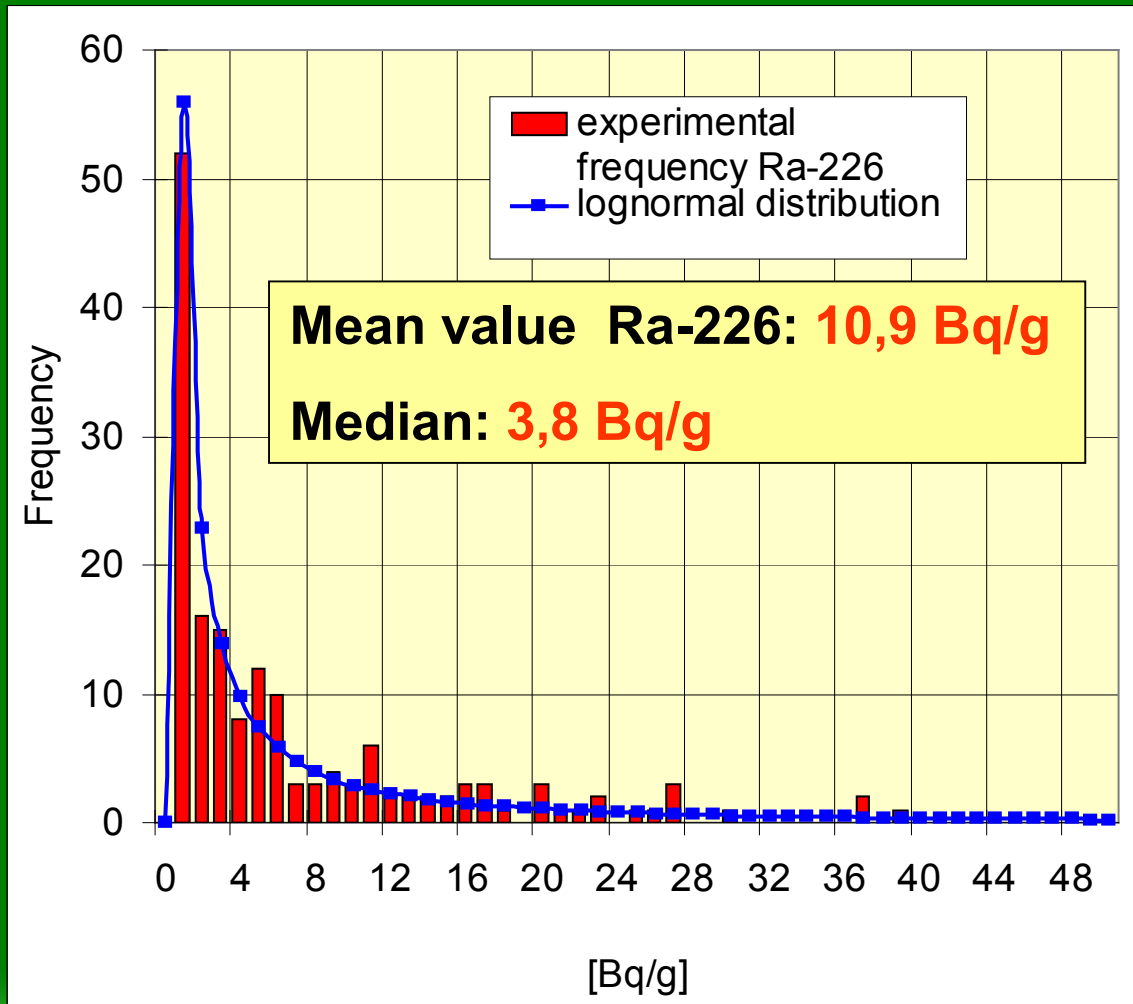


Gamma dose rates of barrels with residues (Scales from hydrocleaning, high pressure cleaning or waterblasting, 2000 bar)





Ra-226 distribution in barrels with sludge

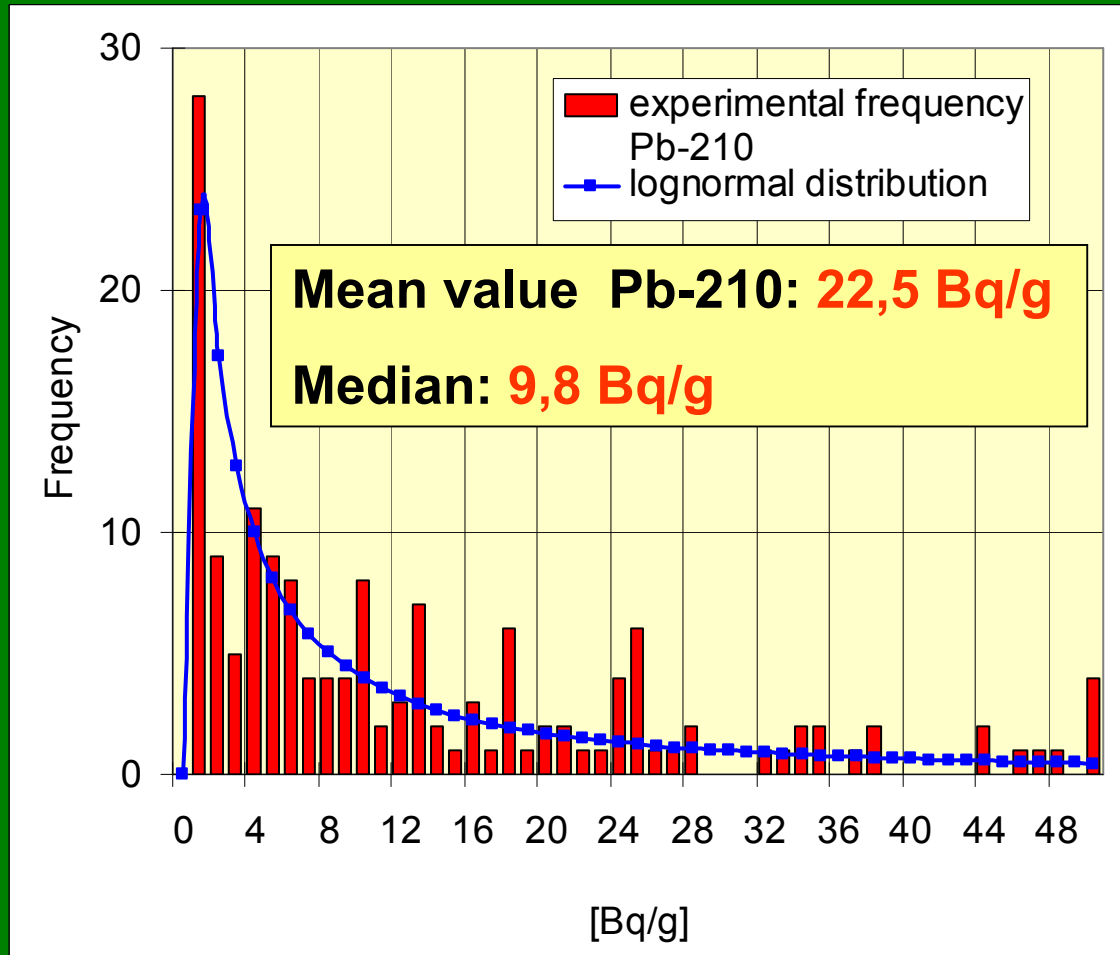


172 barrels with
sludge,

Mass: 17 tons



Pb-210 distribution in barrels with sludge



**172 barrels with
sludge,**

Mass: 17 tons

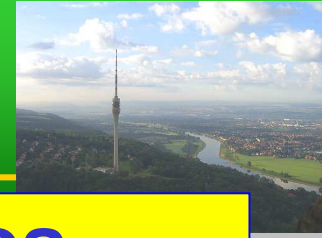
**In average
specific Pb-210
activity of sludge
larger than that
of Ra-226**



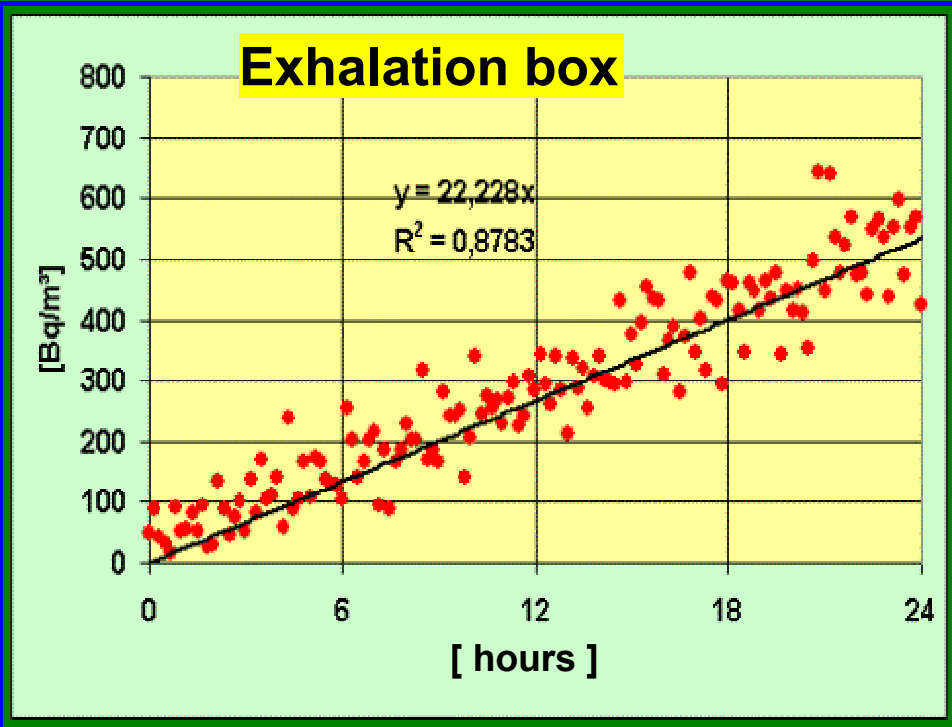
Typical activity ranges in Sludge and Scale

	Sludge [Bq/g]	Scale [Bq/g]
Ra-226	up to 150	5 - 500
Ra-228	30	1 - 350
Th-228	depending on age	depending on age
Pb-210	up to 500	1 - 500
Po-210	(depending on age)	depending on age

Specific activities of U-238 und Th-232 mostly below 1 Bq/g.

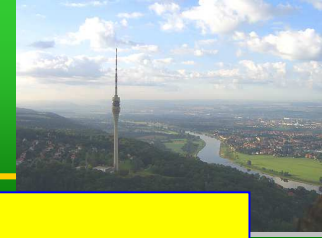


Radon Generation of Dry Scales



in a closed tubing 25.000 Bq/m³ of radon could be reached

[thickness] [mm]	Ra-226 [Bq/g]	[exhalation rate] [Bq/(m ² s)]	[exhalation rate] [Bq/(m ² h)]	[diffusion parameter] [m ² /s]
2,7	440	0,0068	25	5,5·10 ⁻¹⁸



Radon Generation of Sludge

Practically
radon exhalation of sludge
of minor importance



Why?

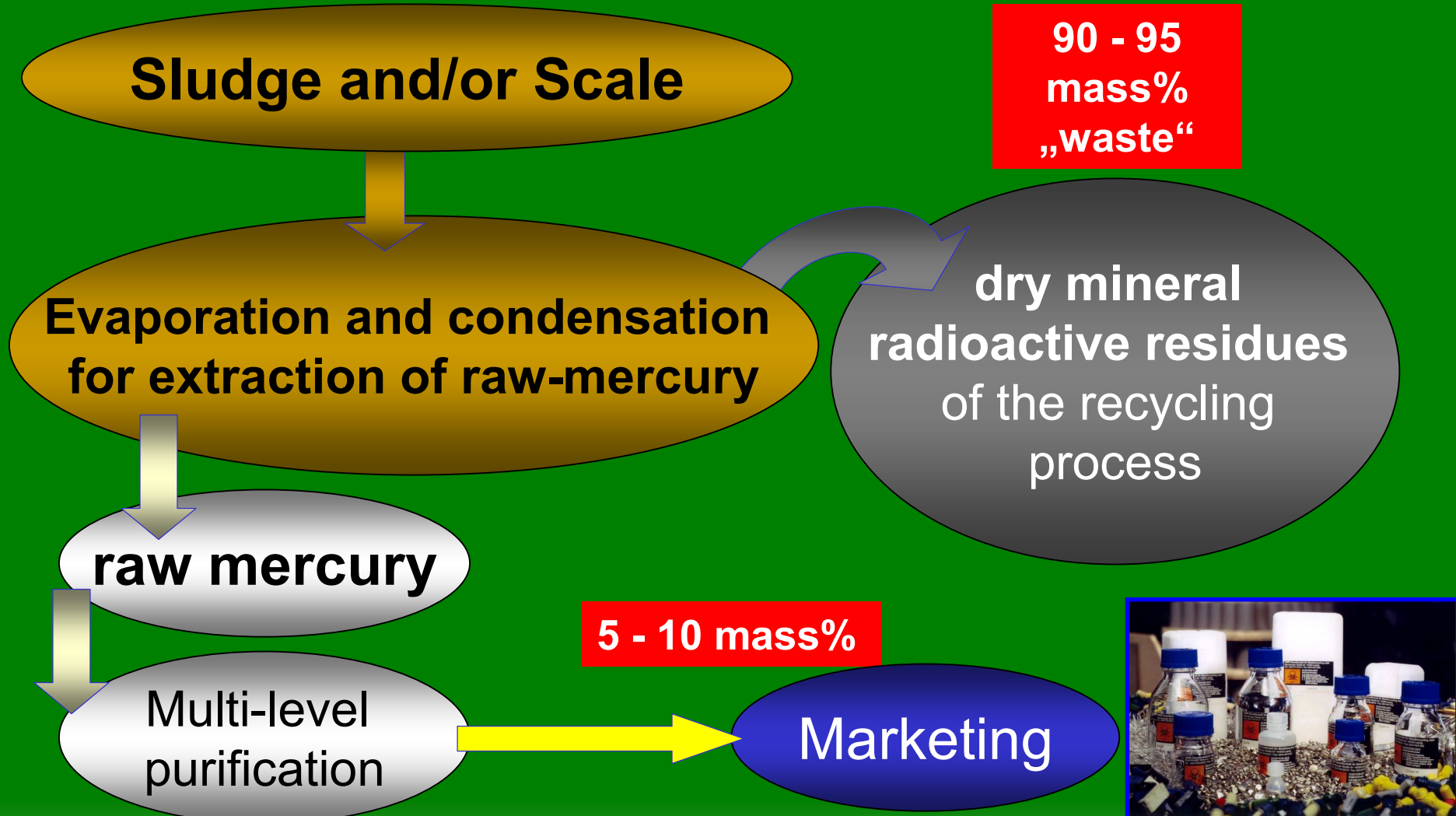
Radon Oil-Water distribution
coefficient (K_d) is rather large!

	K_d
Diesel / Water	$39,8 \pm 4$
Benzine / Water	$52,3 \pm 6$
Air / Water	≈ 4

$$K_d = \frac{c_{\text{Oil}} \left[\frac{\text{Bq}}{\text{l}} \right]}{c_{\text{W}} \left[\frac{\text{Bq}}{\text{l}} \right]}$$



Mercury recycling process





Preparatory work

Preparatory work	Radiation Protection
➤ Taking over the NORM residues	yes, not considered
➤ Transport of NORM (ADR 7) from somewhere to Leipzig	yes, not considered
➤ Reception and Inspection control in Leipzig	yes
➤ Temporary Storage of NORM	yes
➤ Compilation of Charges for the production process (de-mercurisation)	no



Ongoing work

Vacuothermal de-mercurisation

**RP
yes**

Immobilization of the dry residues

**RP
yes**

Disposal at a landfill

**RP
yes**



Take-over room





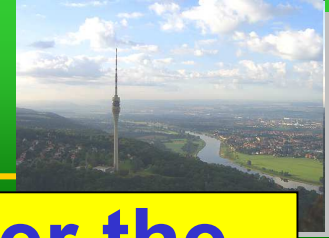
Storage of sludge and scale





Plant for de-mercurisation





Storage of dry mineral residue after the de-mercurisation process





Equipment for making the immobilisates (about 1:1 mixture of NORM residues and geo-polymer)





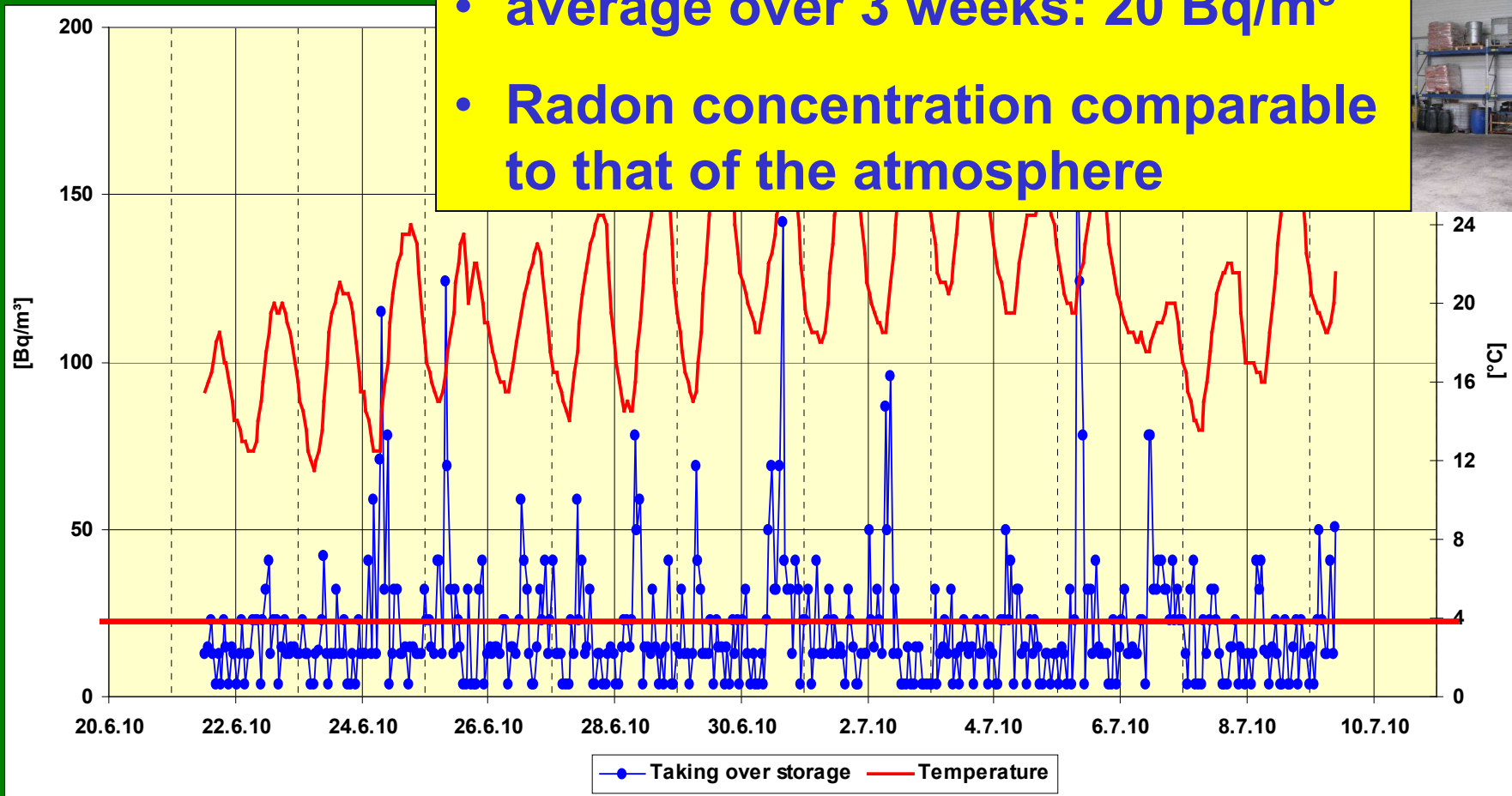
Radiological measurements for dose assessment

- Radon measurements
- Gamma rate measurements
- Dust measurements (α - and γ -spectroscopy)
- Radionuclide analyses (α - and γ -spectroscopy)



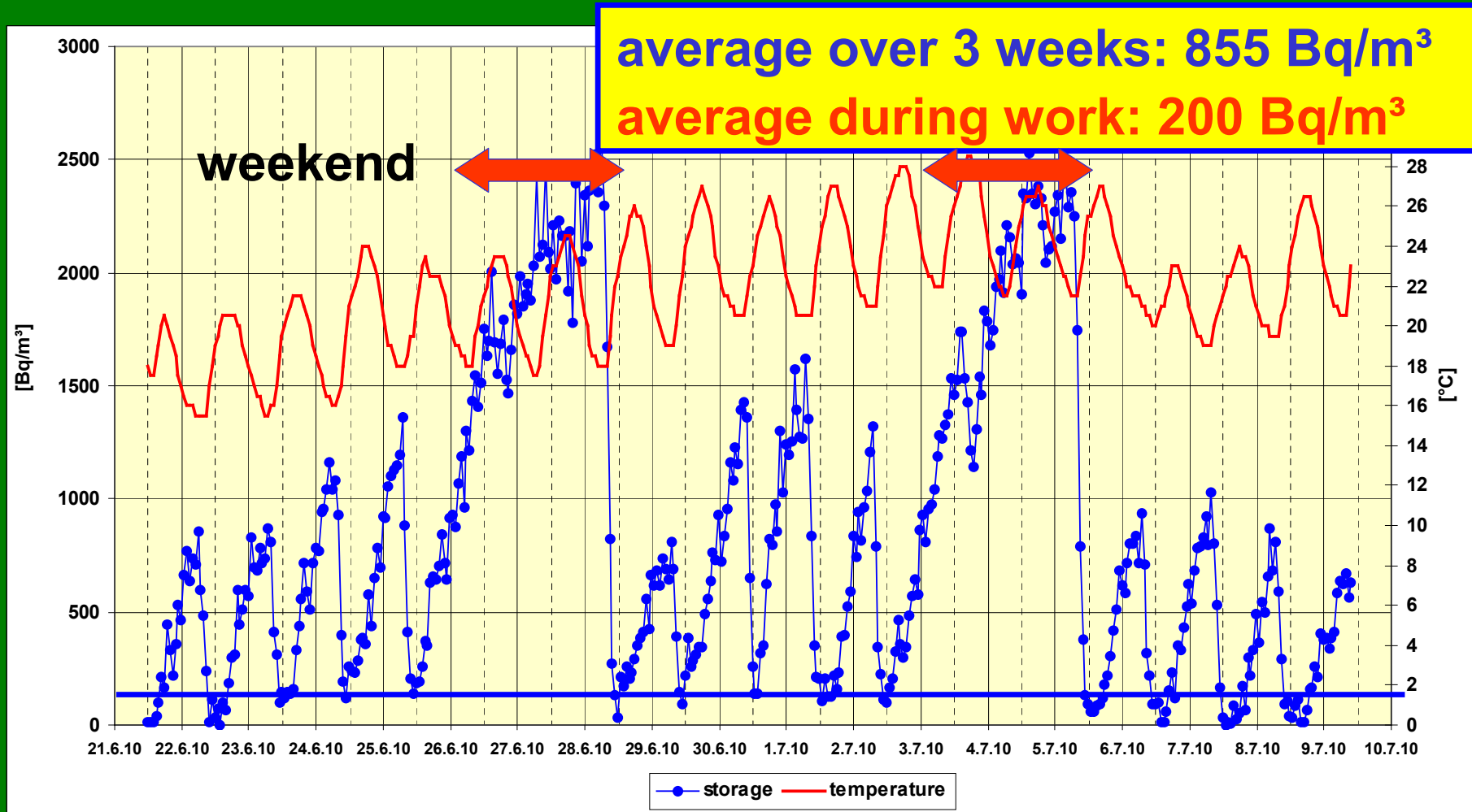
Radon concentration in taking-over storage

- average over 3 weeks: 20 Bq/m³
- Radon concentration comparable to that of the atmosphere





Radon concentration in the storage





Why elevated Radon concentrations?

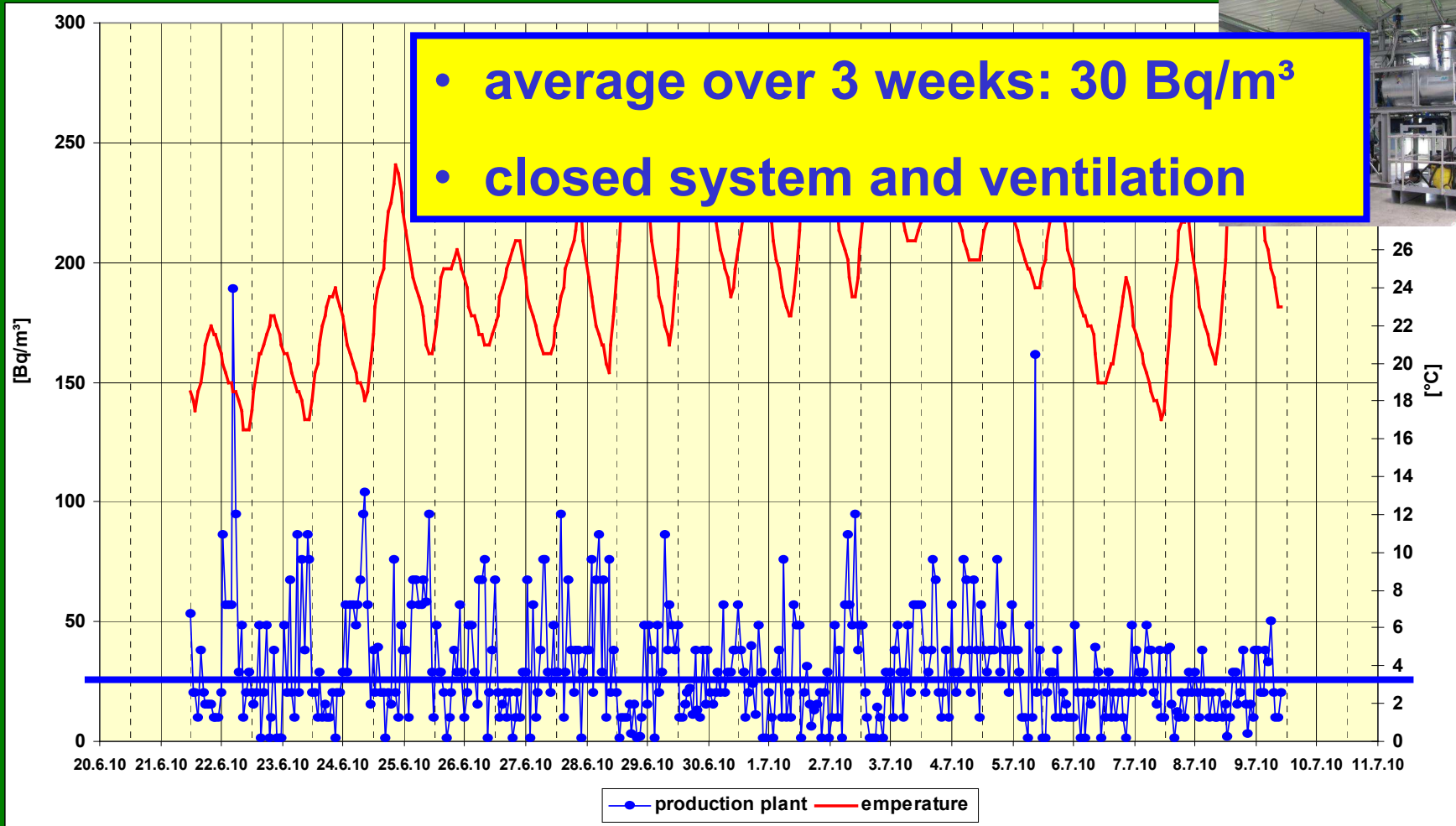
Residues after the de-mercurisation are

- almost dry,
- no volatile organic compounds,
- emanation factor E: 0,2 - 0,6
- specific Ra-226-activity fairly high
- stored in big bags





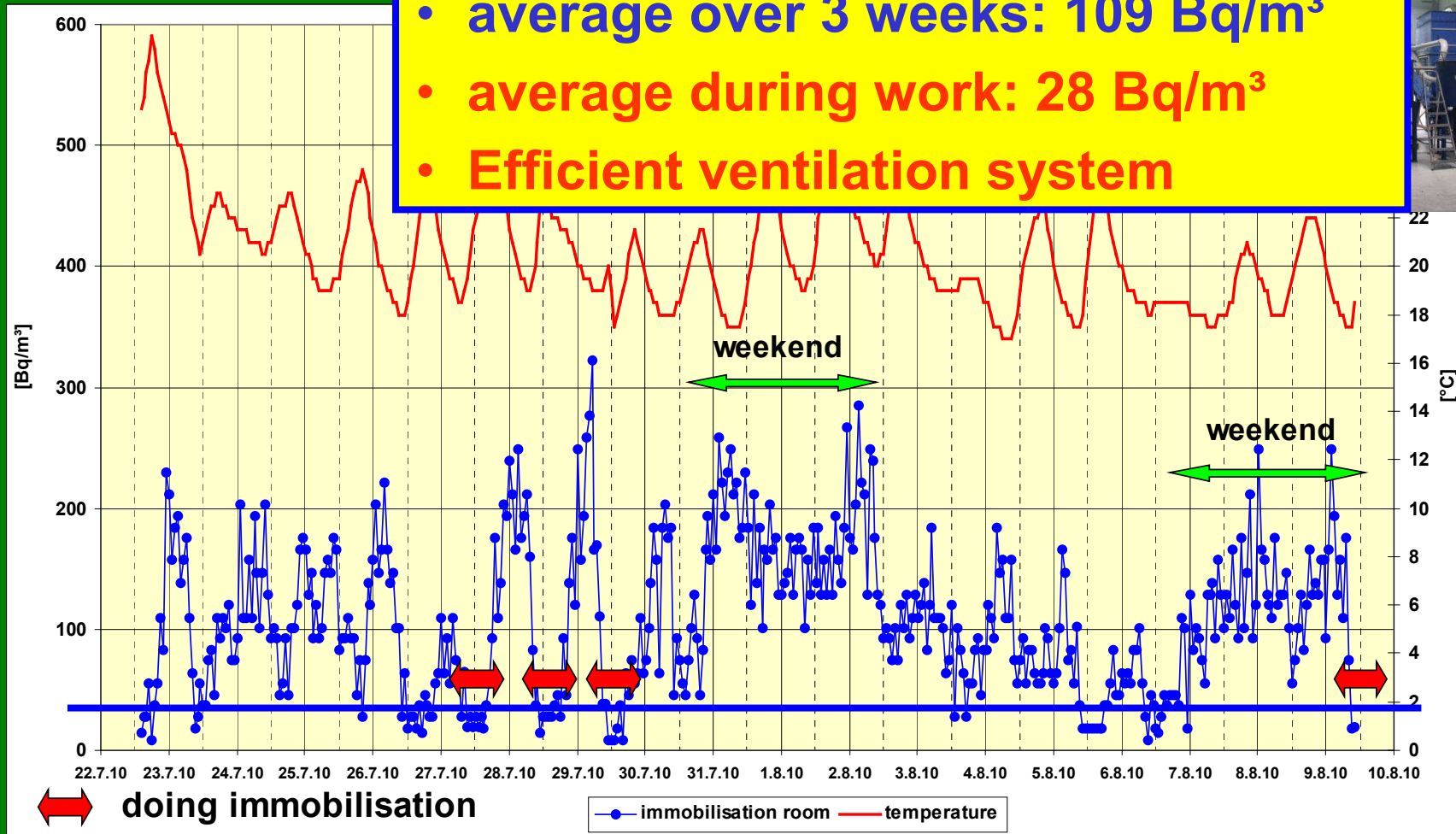
Radon concentration in production plant





Radon concentration in immobilisation room

- average over 3 weeks: 109 Bq/m³
- average during work: 28 Bq/m³
- Efficient ventilation system





Summary: Radon exposure

work	Exposure time [h]	Radon concentration [Bq/m³]	Dose [mSv]
• taking-over, compiling charges	100	20	0,01
• handling in the storage	200	200	0,12
• de-mercurisation	1200	30	0,11
• immobilisation	300	28	0,03
sum	1800		0,27



Summary: gamma rate measurements

taking-over [$\mu\text{Sv/h}$]	storage [$\mu\text{Sv/h}$]	production plant [$\mu\text{Sv/h}$]	equipment for immobilisation [$\mu\text{Sv/h}$]
0,24	0,98	0,33	0,42



taking-over [mSv]	storage [mSv]	production plant [mSv]	equipment for immobilisation [mSv]
0,014	0,118	0,238	0,076



Summary: external exposure

work	Exposure time [h]	Gamma dose rate [nSv/h]	Dose [mSv]
• taking-over, compiling charges	100	240	0,014
• handling in the storage	200	980	0,118
• de-mercurisation	1200	330	0,238
• immobilisation	300	420	0,076
sum	1800		0,45



Inhalation of Dust

We quantify the dust deposition by measuring the total activity of the used masks!!

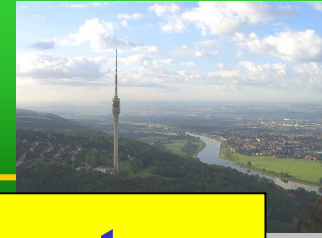


Very often dominating together with Po-210

➤ Most effective and accurate method!!

**Example:
10 days of
deposition**

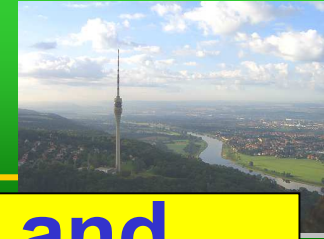
worker	Ra-226 [Bq]	Ra-228 [Bq]	Th-228 [Bq]	Pb-210 [Bq]
1	0,6	0,15	0,14	1,4
2	1,7	0,4	0,44	4,4



Exposure due to Inhalation of Dust

worker	exposure time [h]	Dose <u>without</u> mask [mSv]	Dose <u>with</u> mask [mSv]
1	1800	0,24	0,024
2	1800	0,74	0,074

Without masks one could easily get an exposure exceeding 1 mSv/year!!



Total doses for de-mercurisation and immobilisation

work	Exposure time [h]	External exposure [mSv]	Inhalation radon and dust [mSv]	Total Dose [mSv]
• taking-over, compiling charges	100	0,014	0,014	0,028
• handling in the storage	200	0,118	0,128	0,246
• de-mercurisation	1200	0,238	0,16	0,398
• immobilisation	300	0,076	0,042	0,118
sum	1800	0,45	0,34	0,79

Due to radiation protection measures total exposure dose < 1 mSv/year for workers



Immobilization with geo-polymer

*dry mineral
radioactive residues
of the recycling
process*

**Immobilization of the residues
by addition of a polymer
(geo-polymer)
mixing ratio: about 1 : 1**

**Up to 250 kg in
a 120 l barrel**

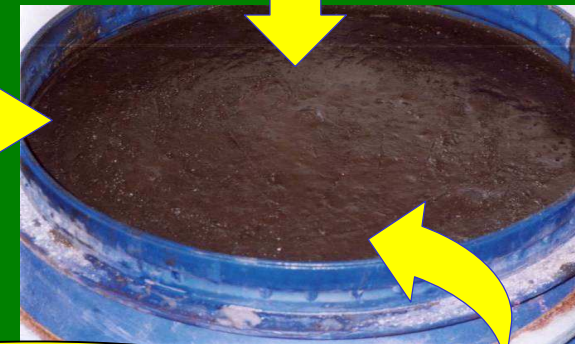


**ready for
disposal**



Characteristics of the Immobilisate made with geo-polymer

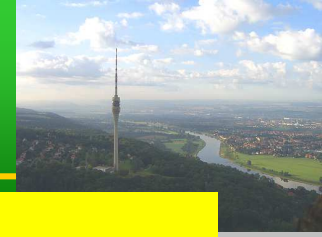
Long term stability,
Like Hardened Concrete,
weather-proof



old mortar similar
to geo-polymer
concrete

negligible elution
of harmful substances
(radionuclides,
heavy metals, arsenic)

No Exposure via the water path



Elution of Immobilisates

Sample	Ra-226	Ra-228	Th-228	Pb-210	Po-210
	[Bq/g]	[Bq/g]	[Bq/g]	[Bq/g]	[Bq/g]
1	18	8,85	9,1	3,2	3,2
2	61	11,4	15	80	80

Eluate	Ra-226	Ra-228	Th-228	Pb-210	Po-210
	[Bq/l]	[Bq/l]	[Bq/l]	[Bq/l]	[Bq/l]
1	<0,02	<0,008	<0,02	<0,02	0,018
2	<0,09	0,015	<0,01	0,12	0,005

➤ **Eluate has a quality comparable to that of „mineral water” concerning concentration of radionuclides**
 → **no exposure along the water path**



Advantage of the Immobilisate made with geopolymer



**Negligible
low exhalation
of radon**

**No formation of dust
at handling, transportation
and disposal**

**No elution
of radionuclides
after disposal!!**

**No internal
exposition**

**External Exposition
due to Gamma
Radiation
is the only
Exposure Pathway**



According to § 98 of the German Radiation Protection Ordinance the de-mercurised residues are released

The criterion is:

The effective dose should not exceed 1 mSv/year (reference value for persons of the population).

The values of the specific activities of the relevant radionuclides play practically no role!



Transport to the landfill



radioactive immobilisates

Mercury





Barrels filled with immobilisates ready for the transport to the landfill



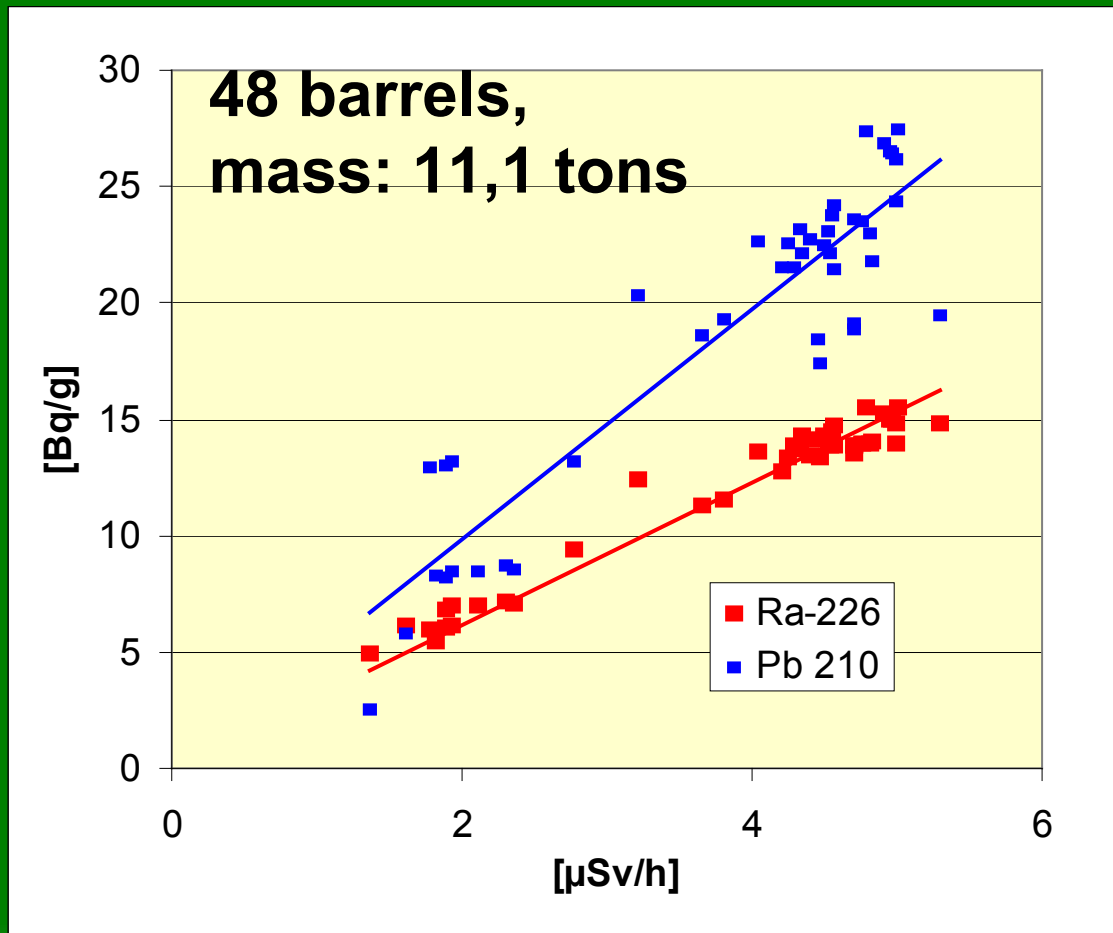


Transport: Radioactive, ADR 7





Radiological characteristics of a typical loading to be disposed



Mean values

Ra-226: 12,2 Bq/g
Pb-210: 19,0 Bq/g

**Gamma dose rate:
3,8 µSv/h**



Gamma dose rate in the driver's cab





Transport on street to the landfill



Distance to the landfill 13 km

Driving time: 15 minutes

Gamma dose rate in the driver's cab: 0,3 - 0,6 $\mu\text{Sv/h}$

15 transportations per year to the landfill

Exposure dose of the driver: $<2 \mu\text{Sv/year}$



Work at the landfill



Handling and storing the barrels: 15 minutes

Distance of the worker to the barrels 3 - 5 meters

Gamma dose rate: $<0,3 \mu\text{Sv/h}$

Exposure dose of the worker at the landfill:

$<2 \mu\text{Sv/year}$



Summary

- Despite the fact, that we are dealing with rather elevated specific activities of the relevant radionuclides, the exposure doses can be kept low by applying necessary Radiation Protection measures.
- The immobilisation of de-mercurised sludge and scale has the great advantage, that a very stable deposition of hazardous residues can be achieved.