

Intercomparison samples & γ -ray analyses in STUK

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Roy Pöllänen, Sinikka Virtanen
Environmental Radiation Surveillance and Emergency Preparedness

Basic ideas of the intercomparison:

- Not too easy (not only e.g. ^{137}Cs) but at the same time not too complicated case.
- Possibility of using different analysis methods (not only γ -spec.).
- Samples contain natural and some artificial radionuclides.

Selection of nuclides to be reported by using γ -spec.:

- Covers a wide range of energies (46 keV – 1460 keV) → validity of the efficiency calibration.
- Some difficult-to-determine nuclides present in the samples.

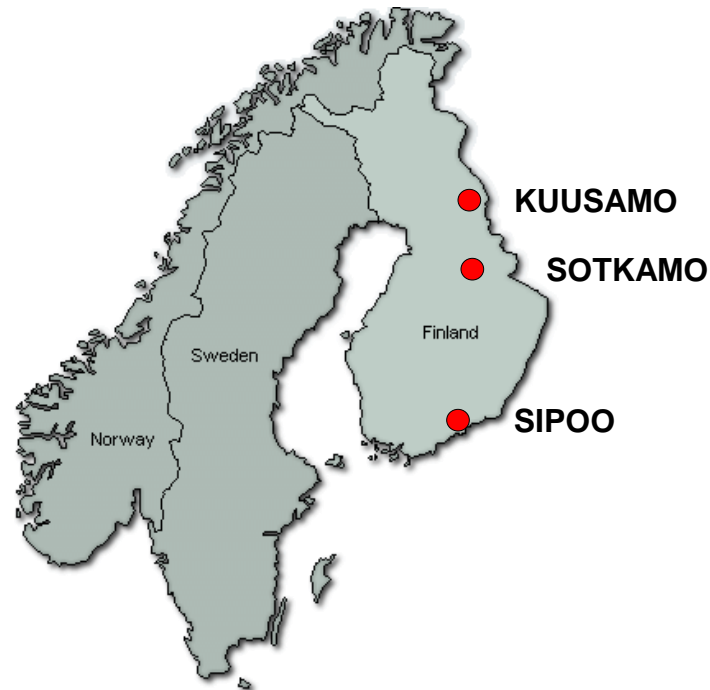
Same samples are used in the CAMNAR NKS-project.

Same samples are also analysed under the umbrella of CBSS labs.

The samples are not certified!

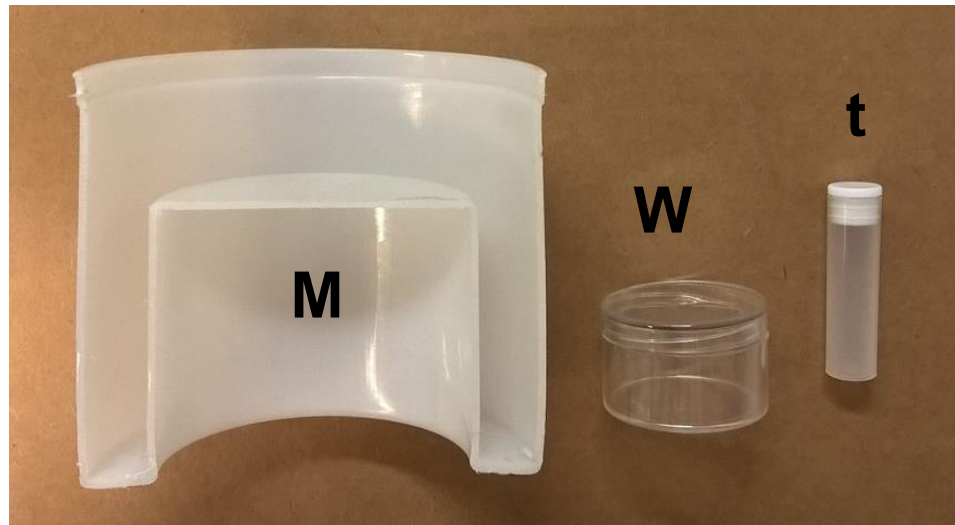
1. Two sample types

- Lake sediment samples from Kuusamo and Sotkamo.
- Water samples from a drilled well in Sipoo.
- (see separate presentation by Sinikka)



2. γ -ray measurements in the lab

- γ -ray measurements were done by using 4 spectrometers (and several measurements) including a well-type detector.
- Measurement geometries:
 - Marinelli (M)
 - Williams (W)
 - test tube (t)



- Final results were obtained by using the BeGe detector labelled as "B6" and W-geometry.



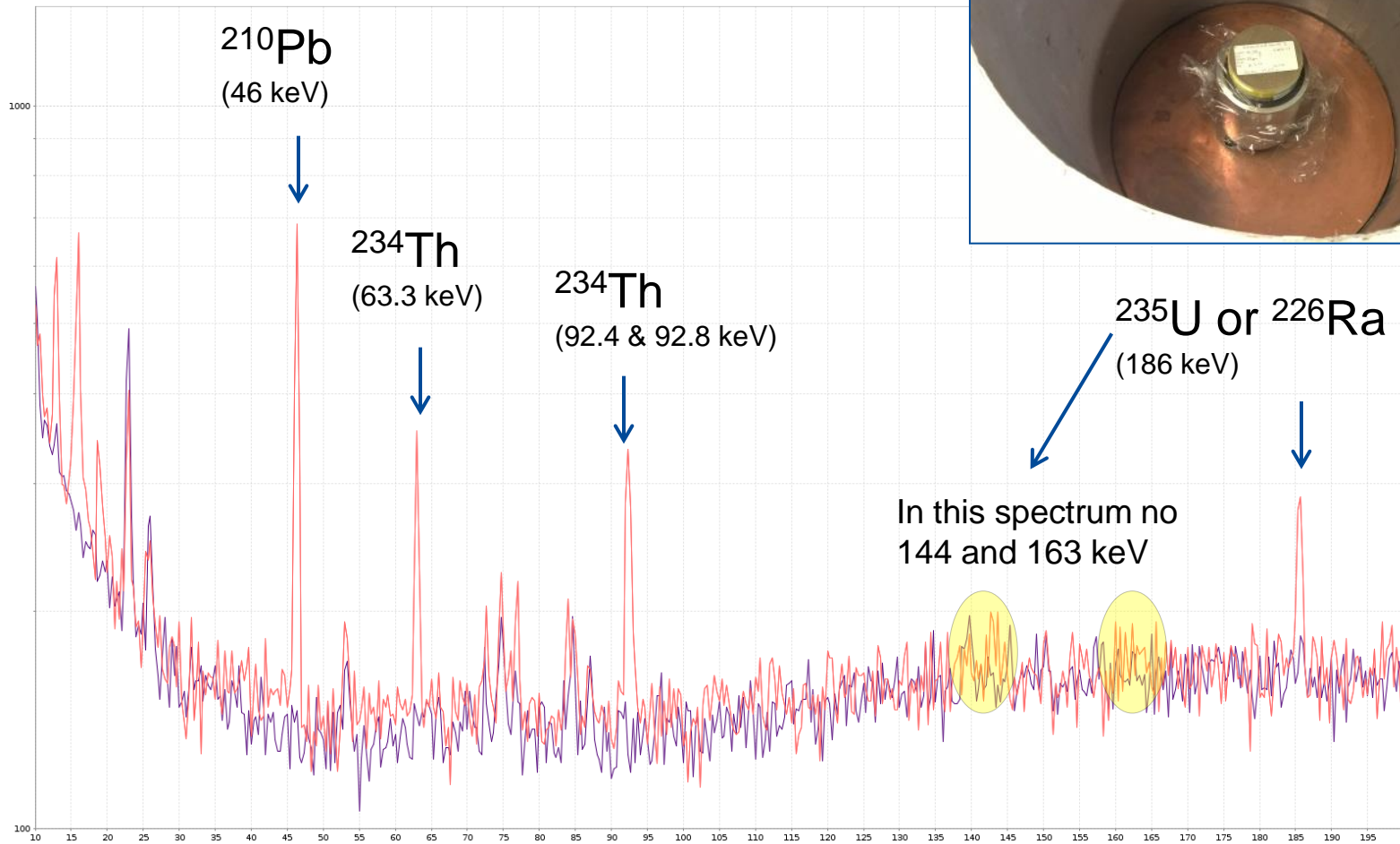
3. Results from the water sample

- Reporting format:

Contact person and organization:		Sinikka Virtanen STUK	
Nuclide	Activity (Bq/L)		Uncertainty (k=1)
Pb-210			
Th-234 (U-238)			
Ra-226			
U-235			
Pa-234m (U-238)			

- ^{238}U can be determined by using its daughters $^{234\text{m}}\text{Pa}$ and ^{234}Th and assuming secular equilibrium (but questionable in water and water sediment samples !!!).
- ^{226}Ra can be determined by using its daughters ^{214}Pb and ^{214}Bi and assuming secular equilibrium → vacuum packaging and waiting for ~ 3 weeks ($t_{1/2 \text{ Rn-222}} = 3.8 \text{ d}$).

Snapshot from the screen



Gammas from ^{234}Th (24.10 d 3)

E_γ (keV)	I_γ (%)	Decay mode
63.29 2	4.8 5	β^-
92.38 1	2.81 15	β^-
92.80 2	2.77 15	β^-
112.81 5	0.277 20	β^-
83.30 5	0.079 4	β^-
62.86 2	0.021 3	β^-
87.02 6	0.019 3	β^-
73.92 2	0.0172 13	β^-
184.8	0.013 7	β^-
108.00 5	0.0106 13	β^-
20.02 2	0.010 3	β^-
57.75 10	0.007 4	β^-
103.35 10	0.0042 13	β^-
29.49 2	0.00158 13	β^-
10		β^-
73.92		β^-
92.00 5		β^-
103.71 6		β^-
132.9		β^-

$^{234\text{m}}\text{Pa}$ (1001 keV) was not detected → the only possibility was to use ^{234}Th in order to determine ^{238}U .

Gammas from $^{234\text{m}}\text{Pa}$ (1.17 m 3)

E_γ (keV)	I_γ (%)	Decay mode
1001.03 3	0.837 10	β^-
766.38 2	0.294 12	β^-
742.81 3	0.080 4	β^-
258.23 7	0.0728 4	β^-
786.27 2	0.0485 10	β^-

- Determination of ^{226}Ra was done from the vacuum-packaged sample and using the peaks of ^{214}Pb (352 keV) and ^{214}Bi (609 keV).
- The 144 keV and 163 keV peaks of ^{235}U were detected only in some measurements but can be used to verify results obtained from the 186 keV peak.

Gammas from ^{226}Ra (1600 y 7)

E_γ (keV)	I_γ (%)	Decay mode
186.211 13	3.59 6	α
262.27 5	0.0050 5	α
600.66 5	0.00049	α
414.60 5	0.00030	α
449.37 10	0.00019	α
34.8 16		α
187.10 20		α

Gammas from ^{235}U (7.038E+8 y 5)

E_γ (keV)	I_γ (%)	Decay mode
185.712 1	57.2 5	α
143.764 2	10.96 8	α
163.358 2	5.08 4	α
205.309 2	5.01 5	α
109.16 2	1.54 5	α
202.111 3	1.08 2	α
194.94 7	0.63 7	α

STUK's result for the water sample:

Nuclide	Activity (Bq/L)		Uncertainty (k=1)	
Pb-210	14.5		1.4	
Th-234 (U-238)	6		1	
Ra-226	0.59		0.17	
U-235	0.47		0.06	
Pa-234m (U-238)				



- Activity ratio $^{238}\text{U}/^{235}\text{U}$ in natural U is 21.5. We got ~ 13 . Is the result correct?
- Regarding secular equilibrium (such as $^{238}\text{U} \rightarrow ^{234}\text{Th} \rightarrow ^{234\text{m}}\text{Pa}$): Be careful !

Two separate measurements by using our new well detector (compared to the results in the previous slide):

Nuclide	Activity (Bq/L)	Measurement1	Measurement2
Pb-210	14.5	16.5 (13%)	16.1 (25%)
Th-234 (U-238)	6	3.9 (18%)	4.5 (18%)
Ra-226	0.59	-	-
U-235	0.47	0.33 (32%)	-
Pa-234m (U-238)	-	-	-



- U-235 determined using the 186 keV peak (and corrected by the above-mentioned proportion of Ra-226)
- Fairly good agreement!

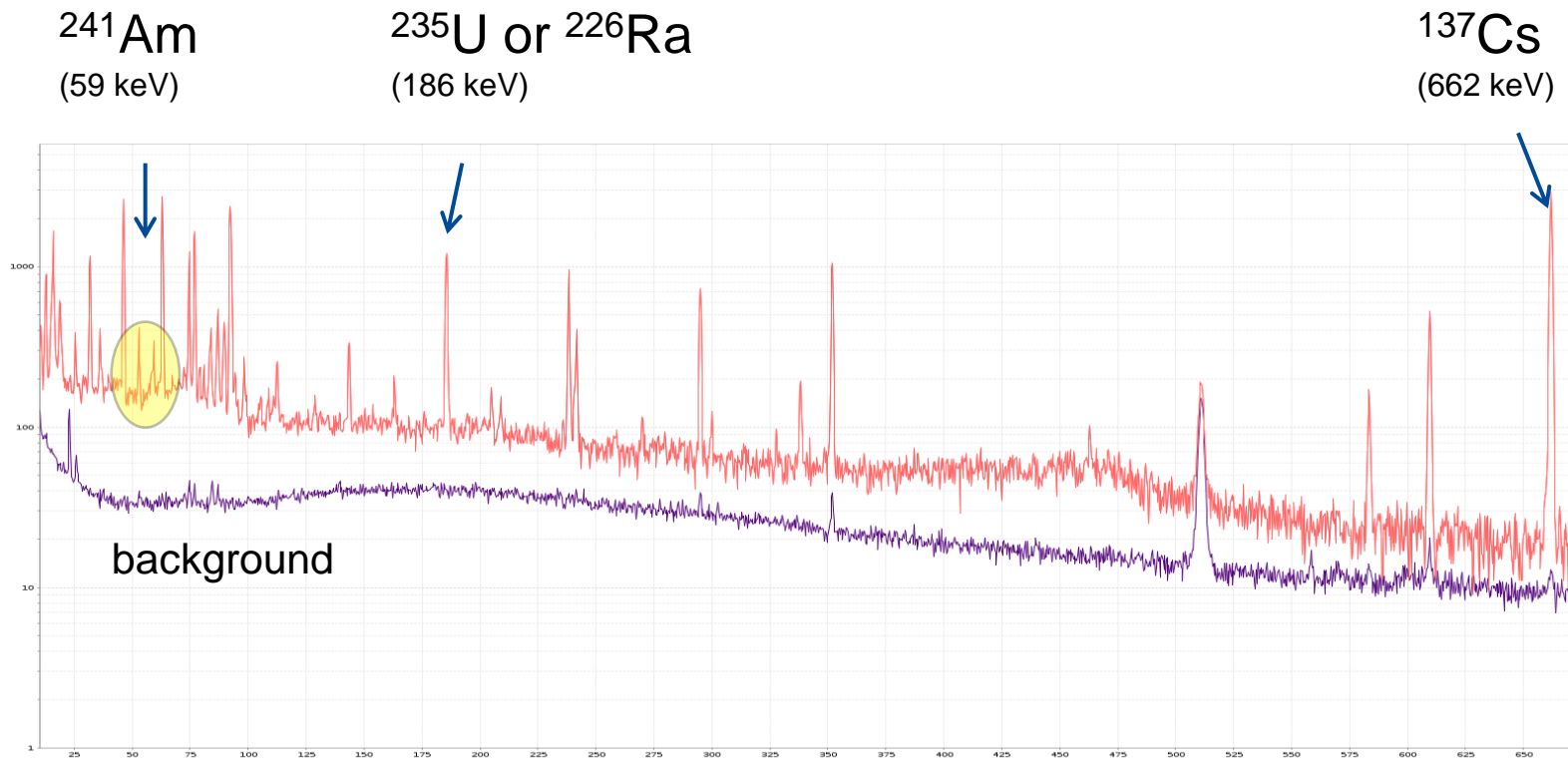
4. Sediment sample – homogeneity?

Sample processing

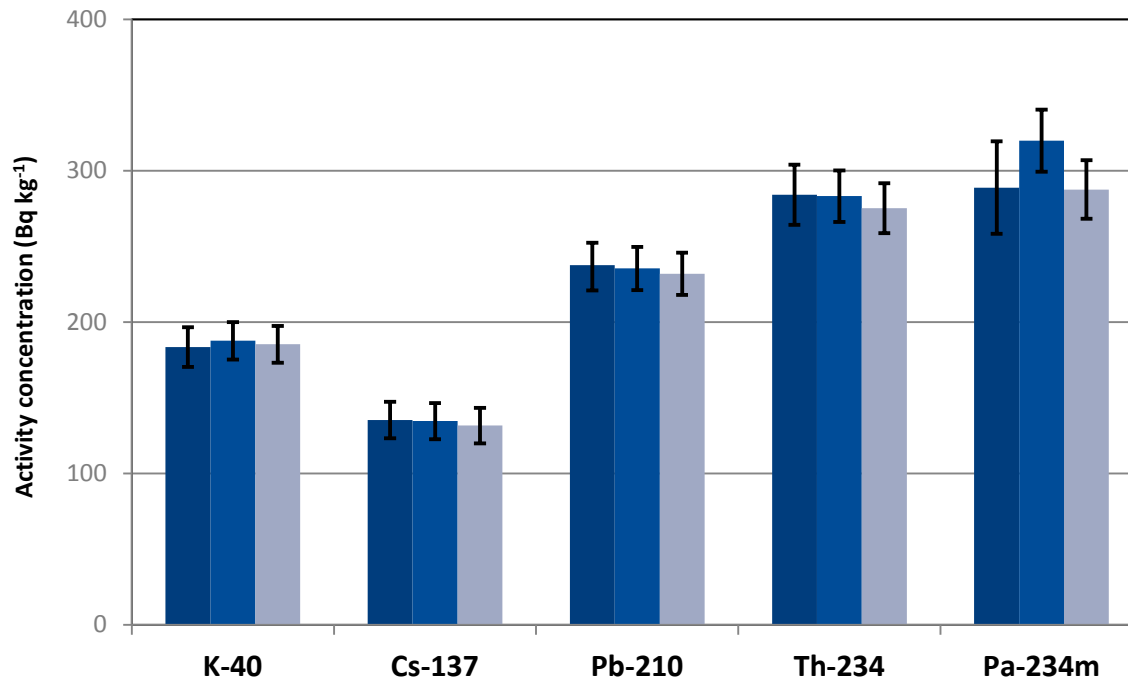
Maria Kaipainen and Sinikka Virtanen in action

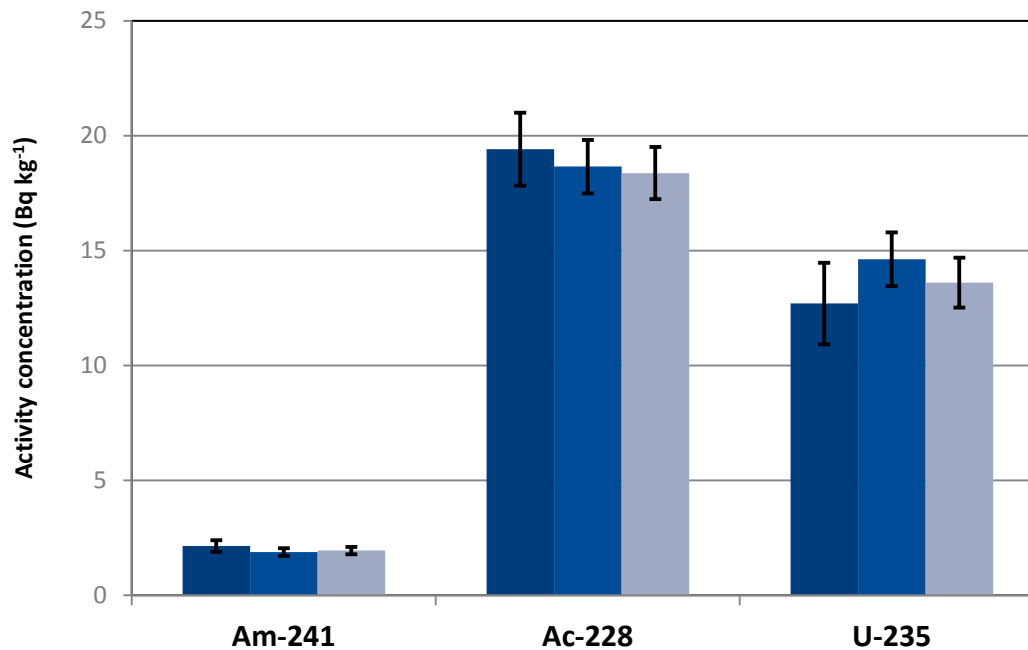


- Artificial nuclides (^{137}Cs and ^{241}Am from atmospheric nuclear tests) were present in this sample .



- The spectra were analyzed by Unisampo-Shaman and Gamma 99 software.
- Results from 3 subsamples (coverage factor $k=1$):





The sediment material was homogeneous.

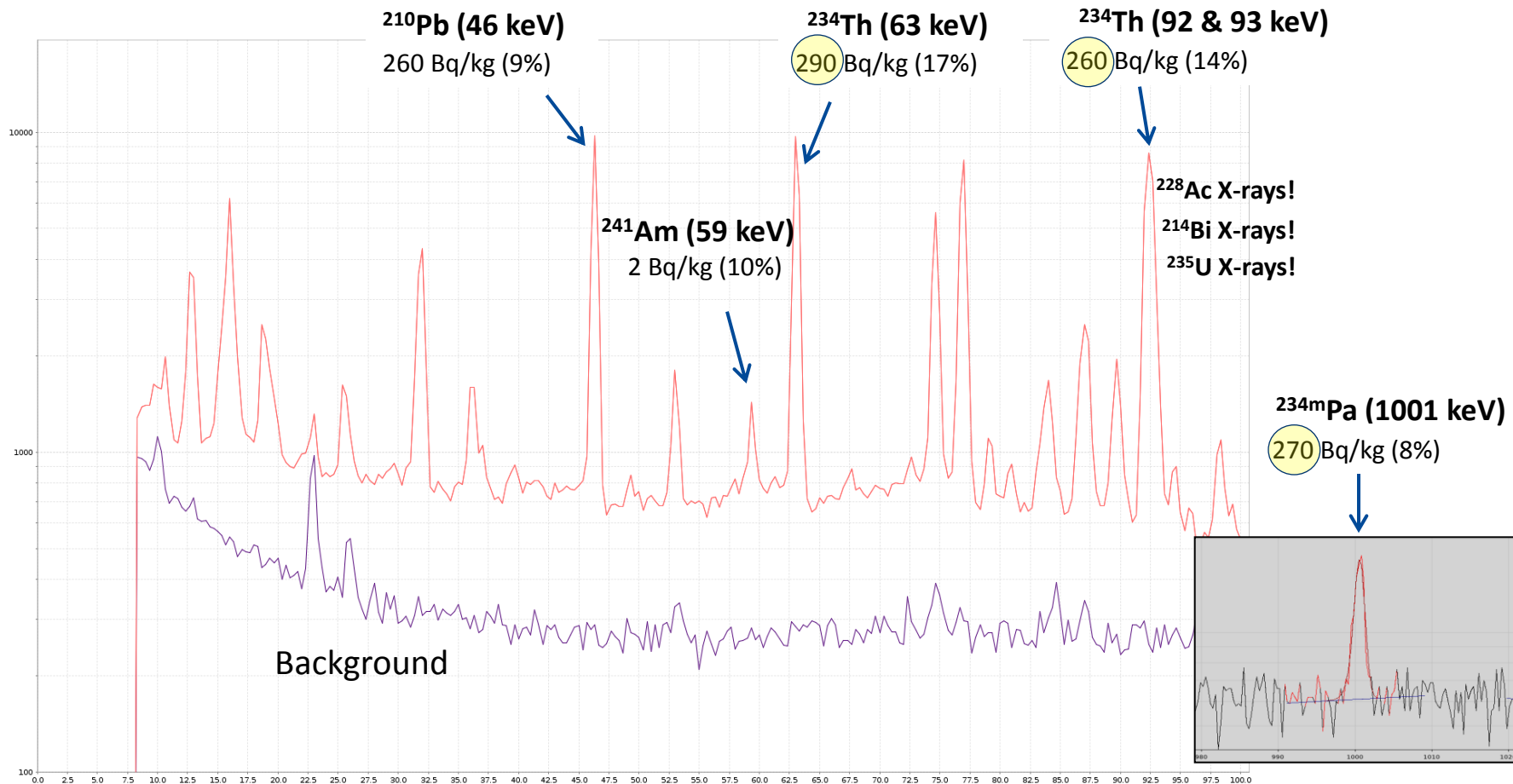
5. Results from the sediment sample

- Reporting format:

		Contact person and organization: Sinikka Virtanen STUK	
	Nuclide	Activity (Bq/kg)	Uncertainty (k=1)
"Easy" cases	Cs-137		
	Am-241		
	K-40		
	Pb-210		
Not necessarily so easy	Th-234 (U-238)		
	Ra-226		
	Ra-228		
	U-235		
	Pa-234m (U-238)		

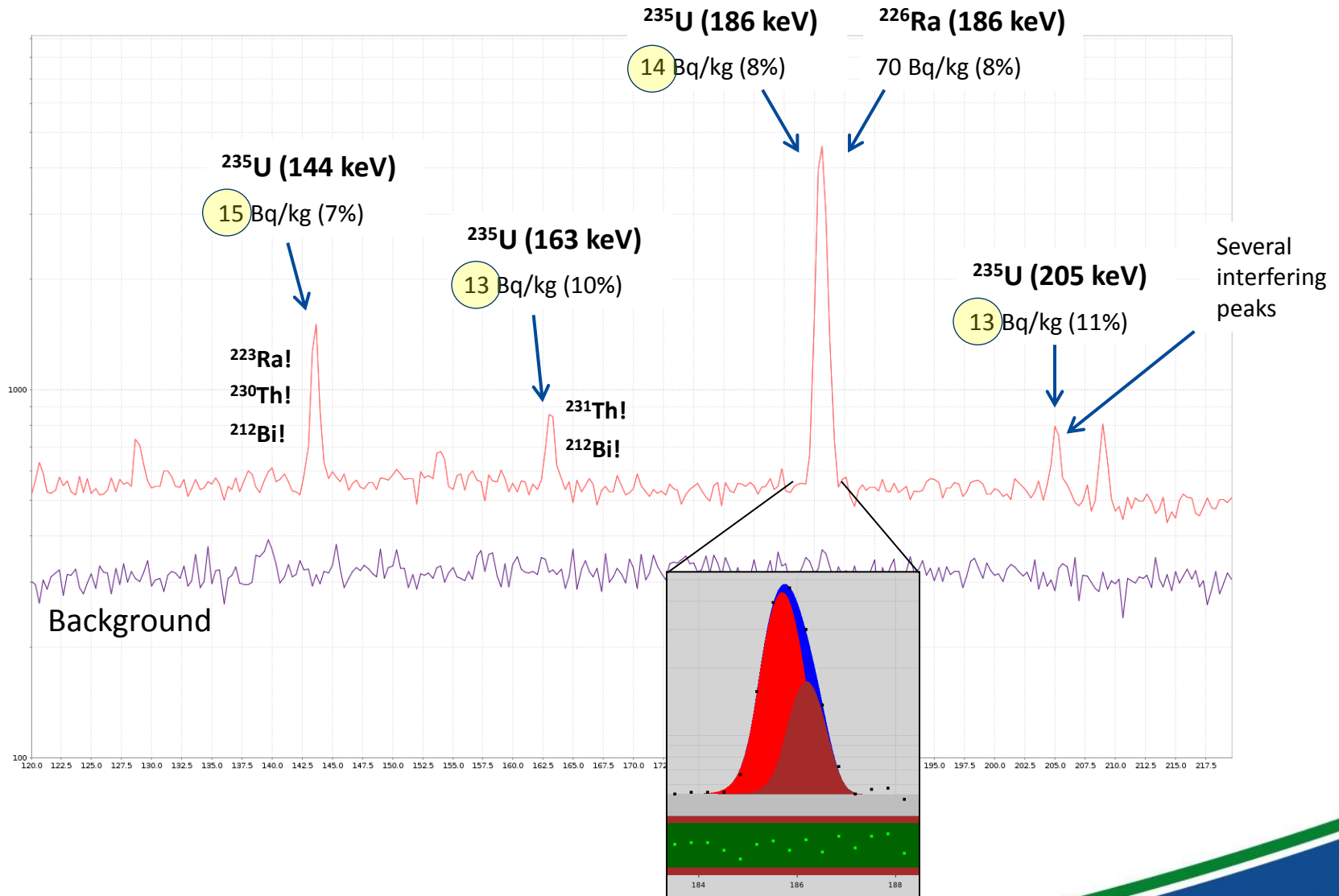
- > 80 peaks in the spectra (^{238}U , ^{235}U and ^{232}Th decay series).
- ^{228}Ra ($t_{1/2} = 5.8$ a) can be determined by using its daughter ^{228}Ac ($t_{1/2} = 6.2$ h) assuming secular equilibrium.

Data acquisition of 6 d by using a BeGe detector (8-100 keV)

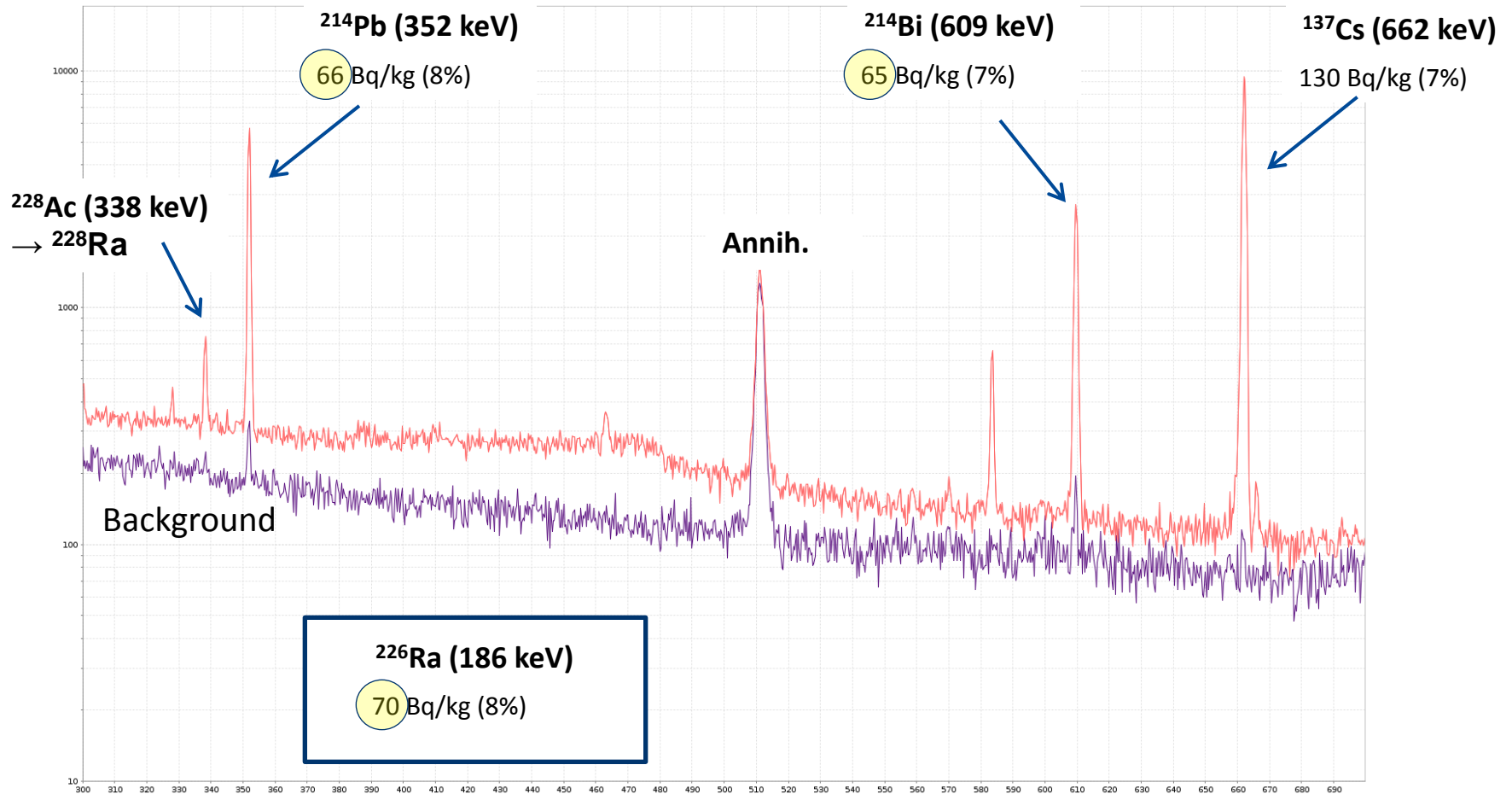


^{238}U determined by using its daughters ^{234}Th and $^{234\text{m}}\text{Pa}$.

Data acquisition of 6 d by using a BeGe detector (120-220 keV)

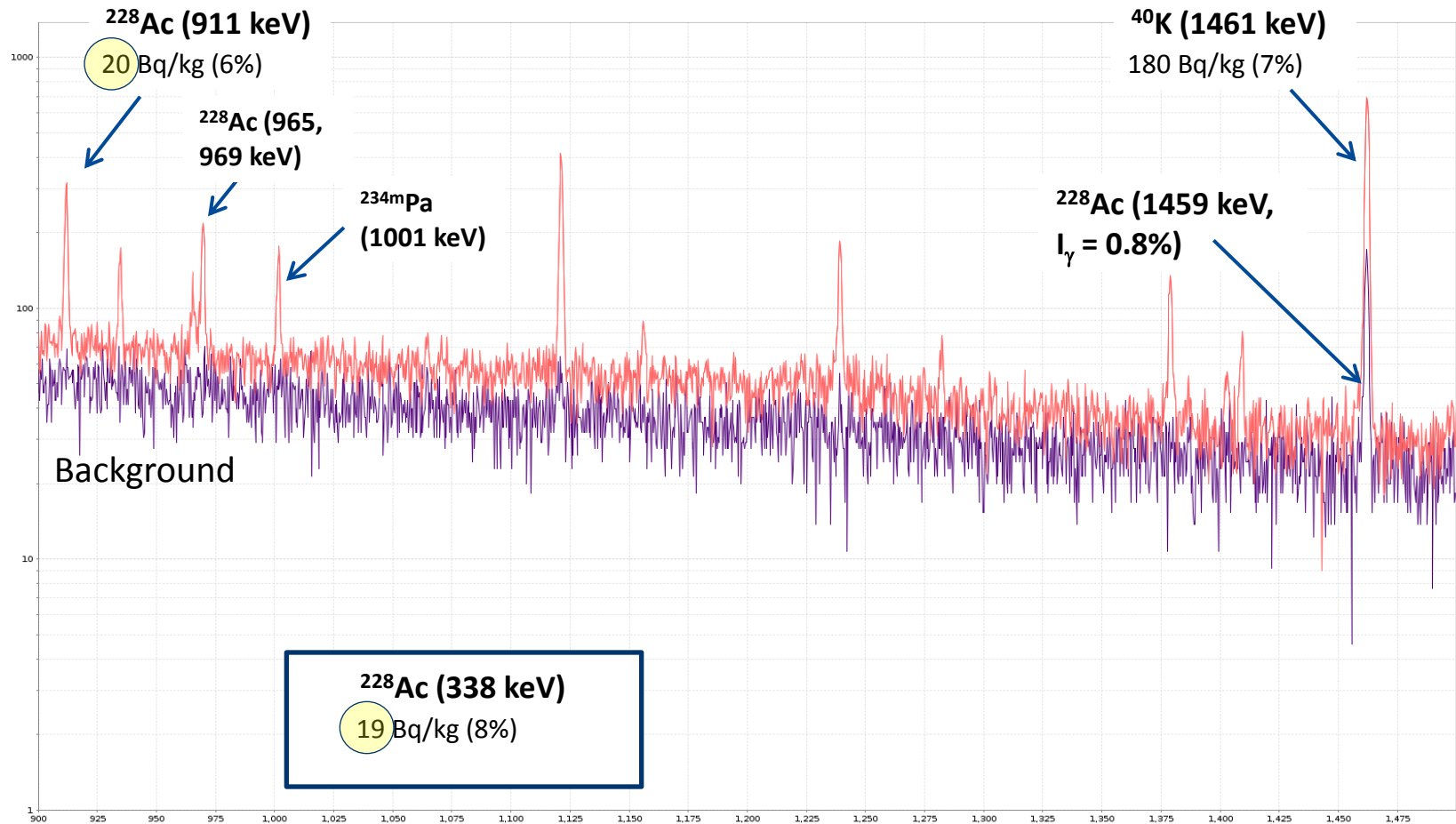


Data acquisition of 6 d by using a BeGe detector (300-700 keV)



Vacuum-packaged sample! ^{226}Ra can be obtained from ^{214}Pb & ^{214}Bi .

Data acquisition of 6 d by using a BeGe detector (900-1500 keV)



^{228}Ra determined by using its daughter ^{228}Ac .

STUK's result for the sediment sample (obtained from the 6-d measurement of the vacuum-packaged sample):

Nuclide	Activity (Bq/kg)		Uncertainty (k=1)
Cs-137	129		9
Am-241	2.05		0.20
K-40	181		12
Pb-210	256		22
Th-234 (U-238)	280		30
Ra-226	67		5
Ra-228	19.7		1.5
U-235	14		1.0
Pa-234m (U-238)	270		21

- Activity ratio $^{238}\text{U}/^{235}\text{U}$ in natural U is 21.5.
We got 19.6 ± 2.2 ($k=1$)

6. Conclusions

- Reporting the results: be always careful when using the assumption of secular equilibrium.
- The phenomena/assumptions in the background of the analyses must be identified → other analysis methods, e.g. mass spectrometry in the case of ^{238}U , are sometimes necessary.