Simultaneous determination of ^{89}Sr, and $^{90}Sr/^{90}Y$ in various matrices using Sr-Spec resin

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Topics of the presentation….

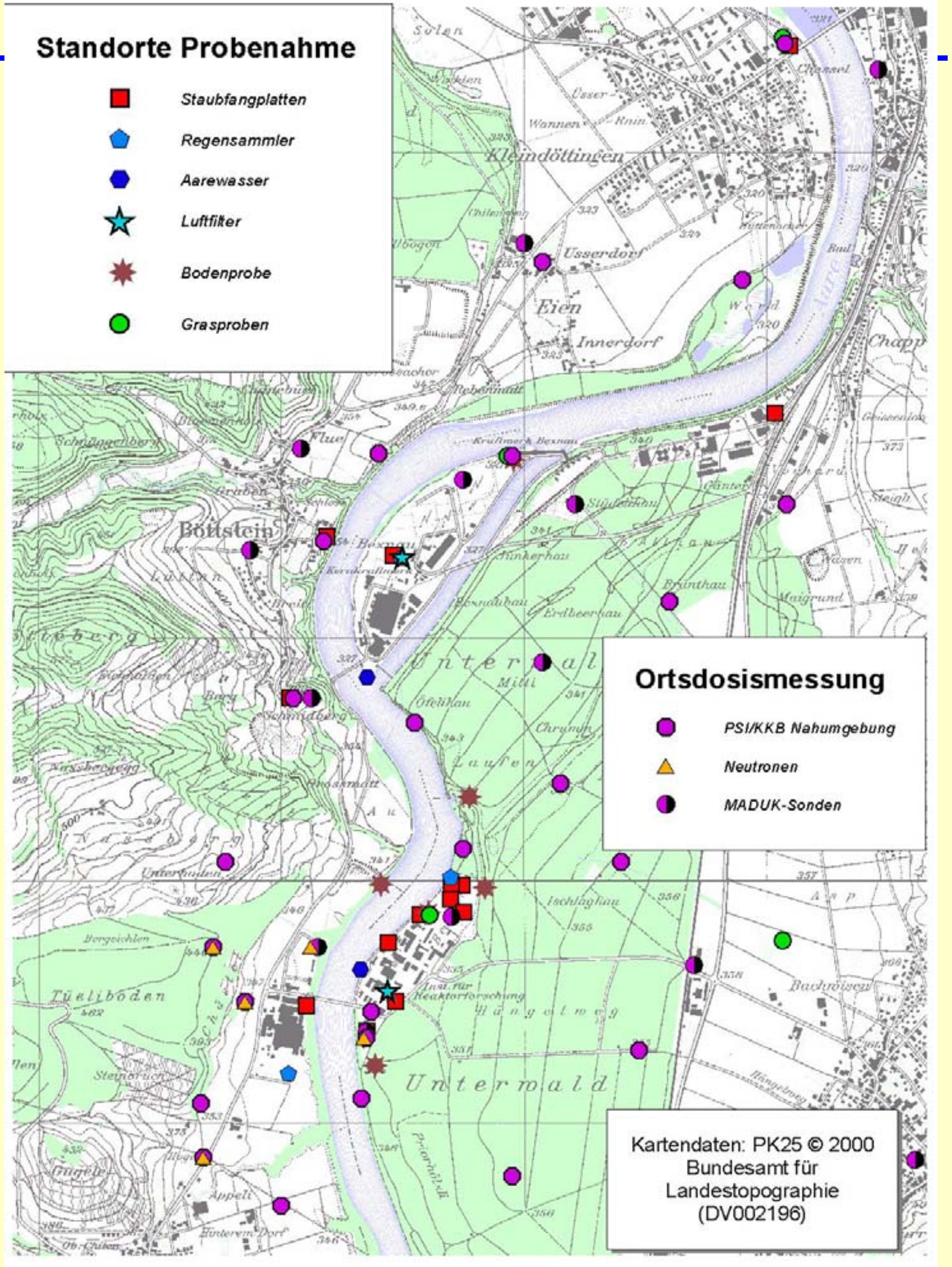
- Immission surveillance in Switzerland / Presentation of the problem of radioactive Sr-isotope analysis
- Presentation of chemical matrices, measurement techniques, tracers to be used for yield determination
- Separation procedure
- Introduction to the LSC 3-window approach
- Case studies ^{90}Sr > ^{89}Sr and ^{89}Sr > ^{90}Sr
- Conclusions

Areal graph of PSI + ZWILAG

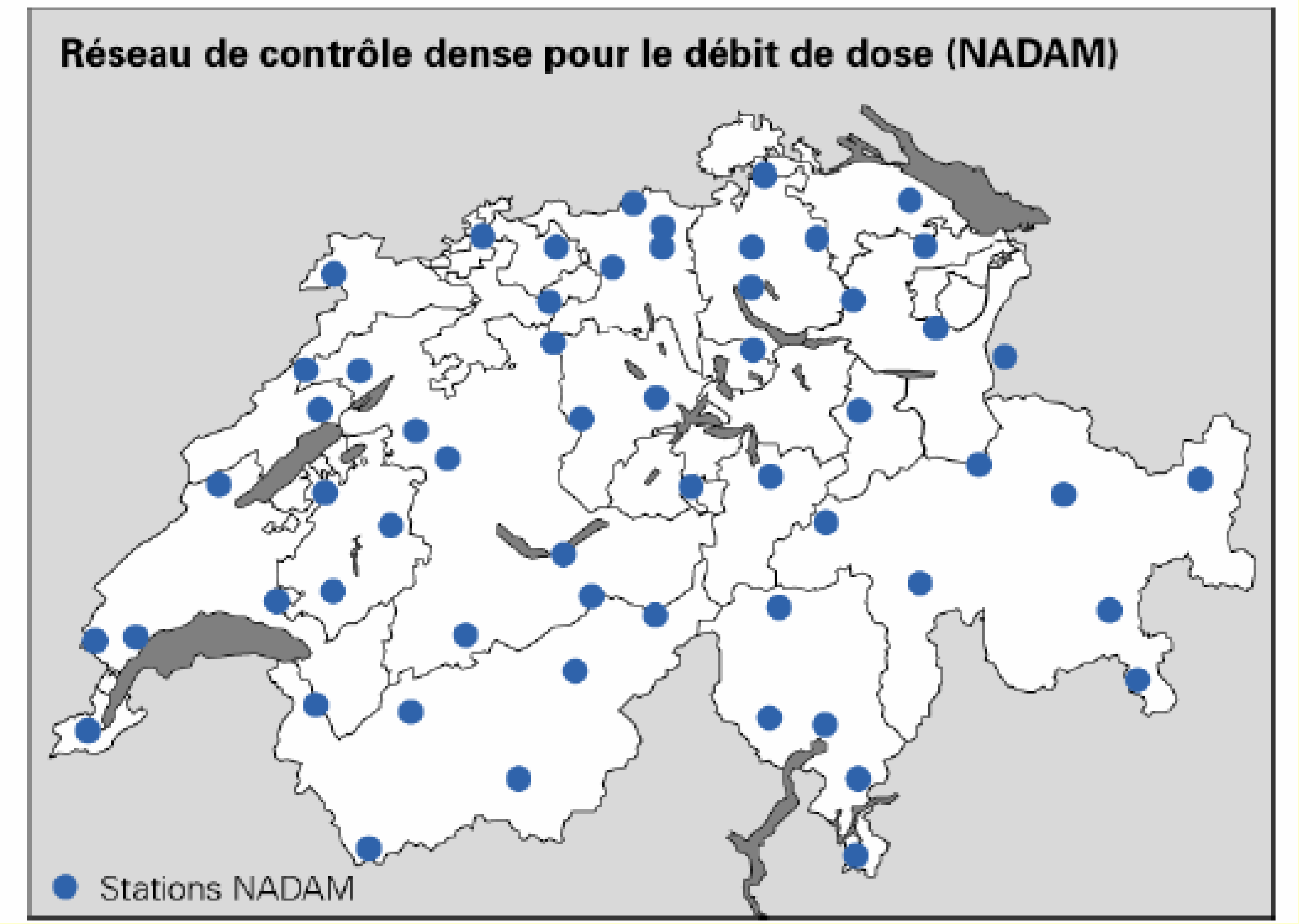
Sample material and isotopes analyzed

samples	**isotopes**
air filters, vaseline plates	^{7}Be, ^{60}Co, ^{131}I, ^{137}Cs, ^{134}Cs...
rain-, river-, waste water	^{3}H, ^{7}Be, ^{60}Co, $^{89,90}Sr$, actinides...
biological (grass, tree leaves, milk etc.)	^{7}Be, ^{14}C, $^{89,90}Sr$, ^{60}Co,...
soil samples	^{60}Co, ^{131}I, ^{137}Cs, ^{134}Cs, $^{89,90}Sr$, $^{238,239,240,241}Pu$, ^{241}Am,

environmental monitoring around PSI and the adjacent nuclear power plant at Beznau: locations for sampling and on-line dose rate recording



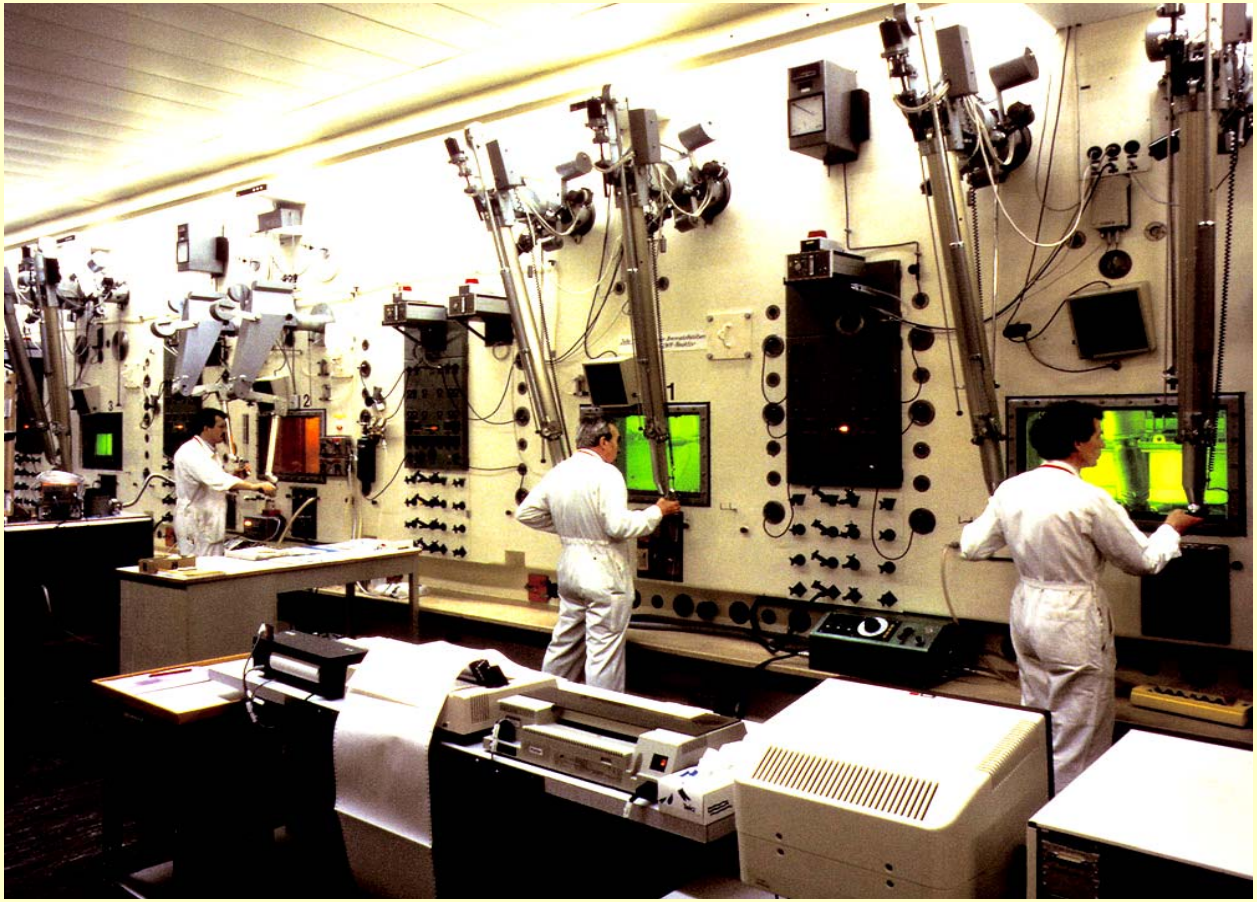
Locations of on-line dose rate measurements in Switzerland



- *immission surveillance at PSI: aerosol collecting unit*

- *immission surveillance at PSI: rain water sampler*

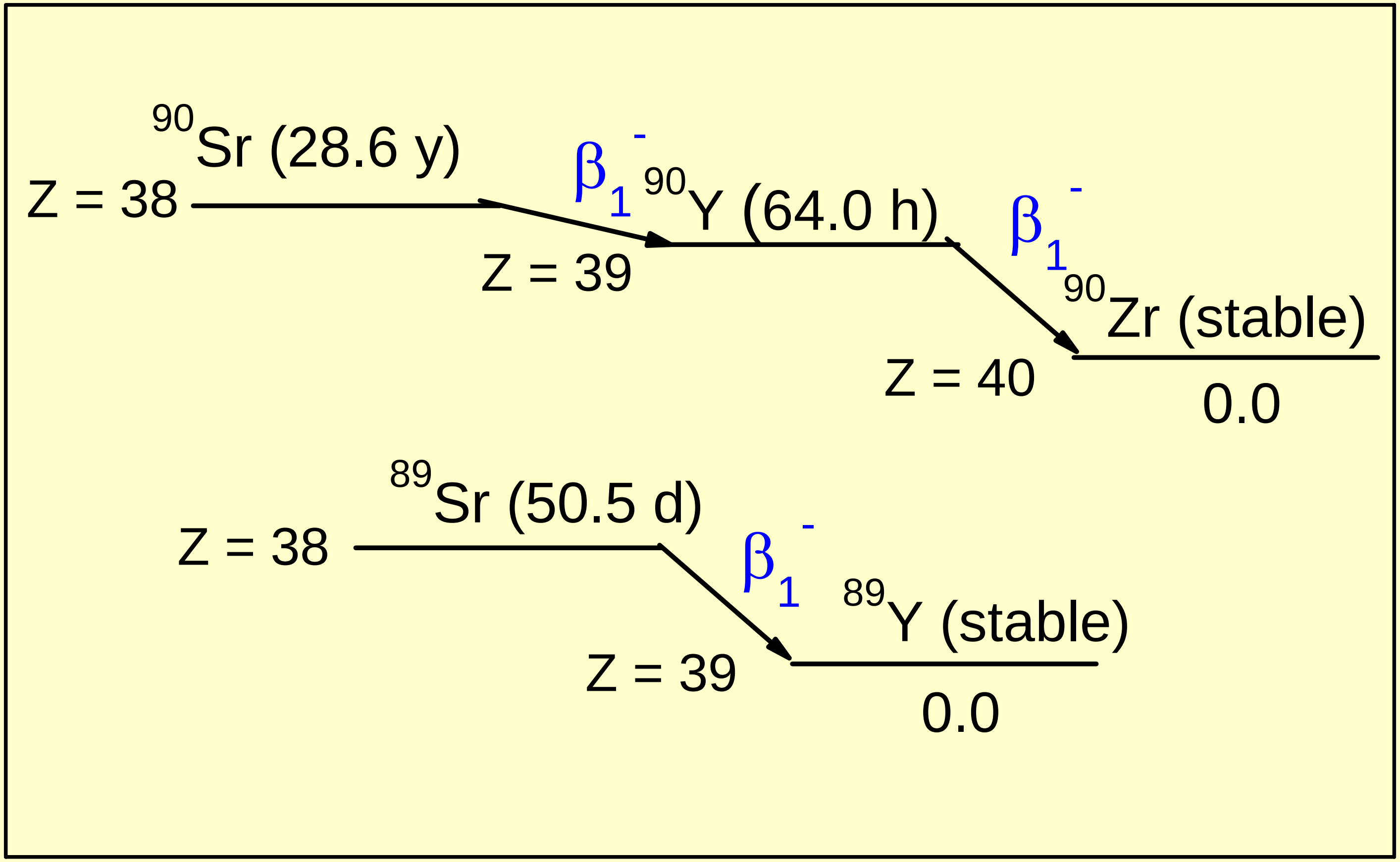
Investigating nuclear materials in the lead glass shielded hot laboratory



Sr-isotope determination: typical matrices investigated at PSI

- Environmental (soil, grass, peat etc.): complex chemical matrix, elimination of radioisotopes from the U-Th-series and ^{210}**Pb** (separation via U/TEVA resin and ammonium citrate after elution of the Sr-fraction from the Sr-Spec column)
- Waste and reactor coolant water: chemically simple, however complex with respect to the radioisotope composition (^{7}Be, ^{54}Mn, $^{58,60}Co$, ^{110m}Ag, ^{124}Sb, $^{134,137}Cs$, ^{133}**Ba** etc, ….) (may require further purification columns)

Decay scheme of ^{89,90}Sr: problem: no gamma ray emission from the excited level to the ground state

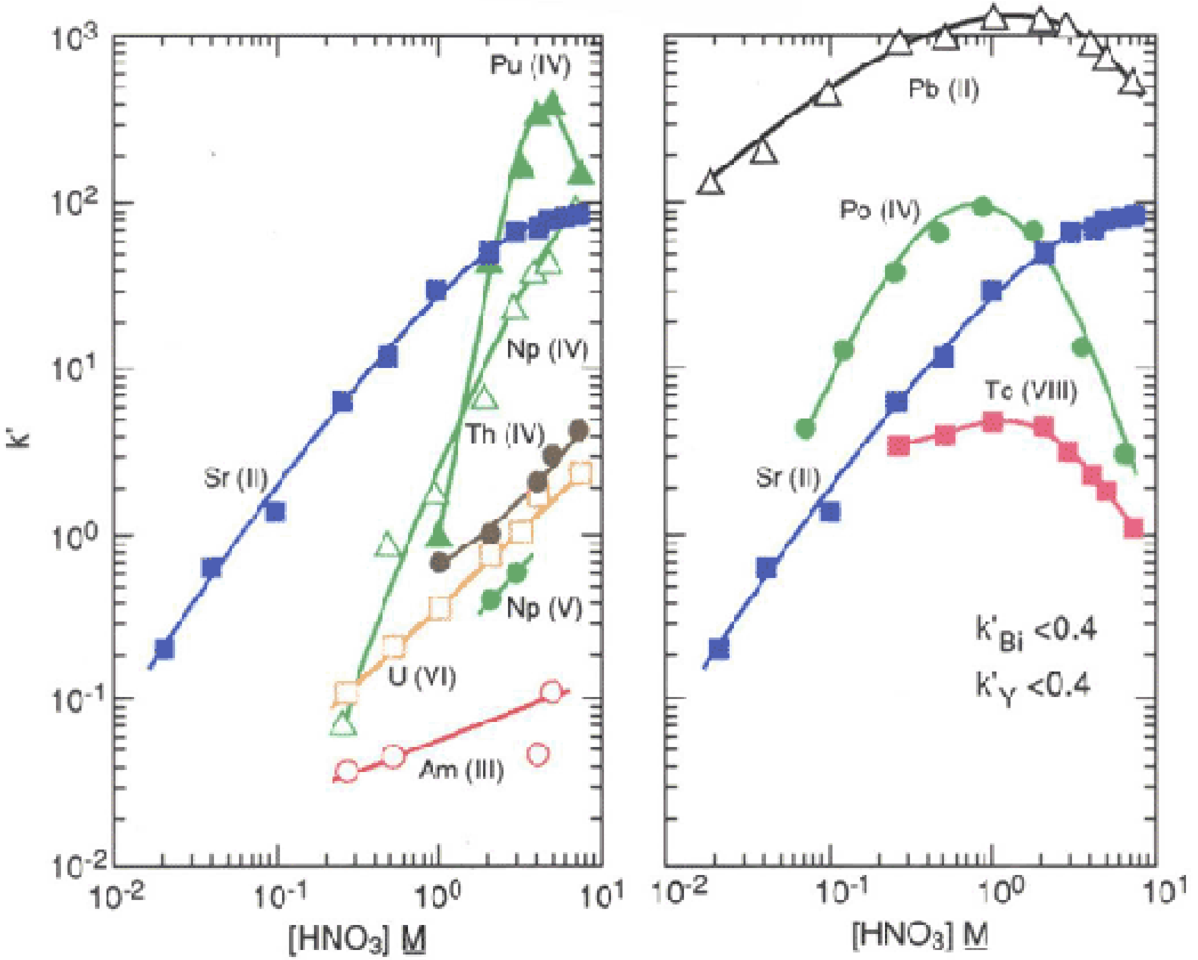


Measurement techniques for determination of ^{85}Sr, $^{89,90}Sr$ and check on radioisotope impurities

measuring technique	**radioisotopes**
gamma-spectrometry	impurities e.g. ^{7}Be, ^{60}Co, ^{124}Sb, …. and ^{85}Sr (yield tracer)
liquid scintillation spectrometry (LSC)	^{85}Sr (yield tracer) ^{89}Sr, ^{90}Sr, ^{90}Y and ^{90}Y (via Cherencov counting)
ICP-OES or MS	stable Sr for yield determination (already present in the sample or via added carrier)

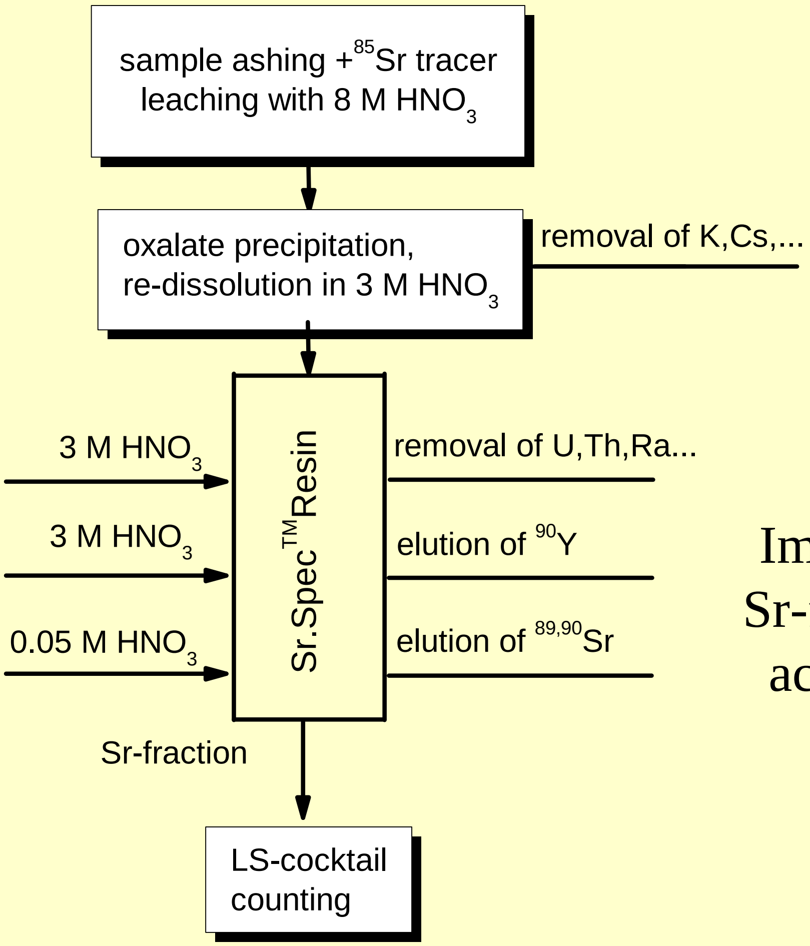
Figures 4 and 5

Acid dependency of k' for various ions at 23-25°C.
Sr Resin



Horwitz (HP199)

Schematic illustration of the separation procedure (soil, mud, grass)

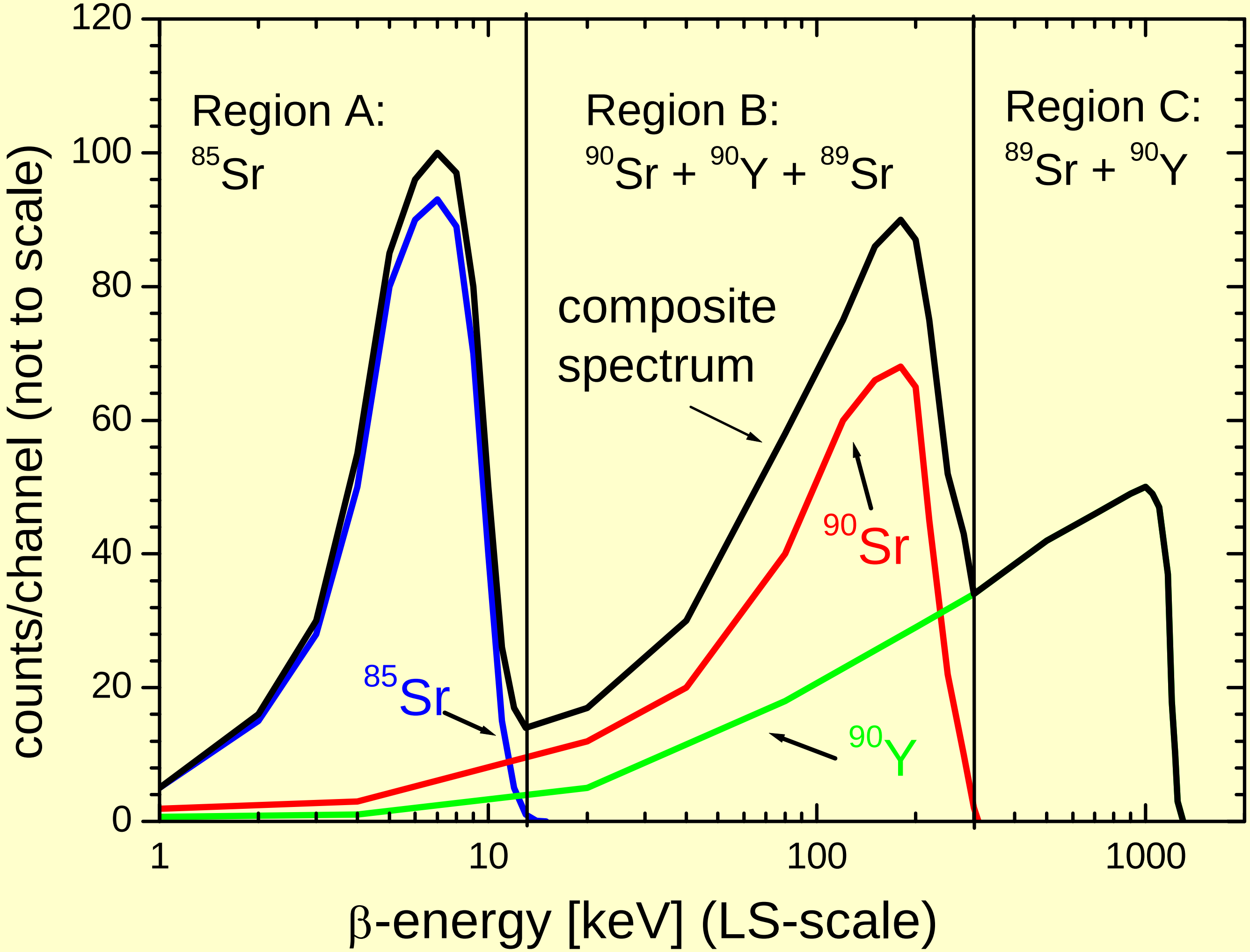


Important: dissolution of the Sr-fraction in toluene sulfonic acid: e.g. $C_6H_5CH_2SO_2NH_2$

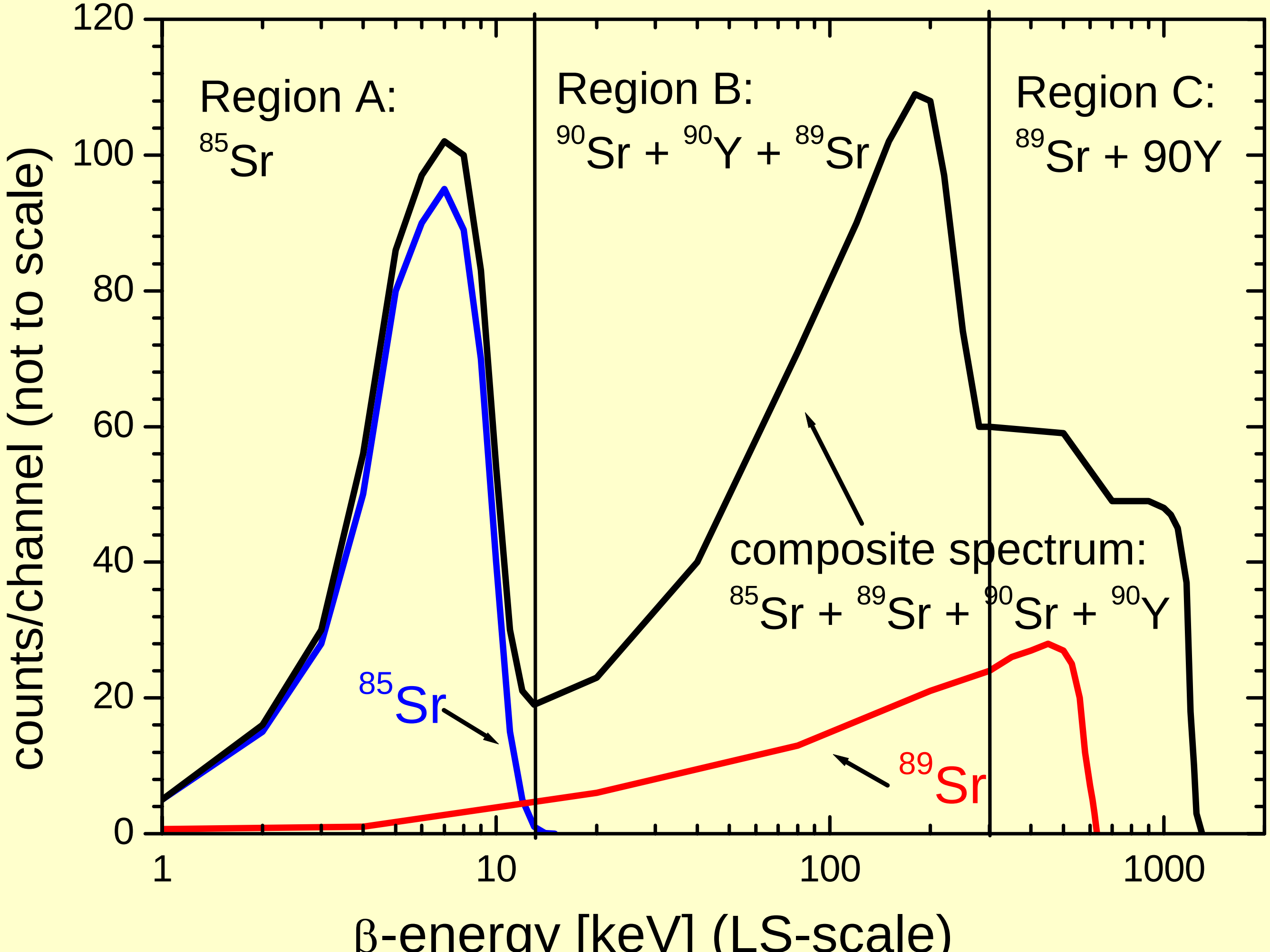
Radioactive Sr-isotopes, decay energies and half-lives

Sr-90	β^-, e_{max} = 550 keV	$T_{1/2}$ = 29 years
Y-90	β^-, e_{max} = 2.3 MeV	$T_{1/2}$ = 64 hours
Sr-89	β^-, e_{max} = 1.5 MeV	$T_{1/2}$ = 50.5 days
Sr-85	ec, γ, e_{max}(LS-photons): 10 keV, γ: 514 keV	$T_{1/2}$ = 65 days

three region window setting: single isotope and composite spectrum



three region window setting: single isotope and composite spectrum



Solving the problem (i.e. 4 isotopes in 1 spectrum)

$$N(t)_m^A = N(t)_{85}^A + N(t)_{89}^A + N(0)_{90Sr}^A + N(t)_{90Y}^A \qquad (1)$$

$$N(t)_m^B = N(t)_{85}^B + N(t)_{89}^B + N(0)_{90Sr}^B + N(t)_{90Y}^B \qquad (2)$$

$$N(t)_m^C = N(t)_{85}^C + N(t)_{89}^C + N(0)_{90Sr}^C + N(t)_{90Y}^C \qquad (3)$$

$$A_i = N_i / \varepsilon_i$$

$$A(t)_{90Y} = A(0)_{90Sr} \cdot \left(1 - e^{-\lambda_{90Y} \Delta t_m}\right)$$

$$f_1 = \left(1 - e^{-\lambda_{90Y} \Delta t_m}\right)$$

Solving the problem (i.e. 4 isotopes in 1 spectrum)

$$N(t)_m^A = A(t)_{85} \cdot \varepsilon_{85}^A + A(t)_{89} \cdot \varepsilon_{89}^A + A(0)_{90Sr} \cdot \varepsilon_{90Sr}^A + A(0)_{90Sr} \cdot f_1 \cdot \varepsilon_{90Y}^A$$

$$N(t)_m^B = A(t)_{85} \cdot \varepsilon_{85}^B + A(t)_{89} \cdot \varepsilon_{89}^B + A(0)_{90Sr} \cdot \varepsilon_{90Sr}^B + A(0)_{90Sr} \cdot f_1 \cdot \varepsilon_{90Y}^B$$

$$N(t)_m^C = A(t)_{89} \cdot \varepsilon_{89}^C + A(0)_{90Sr} \cdot f_1 \cdot \varepsilon_{90Y}^C$$

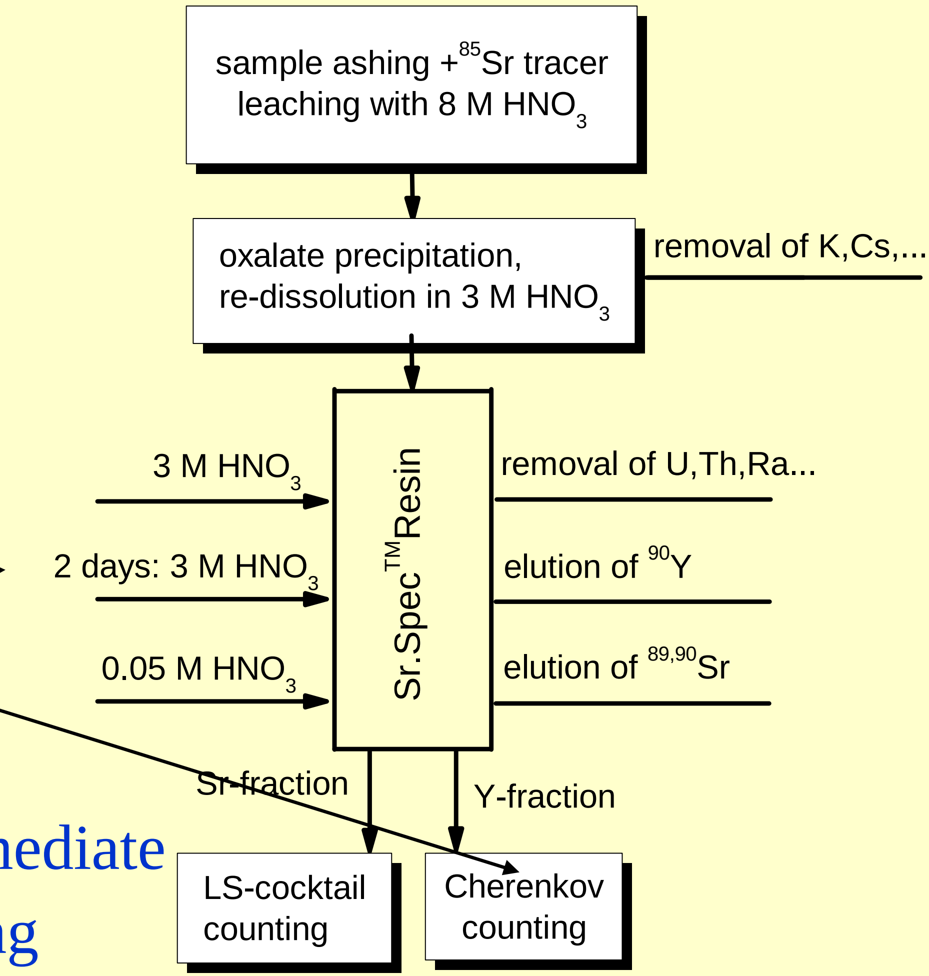
$$A(0)_{85,89} = A(t)_{85,89} \cdot e^{\lambda_{85,89} \cdot \Delta t_m}$$

$$Y_{chem} = \frac{A(0)_{85}}{A(0)_{85,add}}$$

$$A(ref)_{89,90} = A(0)_{89,90} \cdot e^{\lambda_{89,90} \cdot \Delta t_{ref}} \cdot \frac{1}{Y_{chem} \cdot V_{samp}}$$

Compilation of counting sensitivities (E^2/B) for three Perkin Elmer LS-spectrometers

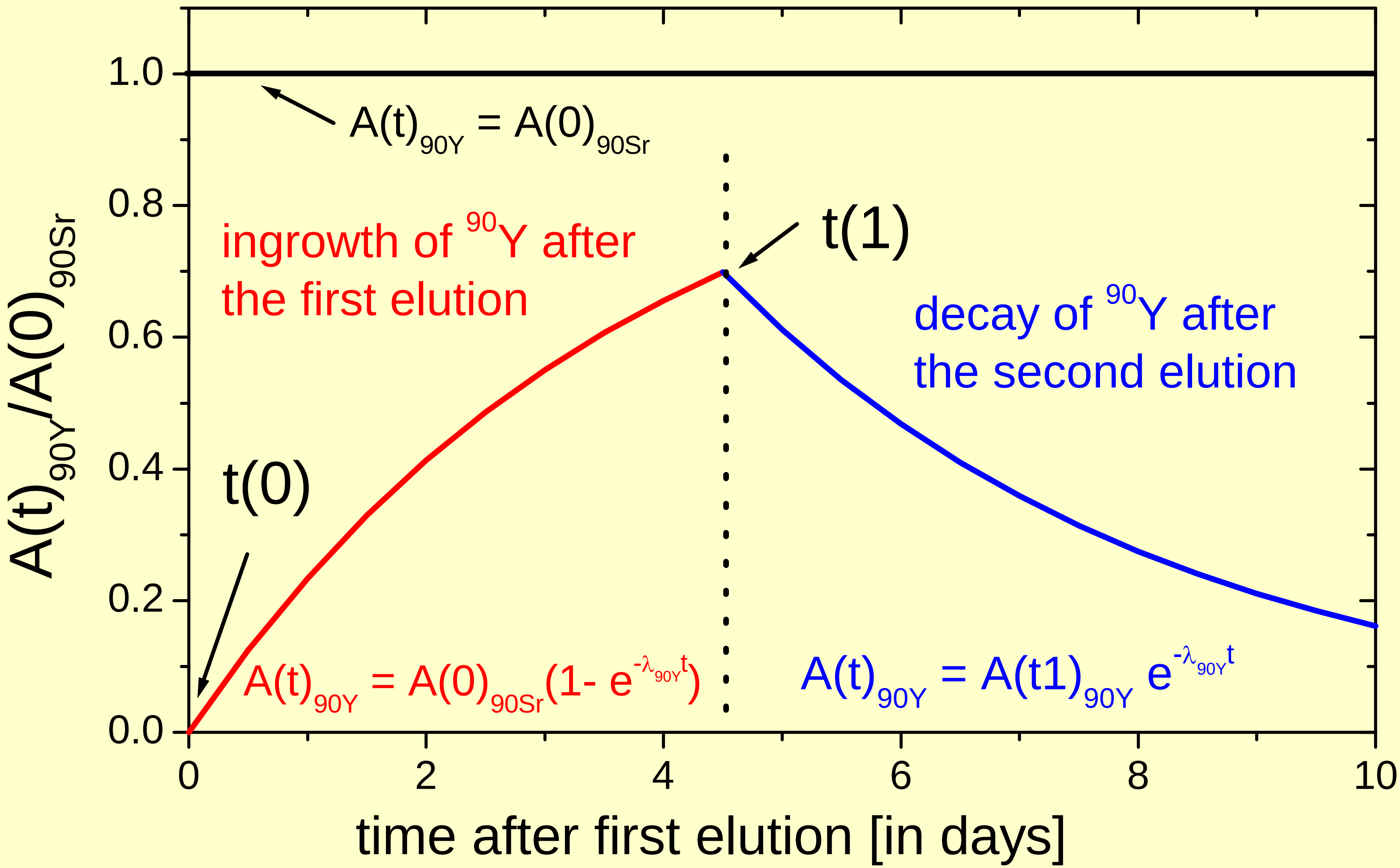
Isotope		TriCarb 2200		TriCarb 2550		TriCarb 2770	
(count mode)	Counting window	normal	low level	normal	low level	normal	low level
Sr-85	A	490	410	610	540	360	290
Sr-89	B + C	480	150	520	190	1220	530
Sr-90	B	600	410	630	400	1760	1000



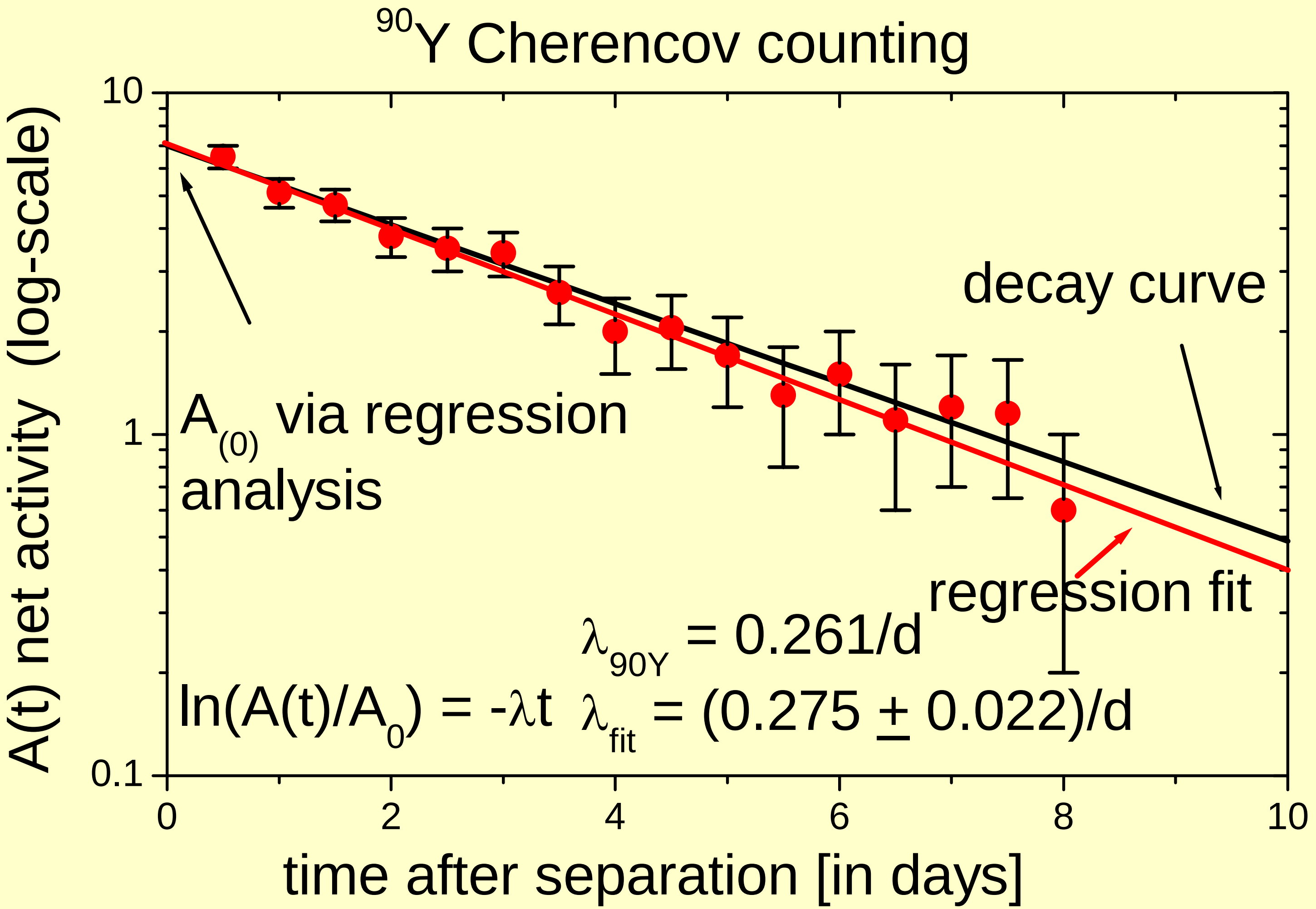
$^{89}Sr >> {}^{90}Sr$

$^{89}Sr << {}^{90}Sr$: immediate Cherenkov counting

Solving the problem for cases with $^{89}Sr >> {}^{90}Sr$

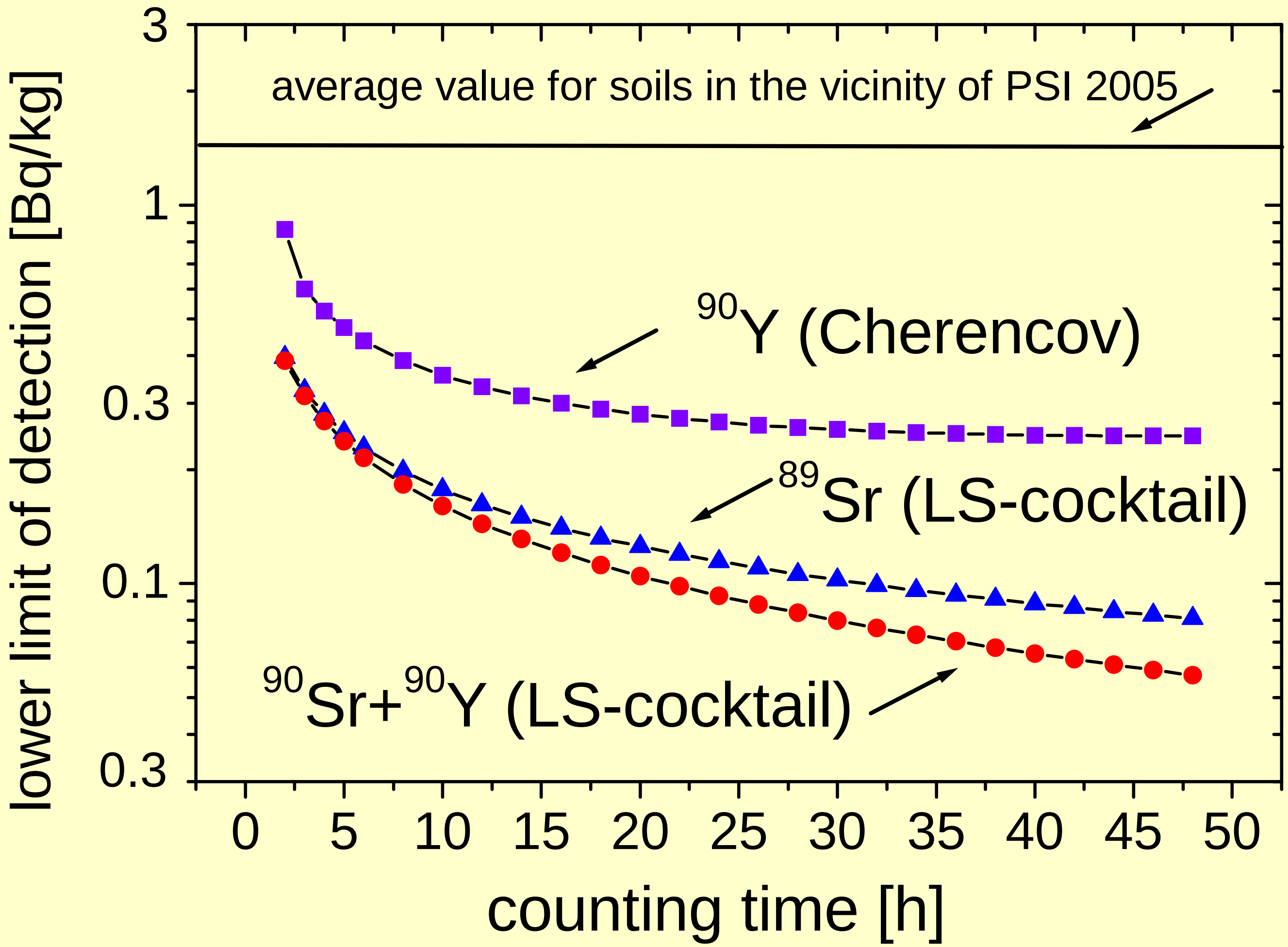


Solving the problem for cases with $^{89}Sr >> {}^{90}Sr$



Calculating MDA's

$$MDA\,[Bq/kg] = \frac{2.71 + 2 \cdot K\sqrt{2 \cdot I_0}}{t \cdot Y_{chem} \cdot \varepsilon \cdot V_s}$$



Results of ^{89}Sr + ^{90}Sr radioisotope intercomparison exercises (aqueous samples)

Organisation (year)	^{89}Sr result PSI	PSI/(Lab mean)	^{90}Sr result PSI	PSI/(Lab mean)
PTB Bruns. (2005) lab code (75) (a)	1.7 $\pm$ 0.3	0.99	2.6 $\pm$ 0.2	0.98
NPL London (2005) lab code (60) (b)	13.6 $\pm$ 1.2	1.06	10.2 $\pm$ 0.7	1.00
PTB Bruns. (2006) lab code (15) (a)	2.8 $\pm$ 0.4	1.11	2.0 $\pm$ 0.2	1.01

Units: (a) Bq/kg, (b) Bq/g

Conclusions

- Spectrum separation between ^{89}Sr and ^{90}Sr can be easily performed using highly aromatic cocktails and LS-spectrometry
- The chemical yield or recovery can be simply obtained measuring added low energetic ^{85}Sr in the same LS-spectrum besides ^{89}Sr and ^{90}Sr
- The analytical solution of the three isotope system using three window settings yields a clear result because the coefficients (i.e. efficiencies) of all components (i.e. Sr-isotopes) are different in each counting window