University of Bologna
School of Engineering and Architecture
Department of Industrial Engineering

National Technical University of Athens
Nuclear Engineering Department

ANALYSIS OF THE VERTICAL DISTRIBUTION AND THE SIZE FRACTIONATION OF NATURAL AND ARTIFICIAL RADIONUCLIDES IN THE SOIL IN THE VICINITY OF HOT SPRINGS

S. Padovani, I. Mitsios, M. Anagnostakis, D. Mostacci

CHERNE 2018 -14th Workshop on European Collaboration in Higher Education on Radiological and Nuclear Engineering and Radiation Protection

- ^{210}Pb is a very useful radioactive isotope for environmental studies. Its vertical distribution in the soil, combined with that of ^{226}Ra, may be used in order to assess the proportion of ^{222}Rn that can escape from a certain soil. These data are very useful in order to assess the annual dose to human.
- In addition, ^{210}Pb and its decay product ^{210}Po appear in the group of the most highly toxic radioisotopes, responsible for most of the internal radiation dose.
- Several studies[1][2][3] report fluctuations of natural radioactivity near hot springs. Generally, activity concentrations of ^{210}Pb and ^{226}Ra are higher in these areas. High values of radioactivity may be a source of potential health problems for people who regularly visit or are working at hot spring facilities.

[1] Beitollahi M, Ghiassi-Nejad M, Esmaeli A, Dunker R. 2007. Radiological studies in the hot spring region of Mahallat, Central Iran. Radiat Prot Dosim 123(4), 505–508

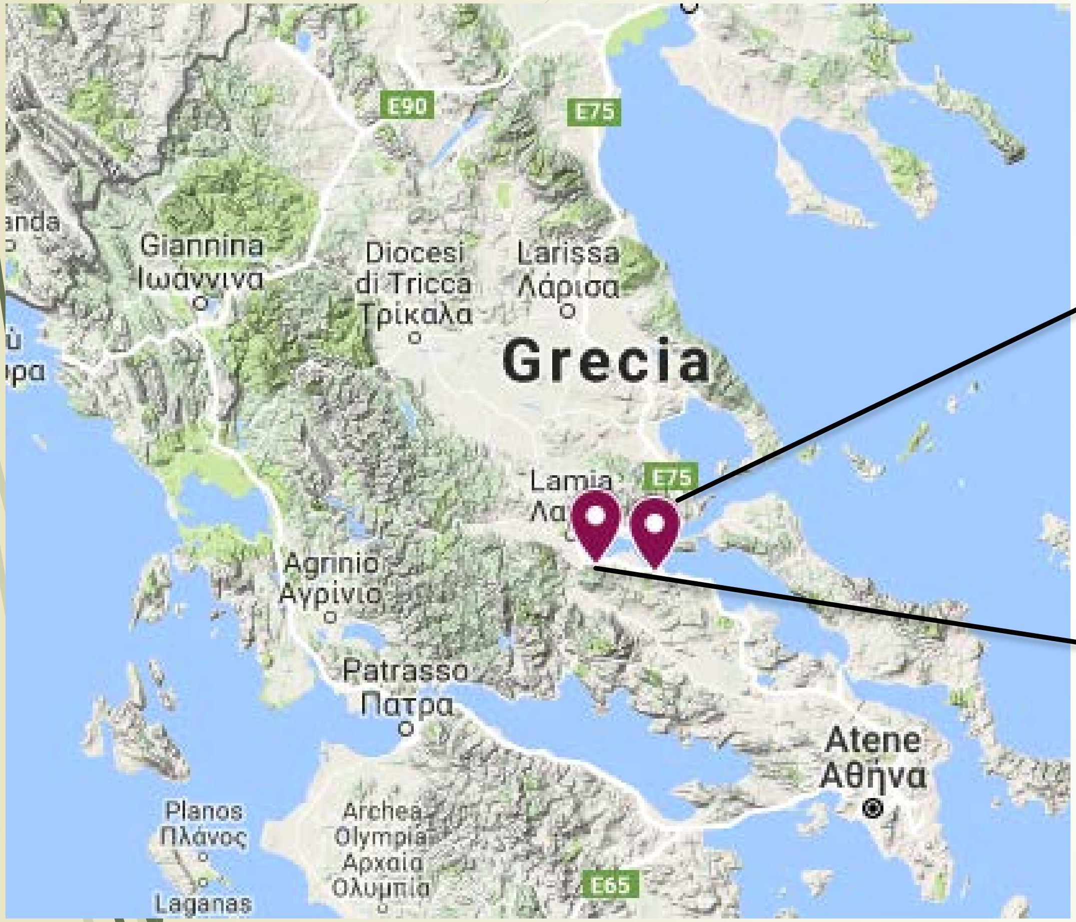
[2] Ramadan KA, Seddeek MK, Elnimr T, Sharshar T, Badran HM. 2011. Spatial distribution of radioisotopes in the coast of Suez Gulf, Southwestern Sinai and the impact of hot springs. Radiat Prot Dosim 145(4), 411–420

[3] Danali-Cotsaki S, Margomenou-Leonidopoulou G. 2003. ^{222}Rn in Greek spa waters: correlation with rainfall and seismic activities. Health Phys 64(6)

1. COLLECTION OF SOIL SAMPLES
2. SAMPLES PREPARATION FOR GAMMA-RAY SPECTROMETRY
3. MONTE CARLO SIMULATION FOR THE CALIBRATION OF THE LEGe DETECTOR
4. ACTIVITY CALCULATIONS
5. INTERPRETATION OF THE RESULTS

1. COLLECTION OF SOIL SAMPLES

SAMPLING LOCATIONS



Kamena Vourla

Vertical distribution of radionuclides

Thermopylae

Size fractionation of radionuclides

SAMPLING LOCATIONS

Kamena Vourla

- Presence of hot springs and an old spa facility
- Flat terrain
- Apparently undisturbed
- Non-rocky soil

➡ Collection of **SOIL CORES** (21 sections)

Kamena Vourla collection site

Thermopylae

- Presence of hot springs
- Small sulfurous stream that overflows the area
- Ground composed of a mixture of rocks and dust
- Complex environment

➡ Collection of **SURFACE MATERIAL** (7 fractions) and **WATER**

Thermopylae collection site and stream

SAMPLING PROCEDURE

Kamena Vourla soil cores

- Hollow metallic tube driven into the ground
- Extraction and opening of the tube
- Measurement and cutting of the core into 1 cm thick sections – total of 21 sections, 0-22 cm
- Collection of 3 soil cores, thus every sample is a mixture of soil from three different cores from the same field

Thermopylae surface material

- Superficial soil from different spots in the area ~2.5 kg

Thermopylae water

- Water from the sulfurous stream ~2 L

2. SAMPLES PREPARATION FOR GAMMA-RAY SPECTROMETRY

SAMPLES PREPARATION

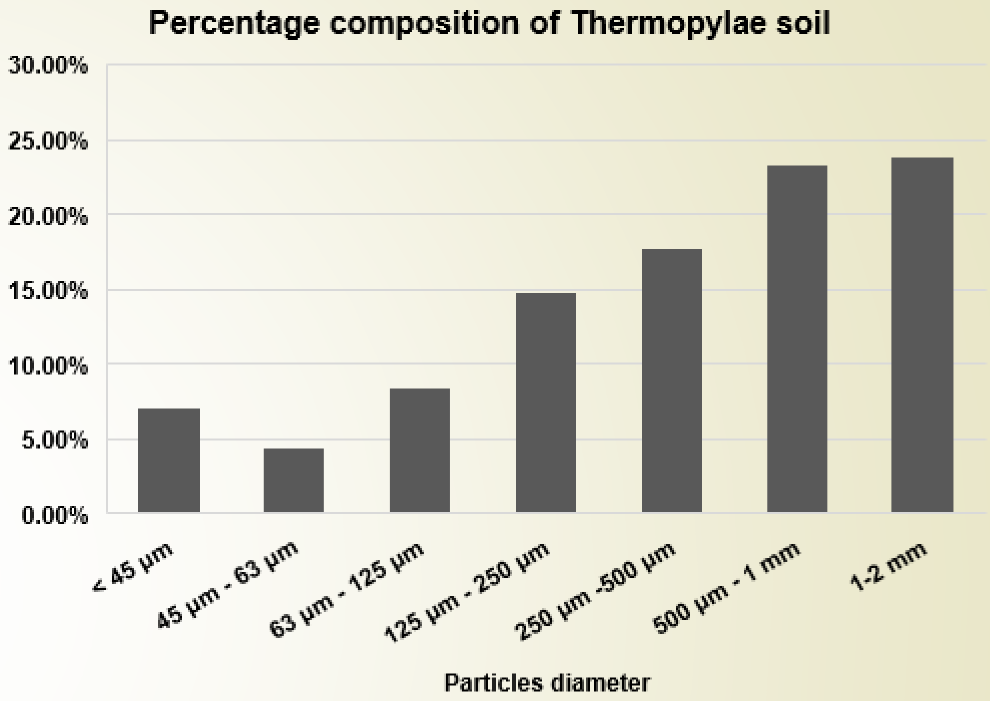
Kamena Vourla core sections

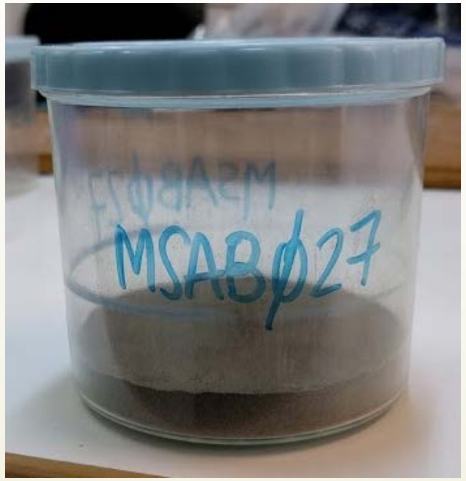
- 24 hours air-drying
- Manual sieving (1x1 mm mesh)
- Coding: from MSAB001 to MSAB021
- **Petri geometry** (vol. 25.8 cm^3)
- Weighing
- Mean humidity content, measured from 5 different soil sections: 2.9 %
- Sealing with silicon
- Painting with epoxy resin

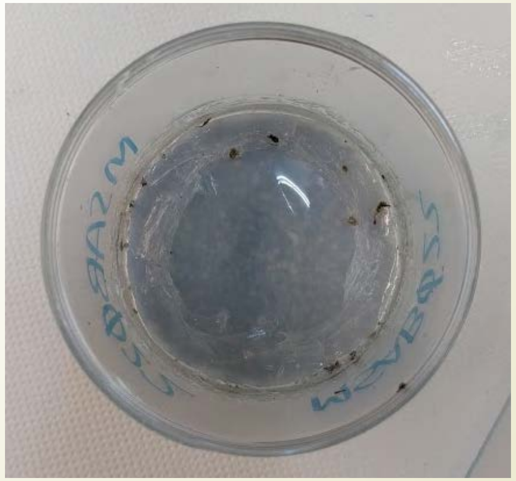
SAMPLES PREPARATION

Thermopylae superficial soil fractions

- 24 hours air-drying
- Sieving with a sieving machine for 20 mins at 2 mm/g
- Coding: from MSAB022 to MSAB028 – tot 7 samples
- **Geometry «8»** (vol. 40.7 cm^3)
- Weighing
- Moisture content measurement
- Sealing with silicon
- Painting with epoxy resin



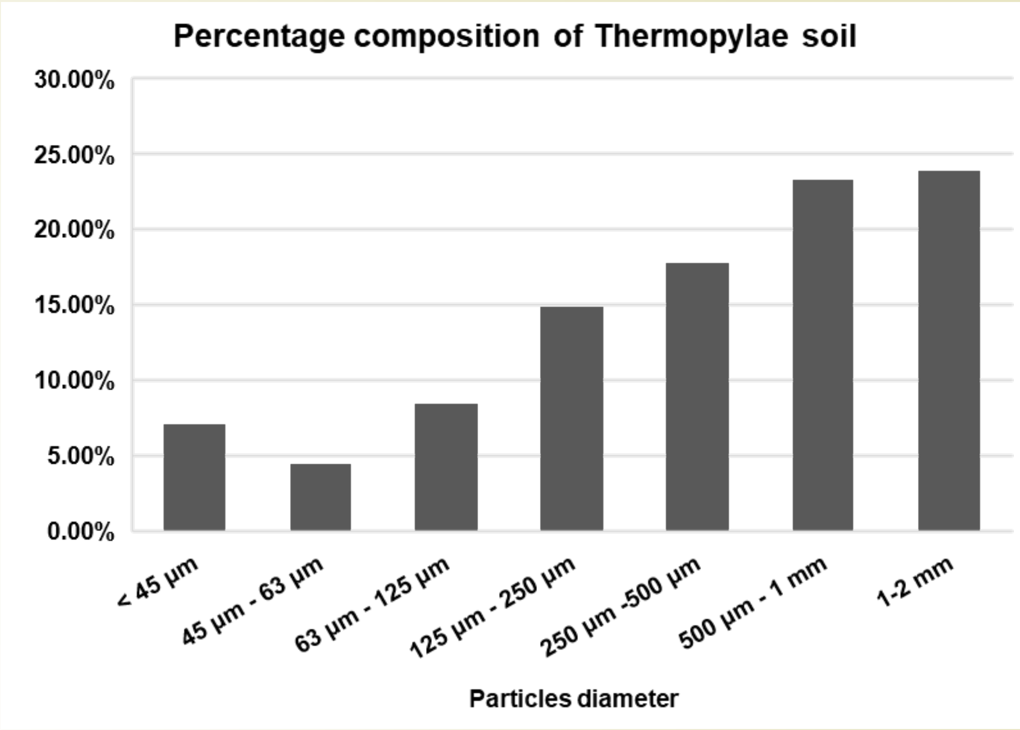




SAMPLES PREPARATION

Thermopylae superficial soil recomposed sample

- 600 g of superficial soil were recomposed
- Building of MSAB029 in **geometry «2»** (vol. 280.9 cm^3)
- Building of MSAB030 in **geometry «8»**



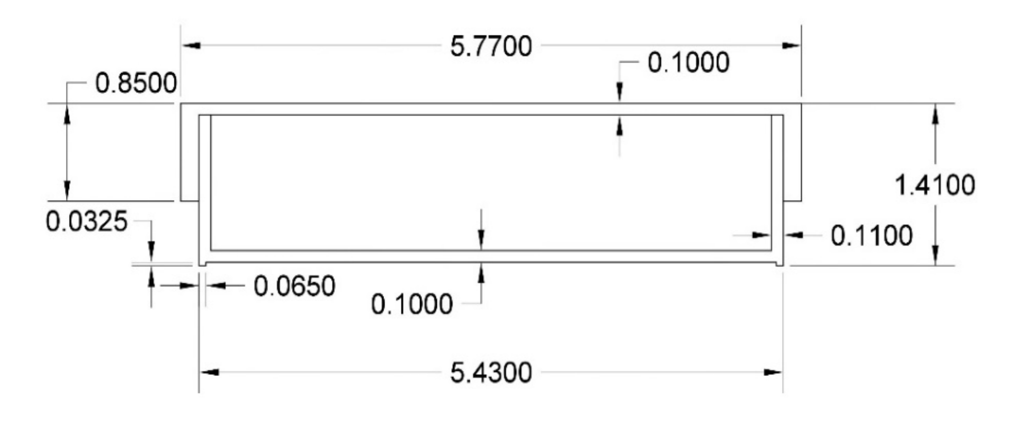
Thermopylae water sample

- Filtration of water using a Whatman 42 Filter Paper, diam. 185 mm
- **Geometry «2»**
- Sealed
- Painted

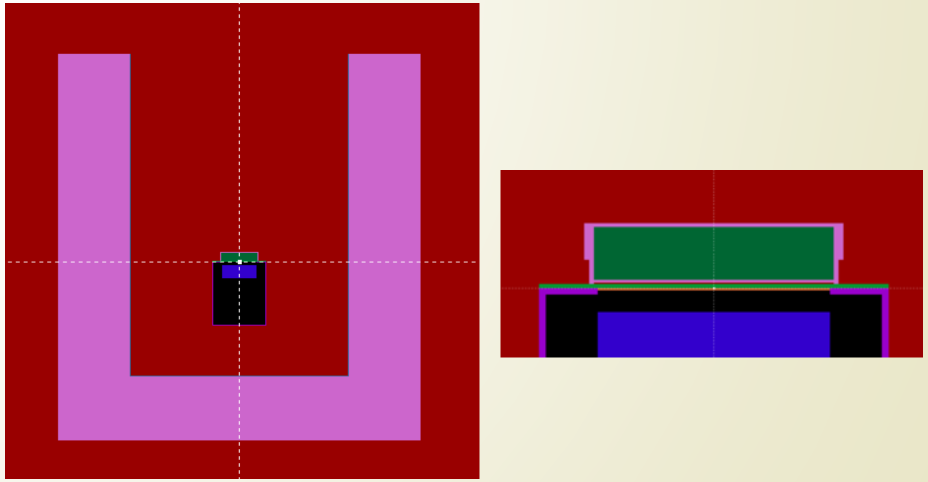
3. MONTE CARLO SIMULATION FOR THE CALIBRATION OF THE LEGe DETECTOR

EFFICIENCY CALIBRATION

- Two detectors were used to analyze the samples: LEGe and XtRa
- LEGe detector was calibrated using the Monte Carlo computer code PENELOPE
- Photons interacting with an energy deposition detector were simulated
- Photon energies from 46.00 keV to 1836.08 keV were used
- Construction of the geometry-definition file (**.geo**), the input file (**.in**) and the material file (**.mat**)



Dimensions of the Petri cup

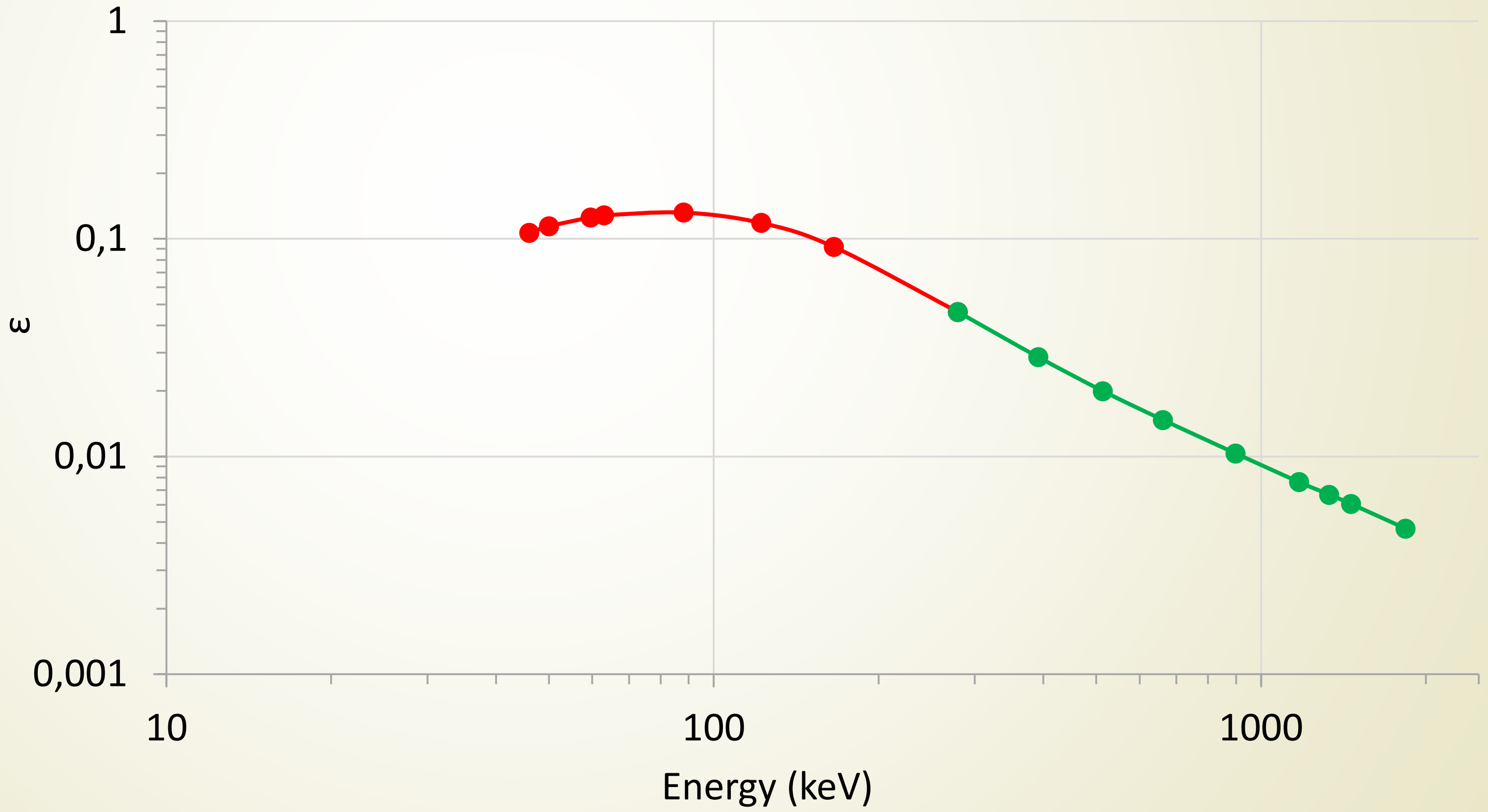


Geometry of LEGe detector and Petri sample with GVIEW2D.

EFFICIENCY CALIBRATION

Calibration curve for LEGe detector – Petri geometry

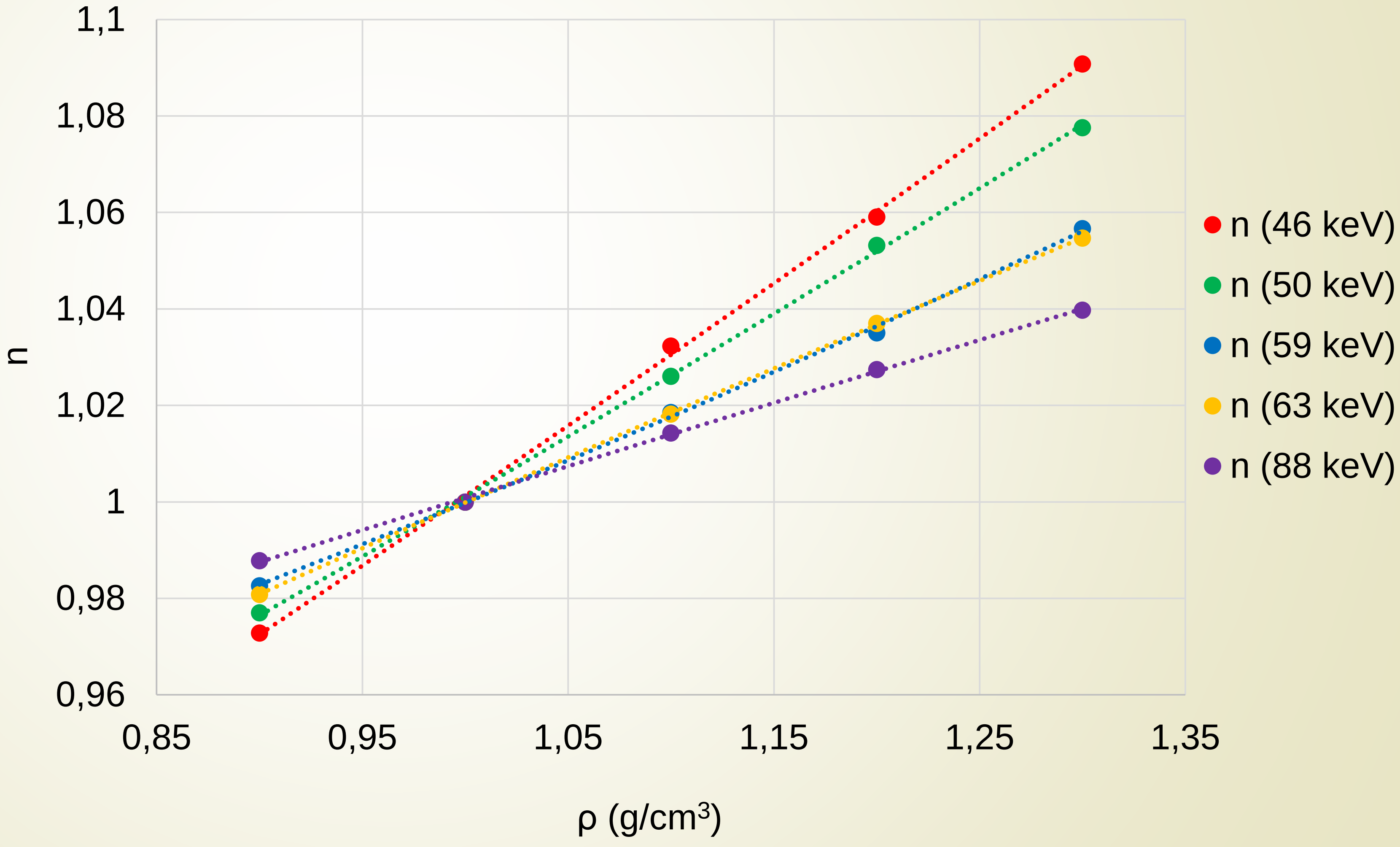
- Uncertainties are not shown in the graph because their values are very low and they cannot be seen with this scale



EFFICIENCY CALIBRATION

Density effect on self-absorption

$$n(\rho, E) = \frac{\varepsilon\left(\rho = 1\,\frac{g}{cm^3}, E\right)}{\varepsilon(\rho, E)}$$



4. ACTIVITY CALCULATIONS

DETERMINATION OF SOIL ACTIVITY

- Analysis with the code SPUNAL
- Kamena Vourla: net area of the photo-peak. Uncertainties:
  - Photo-peak area
  - Calibration of the detector
  - Detector model used in the simulations
  - Weighting procedure
- Thermopylae: activity concentration. Uncertainties:
  - Photo-peak area
  - Calibration of the detector
  - Weighting procedure
  - Self-absorption corrections

$$A = \frac{area}{time \cdot yield \cdot \varepsilon}$$

$$cps_{net} = cps_{tot} - cps_{back} \Rightarrow A = \frac{cps_{net}}{yield \cdot \varepsilon}$$

Activity concentration

$$A = \frac{cps_{net}}{yield \cdot \varepsilon} \cdot \frac{1}{net\ mass}$$

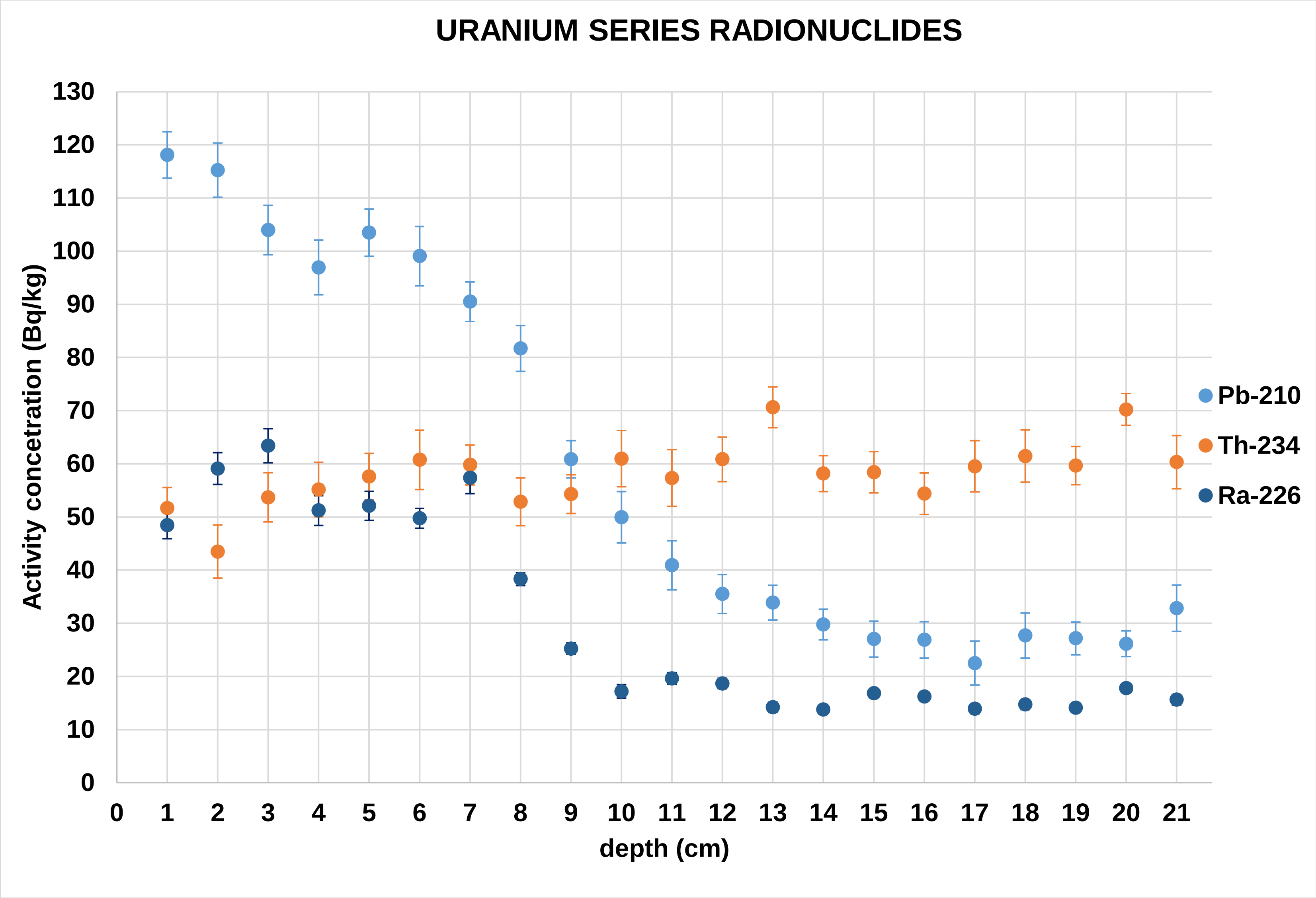
5. RESULTS AND CONCLUSIONS

RESULTS – KAMENA VOURLA

Initial assumptions

- The soil was assumed to be undisturbed
- The expected behavior of the isotopes is different depending on their origin
- Isotopes coming from the atmosphere should be present with higher activity in the first centimeters and decreasing with the depth. Radionuclides brought by the water may also have this kind of vertical distribution, or another profile, depending on the water migration in the soil.
- If the radionuclide appears having the same specific activity at all depths, it is reasonable to believe that the element is naturally contained in the soil itself.

RESULTS – KAMENA VOURLA



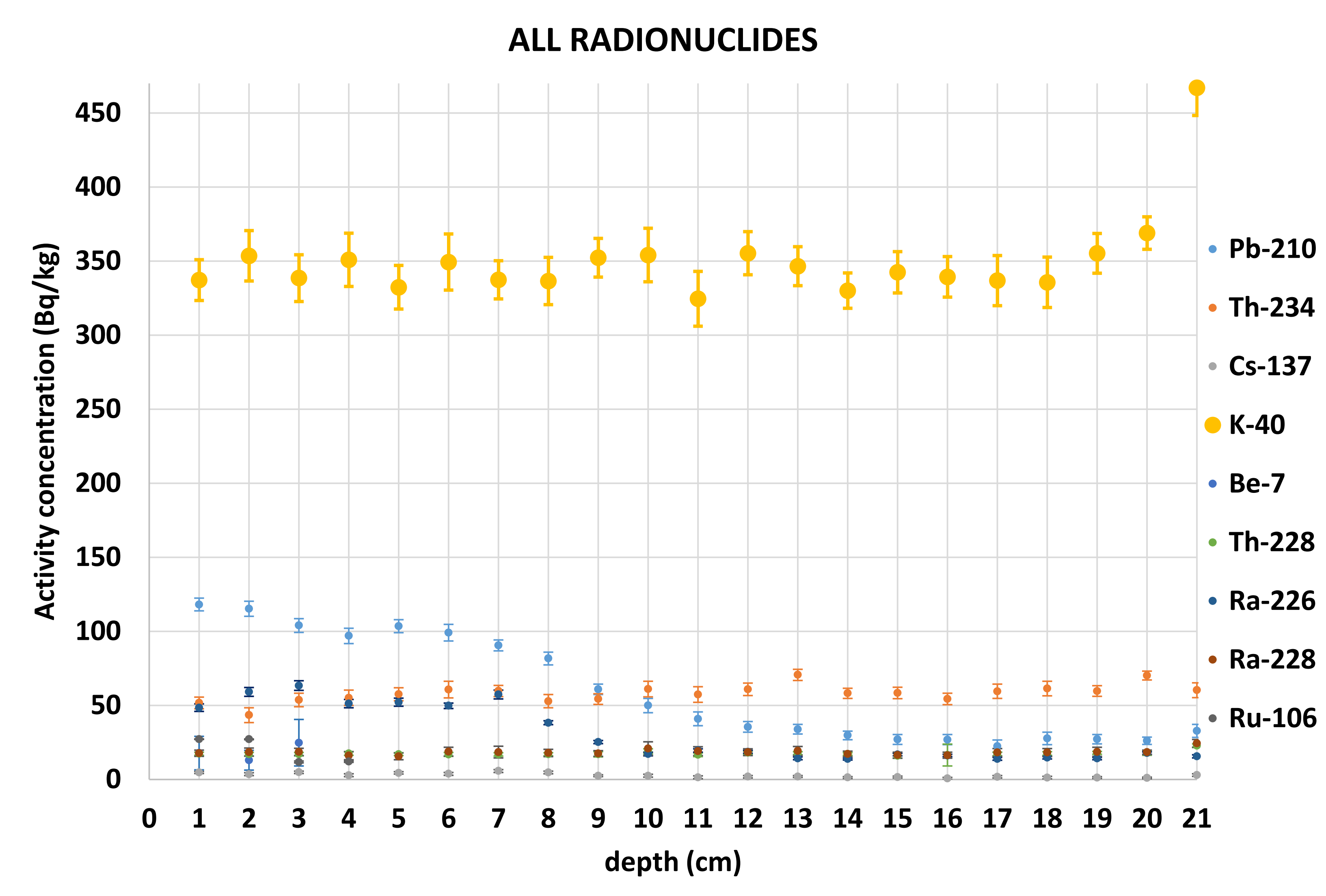
^{210}Pb

- General decreasing trend up to the depth of ~18cm, constant at larger depths.
- From around 120 Bq/kg to ~27 Bq/kg, almost four times lower.
- No significant disturbance of the soil.

^{226}Ra

- Equilibrium with ^{234}Th in the first 7 cm.
- In the deep soil there is significant disruption of equilibrium between the three radionuclides.
- Higher values of activity concentration in the ground surface might be related to the addition of water.
- A complete explanation cannot be given at this point.

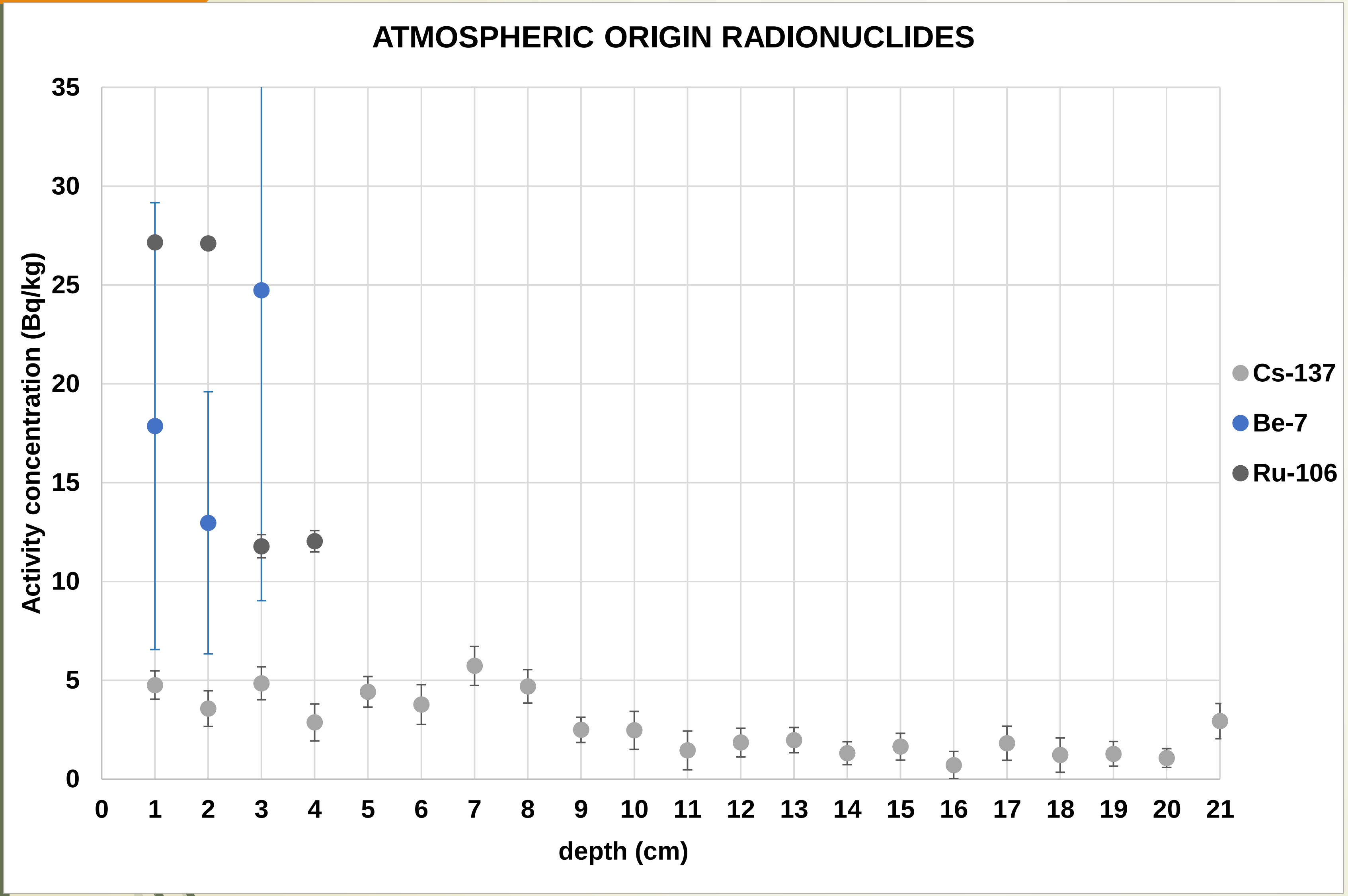
RESULTS – KAMENA VOURLA



^{40}K

- Approximately constant value.
- Under the hypothesis of undisturbed environment it was expected.

RESULTS – KAMENA VOURLA



^{137}Cs

- Non-uniform tendency may suggest a disturbance in the soil (until 7-8 cm)
- Low values: expected

^{106}Ru

- Detected in the first 4 cm
- Normally not detected in the environment

RESULTS – KAMENA VOURLA

^{106}Ru detection

Greek Atomic Energy Commission (EEAE)

- **09th Oct**: between 27th Sept and 5th Oct ^{106}Ru was detected in the atmosphere in Athens at very low concentrations (< 5 mBq/m^3).
- **12th Oct**: after 5th Nov the activity concetration was below the detection limit (< 0.2 mBq/m^3). According to EEAE these concentrations do not pose any risk to health or the environment.
- The source of the Ru-106 was not located in the Greek territory and it remained unknown at that time.

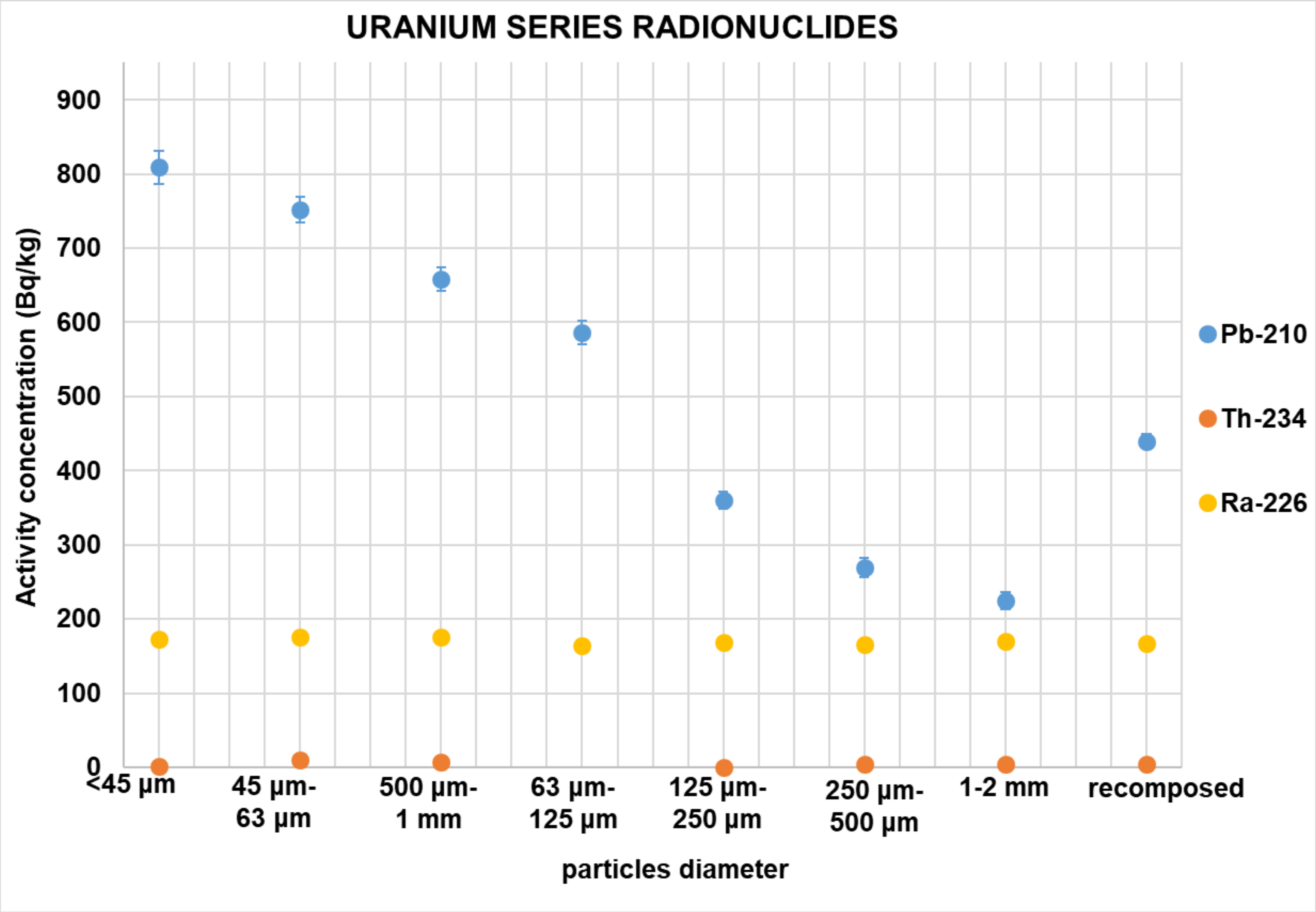
RESULTS – THERMOPYLAE

Size fractionation of radionuclides in Thermopylae surface soil

- The expected behavior of the isotopes is different depending on their origin
  - Isotopes presenting a higher activity concentration in the smaller particles: brought into the ground from an external source. The source could be water or the atmosphere.
  - Almost constant value of activity concentration: the isotope originates from the soil itself.

- Extremely complex environment: the ground could not be described as soil, but it appeared as a mixture of rocks, dust and organic matter. The unknown composition of the superficial soil may have influenced both the behavior of radionuclides and the gamma-ray analysis procedure.
- Coalescence of the finer fractions due to the action of water during the sieving procedure.
- Influence of hot water floods on the radioactivity of the superficial soil.

RESULTS – THERMOPYLAE



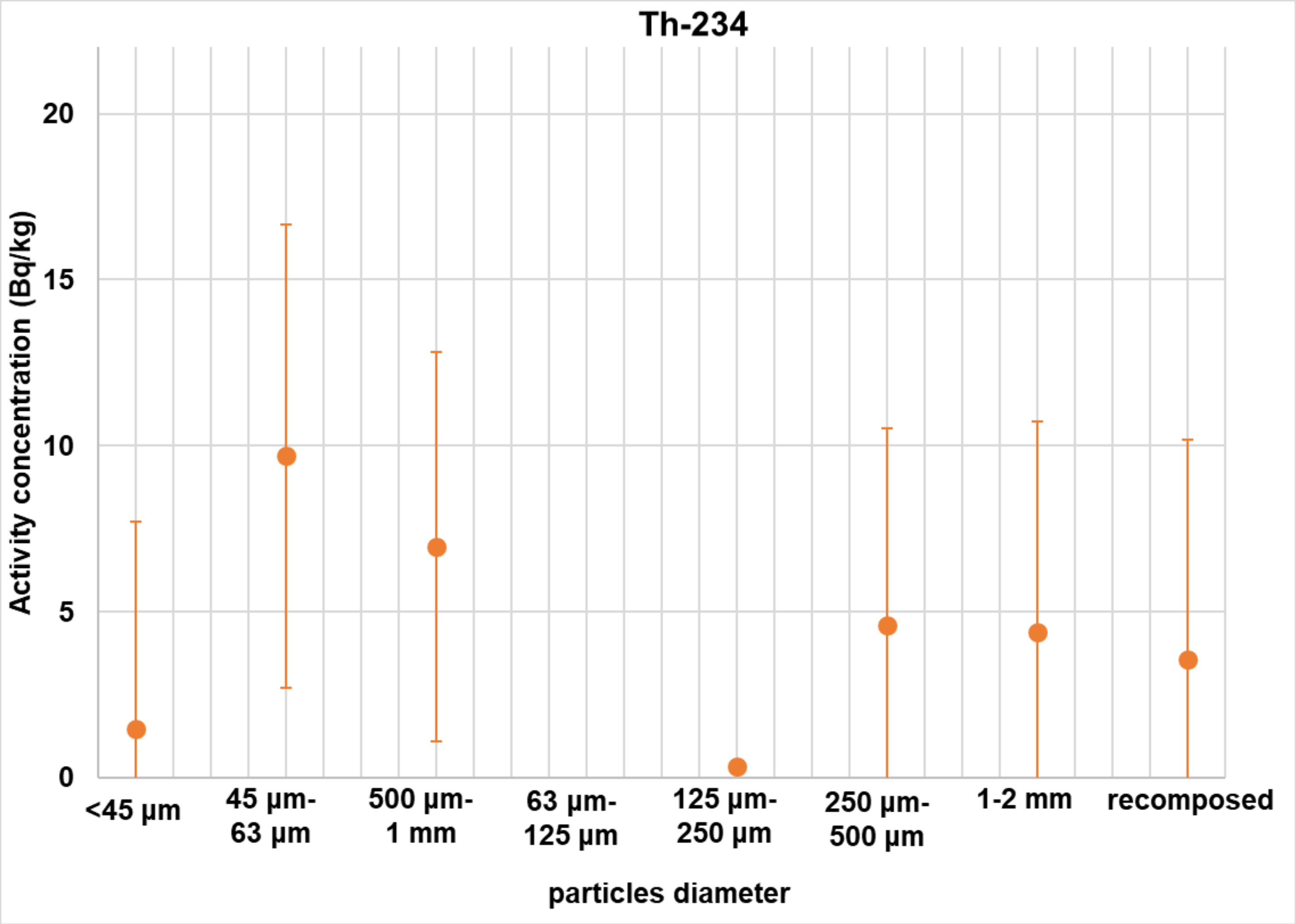
^{210}Pb

- Decreasing activity concentration in larger particles. A significant amount of the ^{210}Pb detected in the surface soil comes from the atmosphere, as a decay product of atmospheric ^{222}Rn (unsupported ^{210}Pb).

^{226}Ra

- Extremely soluble in water, expected to be higher in smaller particles
- Approximately constant around 170 Bq/kg.
- Possible interpretation: ^{226}Ra originates from the ground itself.
- Disruption of radioactive equilibrium between ^{234}Th and ^{226}Ra, indicating that ^{226}Ra is not produced locally by the decay of ^{238}U, but is brought in the area, probably with the water.

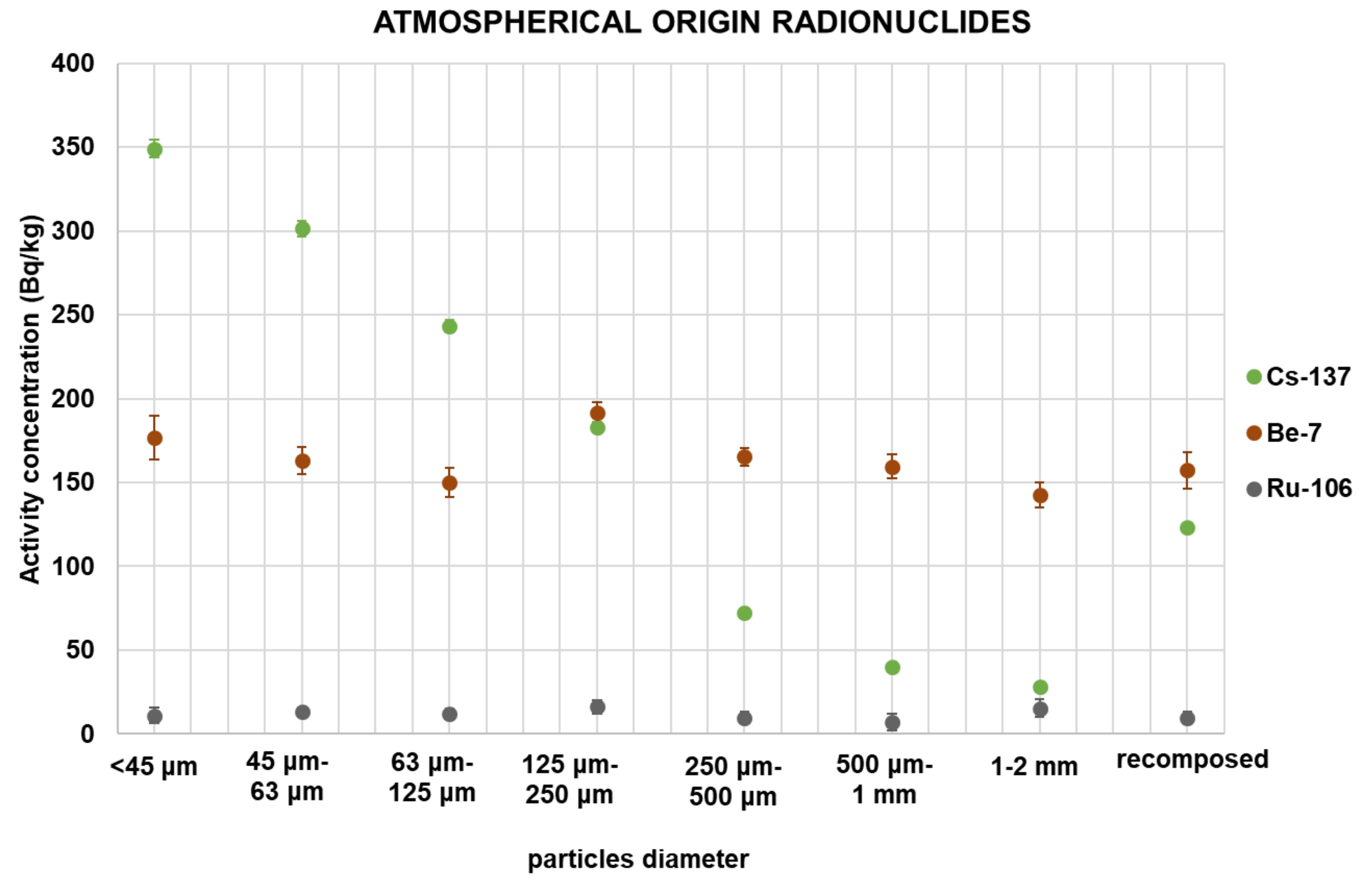
RESULTS – THERMOPYLAE



^{234}Th

- Uncertainties are extremely high and activity concentration is very low.
- MDA: 7.5 Bq/kg.

RESULTS – THERMOPYLAE



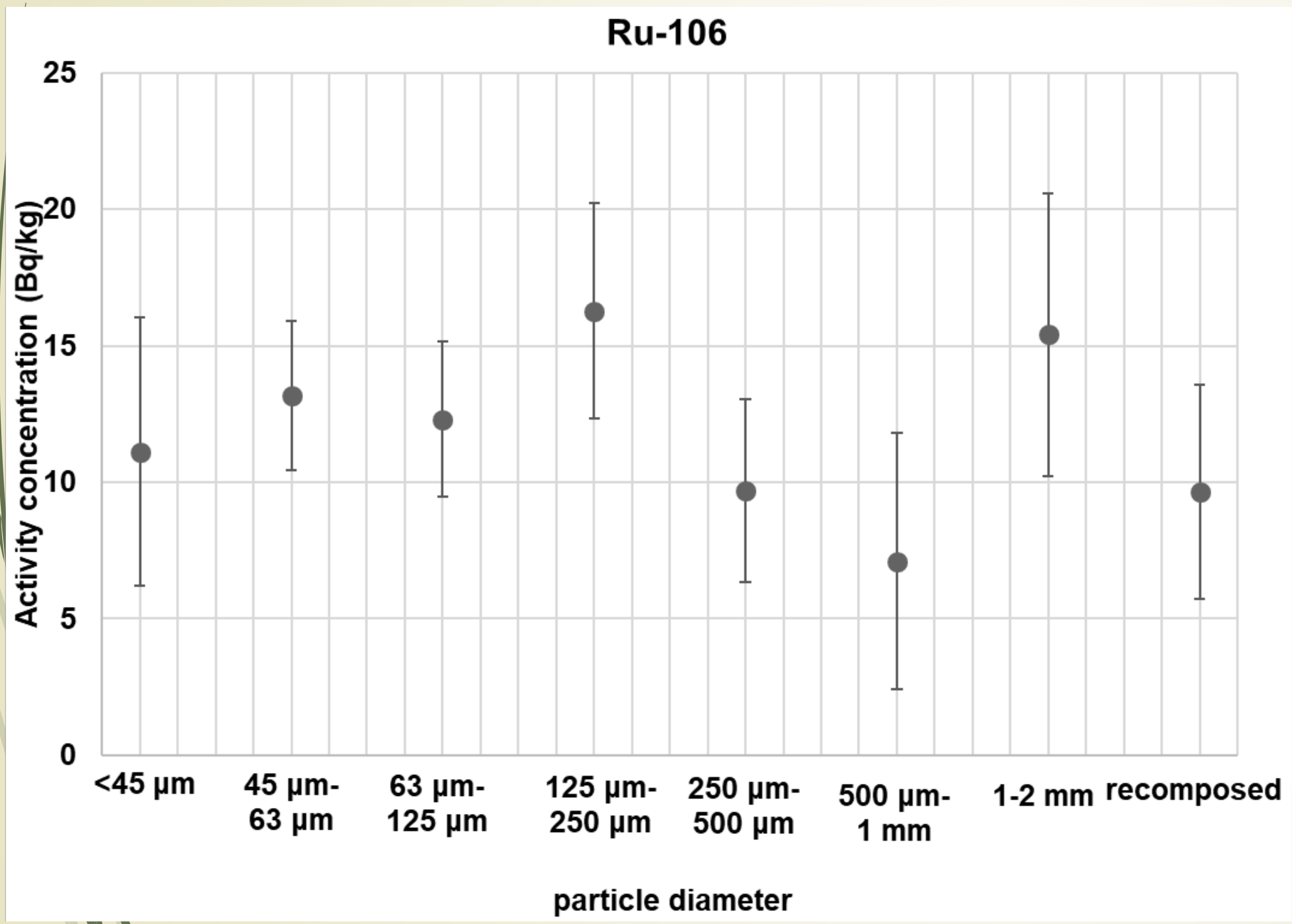
^{137}Cs

- Decreasing activity concentration in larger particles, expected

^{7}Be

- Higher activity concentration in 125-500 μm
- No easy explanation

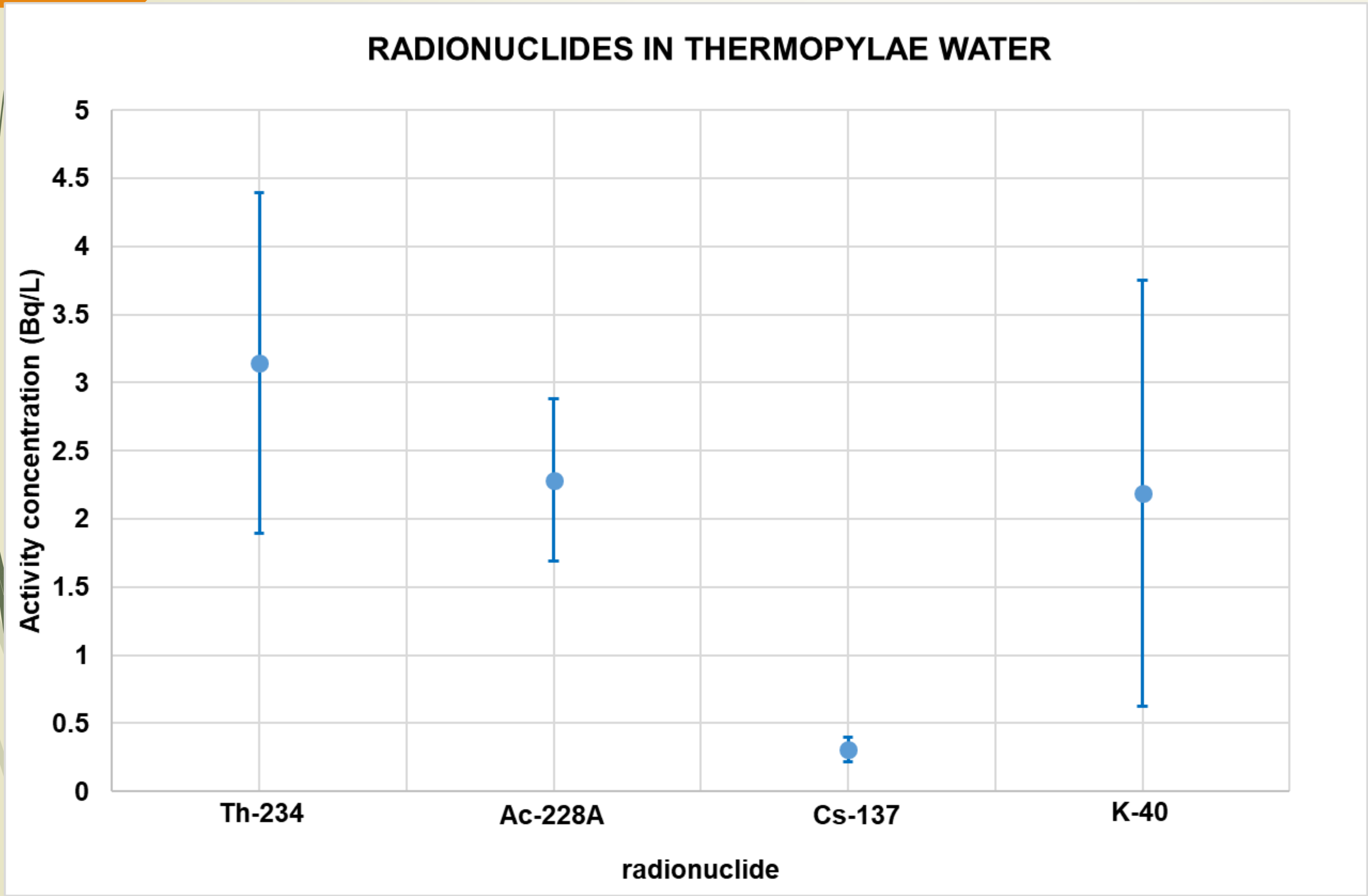
RESULTS – THERMOPYLAE



^{106}Ru

- Detected
- High uncertainties and low values

RESULTS – THERMOPYLAE



A few radionuclides were detected.

For all the other nuclides, the activity concentration was below detection limit.

The enrichment of the ground in radionuclides due to the hot water is a long-term procedure.

CONCLUSIONS

- Unexpected behaviour of some radionuclides, in particular of ^{226}Ra in both Kamena Vourla and Thermopylae.
- Other radionuclides showed an expected trend, i.e. ^{210}Pb.
- The complexity of the sampling sites environment strongly influenced the capability to predict the activity concentration profiles.
- Possible future investigations
- repeat the sampling in the same locations but in different sampling points
- study the size fractionation of the radionuclides in Kamena Vourla
- perform further investigations about the nature of the Thermopylae ground material and the influence of self-absorption on the detection of photons.

THANK YOU FOR YOUR ATTENTION

Sofia Padovani

sofia.padovani@studio.unibo.it

sofiapadovani08@gmail.com