AGH UNIVERSITY OF SCIENCE AND TECHNOLOGY

DETERMINATION OF RADIUM ISOTOPES OF VERY LOW CONCENTRATIONS IN WATER SAMPLES BY LSC TECHNIQUES

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Outlines of presentation

- Radiological criteria for drinking water
- Guidance levels for natural radionuclides in drinking water
- Critical Level and Low Limit Detection
- Chemical and measurement procedures for radium isotopes determination
- Factors controlling the bacground level in the LSC method
- Influence of $BaCl_2$ on the measured background samples
- Influence of $BaCl_2$ on chemical efficiency
- Test of the method
- Conclusions

Radiological criteria for drinking water

- **According to WHO Guidelines the annual committed effective dose resulting from absorb of all radionuclides in drinking water should be below 0.1 mSv/year**
- **According to Guideline, the max level (GL) for radionuclides in drinking water can be calculated as follow:**

$$GL = \frac{IDC}{h_{ing}(ag) \cdot V(ag)}$$

where:

GL – maximum level of radionuclide in drinking water $[mBq/L]$,
IDC – annual permissible committed dose; $IDC = 0.1\,mSv$,
$h_{ing}(ag)$ – dose coefficient for ingestion by person in the ag – age group $[mSv/mBq]$,
$V(ag)$ – annual ingested volume of drinking waters

$$\sum_i \frac{C_i}{GL_i} \leq 1$$

Sum of all ratios of the real concentration of i-isotope to its GL should be lower than 1.

Guidance levels for some selected natural radionuclides in drinking water $[mBq/L]$

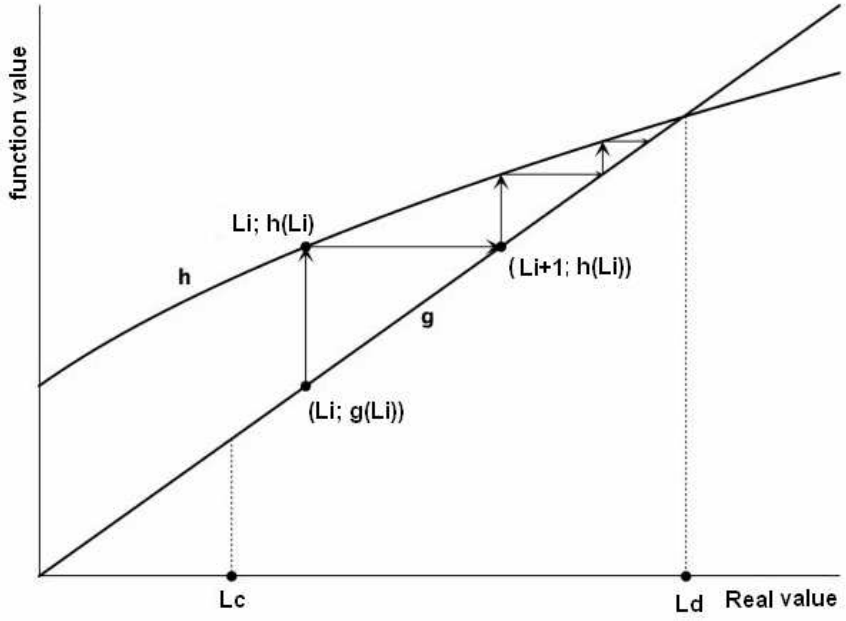
Isotope	Age group					
	< 1 y	1-2 y	2-7 y	7-12 y	12-17 y	> 17 y
U-238	403	1142	1712	2014	2045	3044
U-234	370	1054	1557	1851	1851	2795
Th-232	30	304	391	472	548	595
Ra-226	29	143	221	171	91	489
Ra-228	5	24	40	35	25,6	198
Ra-224	51	208	391	527	685	2107
Pb-210	1.6	38	62	72	72	198
Po-210	5	16	31	53	86	114

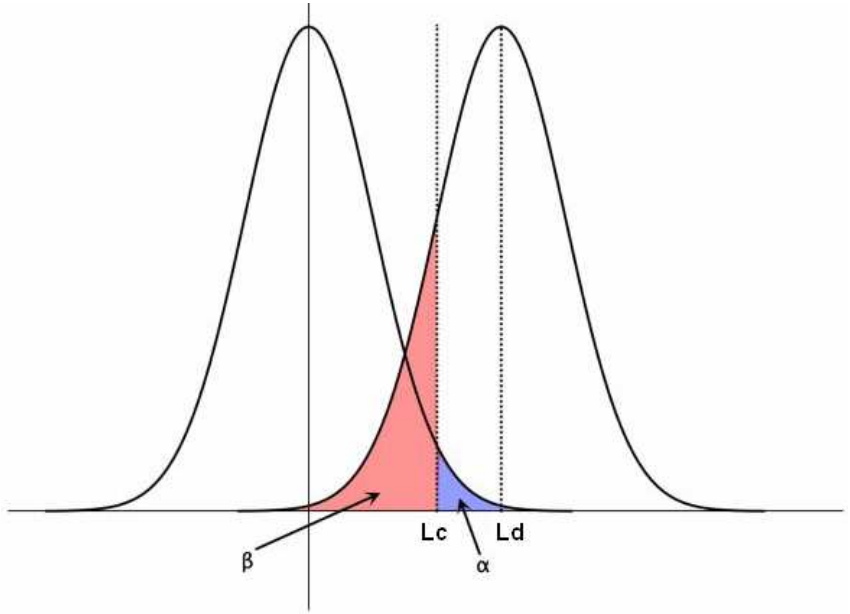
Critical level and limit of detection

Follow Lloyd Currie:

Critical level: $L_C = k\alpha \cdot \sigma_B$

Detection limit: $L_D = L_C + k\beta \cdot \sigma_D$





Follow ISO 11929:2010:

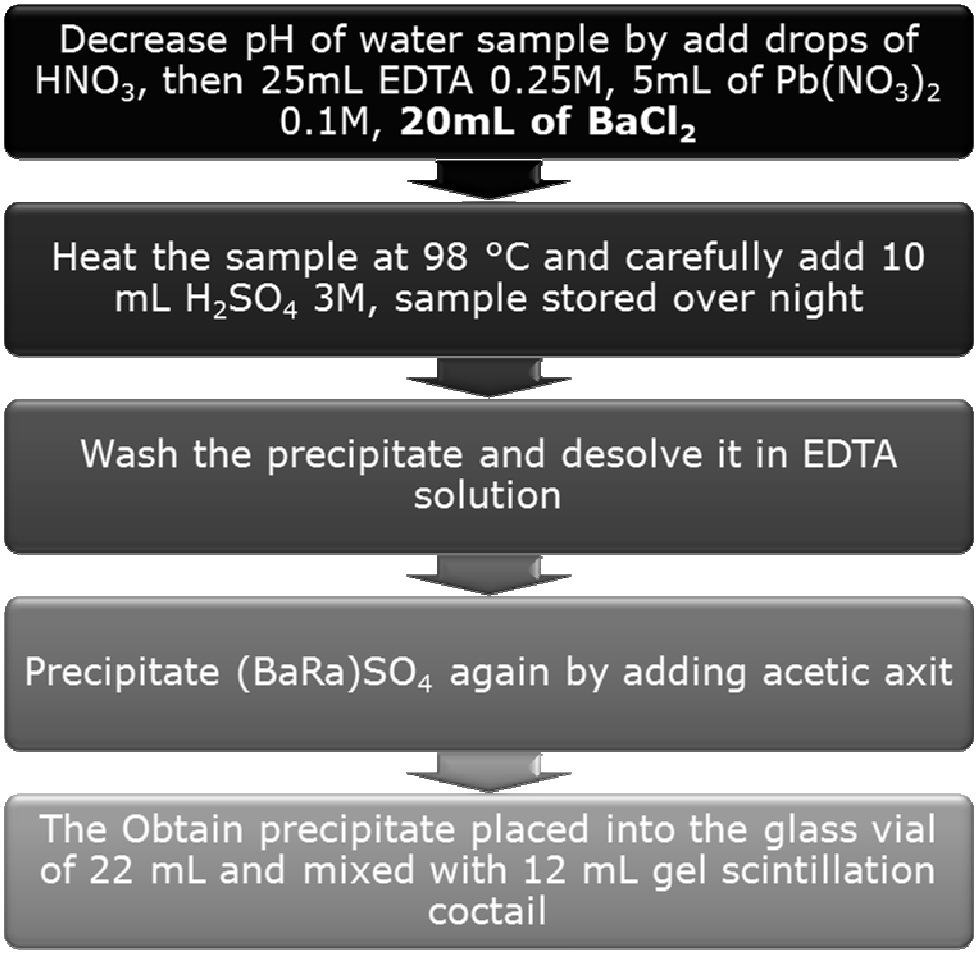
$$g(L_i) = L_i$$

$$h(L_i) = L_i + k \cdot \sigma_i$$

The LLDs requirement of Council Directive for analyzing methods and LLDs of routine methods

Isotope	COUNCIL DIRECTIVE 2013/51/EURATOM	Routine methods
U-238	0.02 Bq/L	0.005 Bq/L
U-234	0.02 Bq/L	0.005 Bq/L
Ra-226	0.04 Bq/L	0.01 Bq/L
Ra-228	0.02 Bq/L	0.03 – 0.8 Bq/L
Pb-210	0.02 Bq/L	0.03 – 0.8 Bq/L
Po-210	0.01 Bq/L	0.01 Bq/L

Chemical procedure for radium in water sample



The chemical agents used in chemical procedure:

- HNO3
- EDTA
- Pb(NO3)2
- BaCl2
- H2SO4
- CH3COOH

In rooutine analyzing, the background sample is to be prepared together with analyzed samples using distilled water.

LSC spectrometers

Guardian 1414 α/β Liquid Scintillation counter

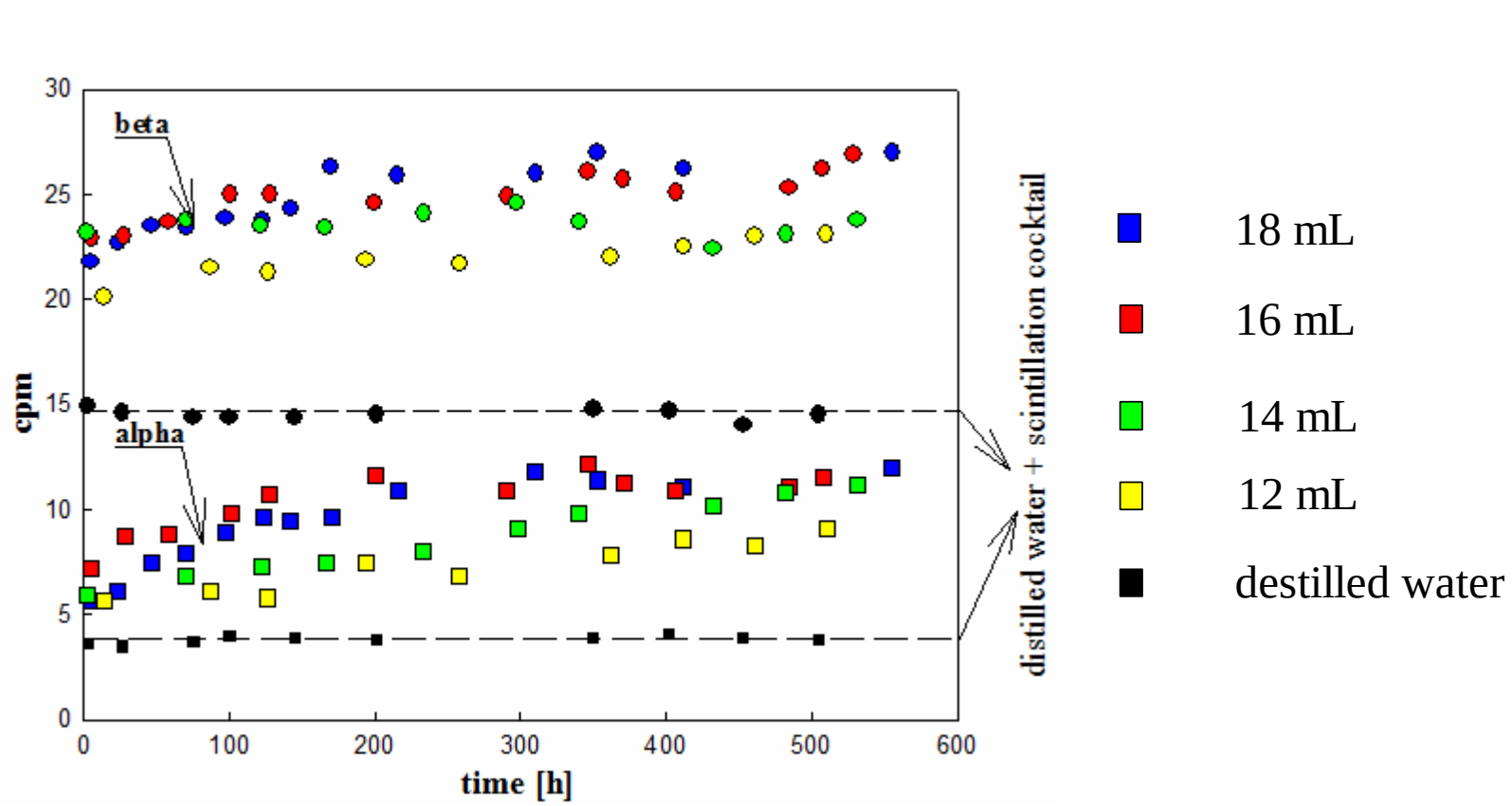
Quantulus

Factors controlling the background level in LSC

- **The electronic noise;**
- **The cosmic and radiation ray from surrounding;**
- **Chemical Reagents;**
- **Scintillation cocktail and glass vial.**

The influences of the chemical reagent $BaCl_2$ were carried out; the blank samples were prepared using 0.5 L of distilled water with different amounts of $BaCl_2$, but the amounts of the other chemical reagents were the same as described in procedure.

Measured count rates of the α and β canals for blank samples with different masses of $BaCl_2$-0.1M



Concentration of Ra-226 in chemical reagent $BaCl_2$ – 0.1 M

Volume of BaCl2 -0.1 M	18 mL	16 mL	14 mL	12 mL
Average α nett count rates (cpm)	7.2	6.6	6.2	5.3
Stand. Uncert.	1.2	0.9	1.3	0.8

Based on the data in the above table, the concentration of Ra-226, amounts to 84 ± 8 Bq/kg of $BaCl_2$. This value was confirmed through gamma spectrometry.

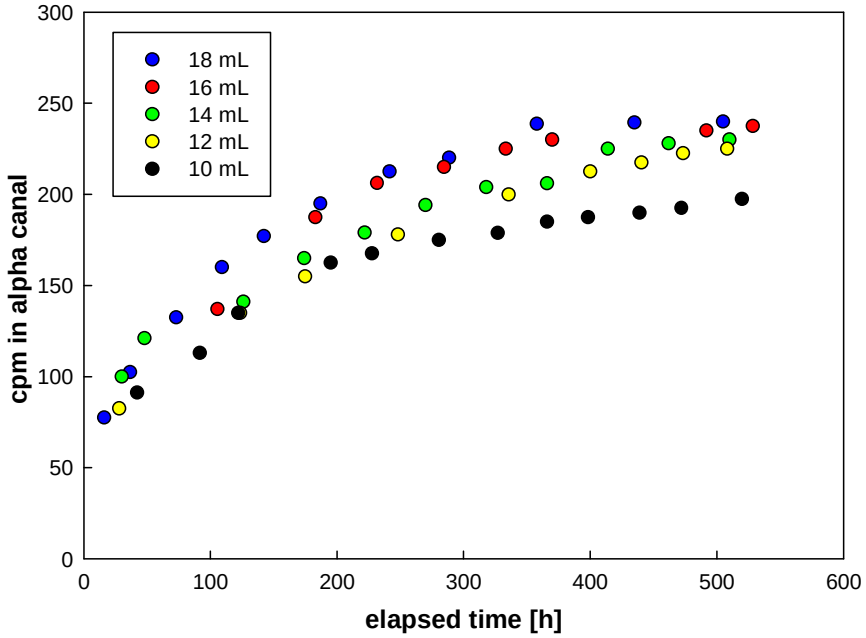
Critical level (L_C)

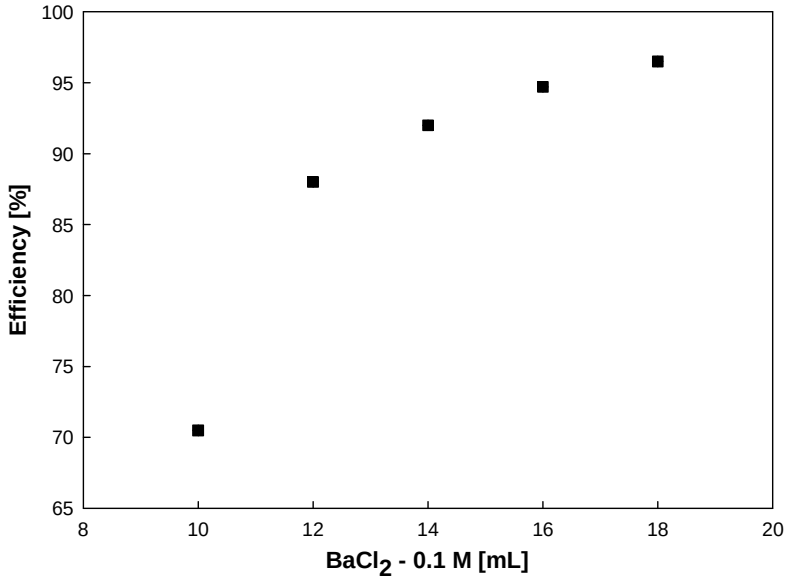
$$L_C = 1.65 \cdot \sigma_0 \ *$$

$BaCl_2$-0.1 M	18 mL	16 mL	14 mL	12 mL	10 mL
Iα (cpm)	7.2 ± 1.2	6.6 ± 0.9	6.2 ± 1.3	5.3 ± 0.8	5.0 ± 0.8
Iβ (cpm)	15.4 ± 3.1	15.8 ± 2.9	14.2 ± 2.8	12.2 ± 2.2	12.0 ± 2.5
Ra-226 (mBq)	8	6	8	6	5
Ra-228 (mBq)	33	30	28	20	25

* by Lloyd Currie

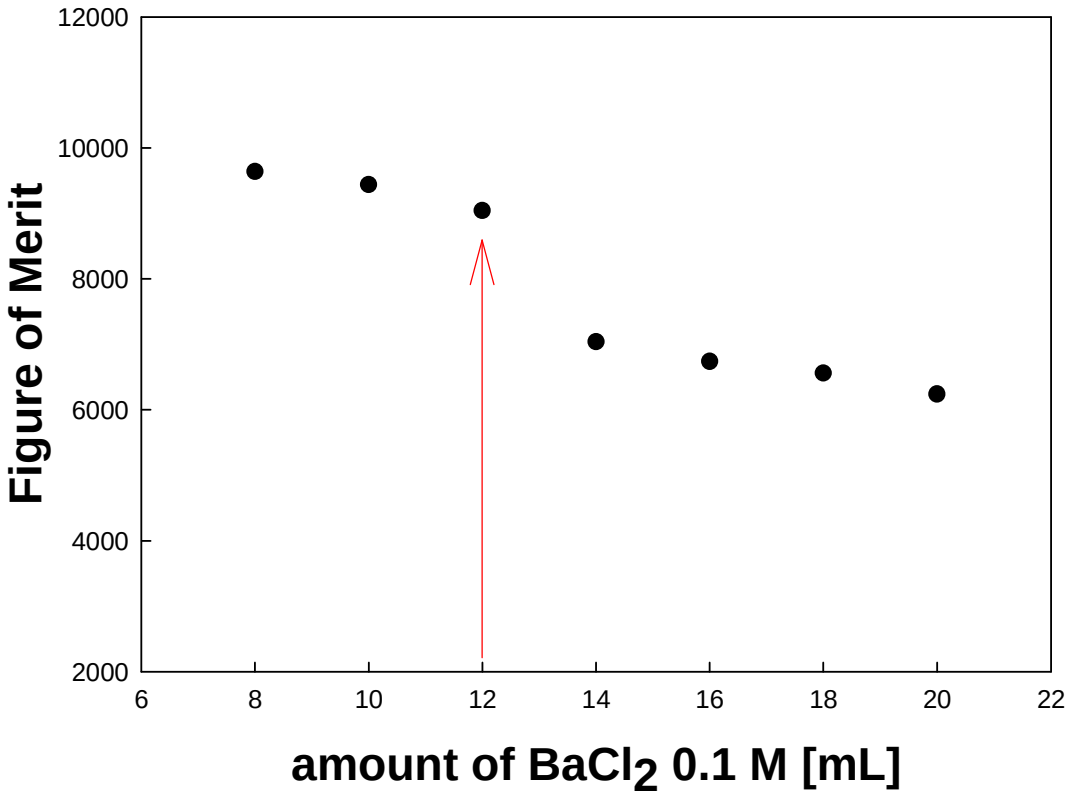
Chemical efficiency as a function of the added amounts $BaCl_2$ 0.1 M





Optimal amount of $BaCl_2$ – 0.1 M

$$F = \frac{signal^2}{background}$$



Test of the method

Sample	Spiked contents of radium isotopes [mBq]		Determined [mBq]	
	$^{226}Ra \pm \sigma$	$^{228}Ra \pm \sigma$	$^{226}Ra \pm \sigma$	$^{228}Ra \pm \sigma$
S1	0	23 ± 2	nd* (≤ 5.0)	17 ± 3
S2	51 ± 2	0	48 ± 2	nd (≤ 20)
S3	53 ± 2	52 ± 3	48 ± 2	50 ± 8

Conclusions

- The current reagent of $BaCl_2$ is Ra-226 contaminated with 84 Bq/kg;
- Critical level for determination of Ra-226 and Ra-228 depend on the used amount of $BaCl_2$;
- 12 mL $BaCl_2$ 0.1 M solution is the optimal used amount to preparing;
- The LSC method is still to be improved, unless the method only can be used for Ra-226 analyzing;
- For high precise determination of Ra-228 we should combine the gamma and LSC methods.


TRANS