Università degli Studi di Ferrara

In-situ gamma-ray spectrometry challenges for NORM hotspot detection

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Outline

Hotspot detection due to ^{226}**Ra**

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NORM and **TENORM**

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Challenges related to field activity

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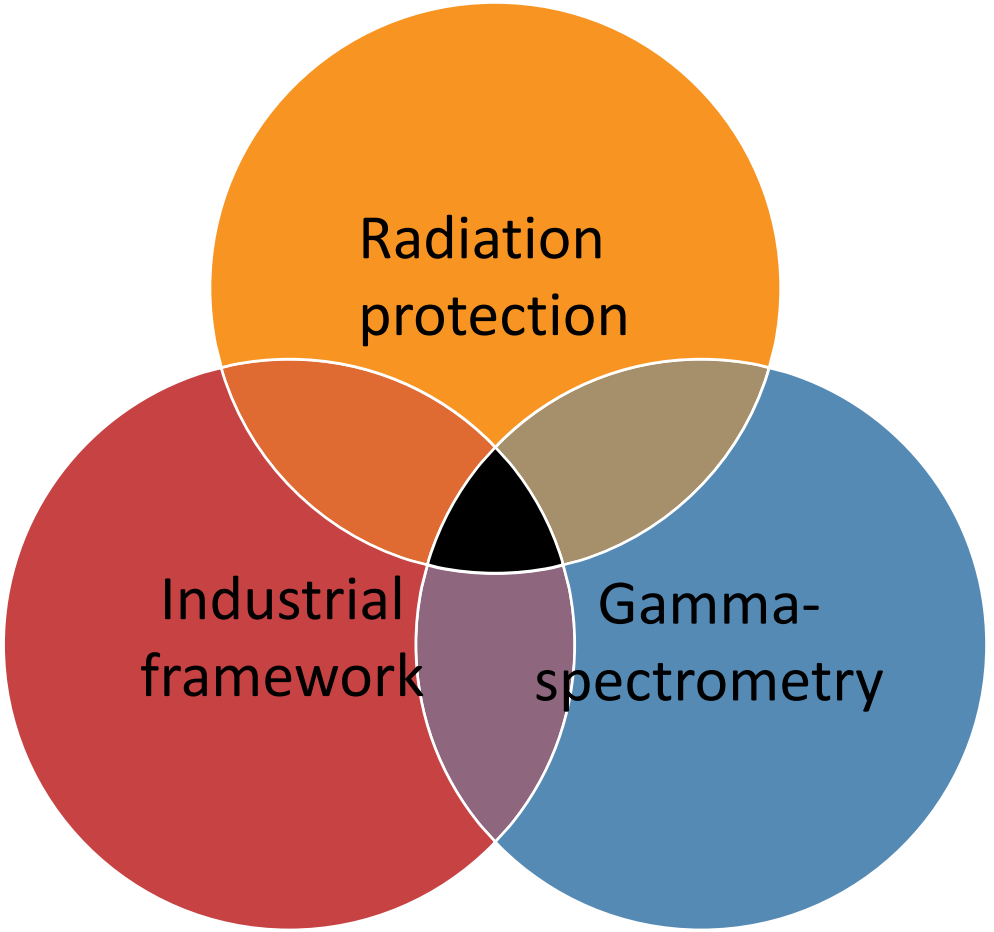
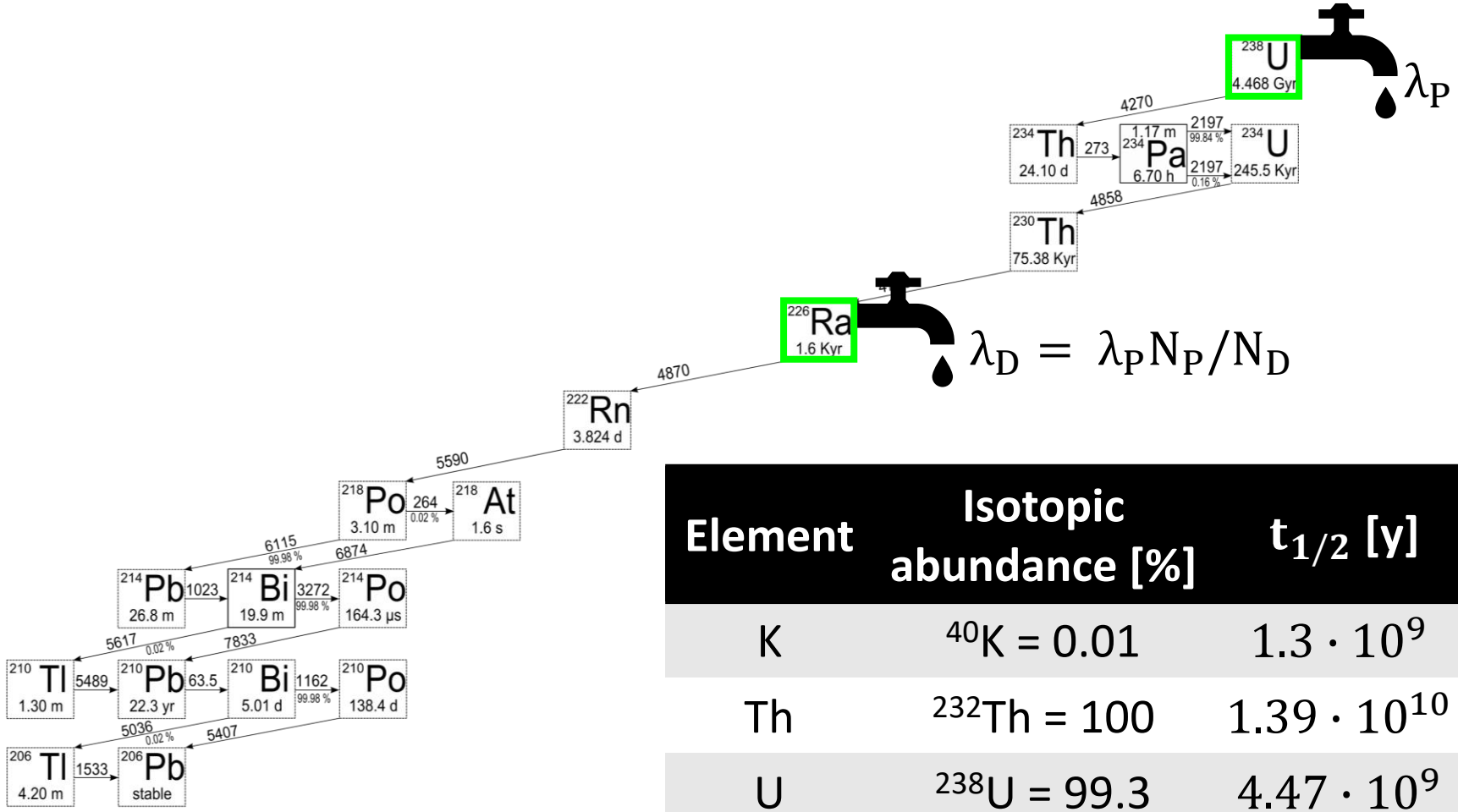
Instruments and **methodologies**



Radionuclides of terrestrial origin

Terrestrial radionuclides originate from natural radioactive decay within the **Earth's crust**, with abundance varying by location and geology. They can form complex decay chains and can be detected through **gamma spectrometry.** When unperturbed, ^{238}U and its daughters are in **secular equilibrium:**



Element	Isotopic abundance [%]	$t_{1/2}$ [y]	Daughter	Typical abundance
K	^{40}K = 0.01	$1.3 \cdot 10^9$	-	0.04 – 2.9 [%]
Th	^{232}Th = 100	$1.39 \cdot 10^{10}$	^{208}Tl	2 - 12 [ppm]
U	^{238}U = 99.3	$4.47 \cdot 10^9$	^{214}Bi, ^{214}Pb	0.7 – 10.7 [ppm]

Ropture of secular eq. in Phosphogypsum

In the **phosphate fertilizer production** industry, phosphate rocks are processed to obtain phosphate fertilizers ($\mathbf{P_2O_5}$) through a procedure known as **Wet Process for Phosphoric Acid** (H_3PO_4):

$$Ca_3(PO_4)_2(s) + 3H_2SO_4(l) \rightarrow 2H_3PO_4(l) + 3CaSO_4(s)$$

World data for phosphate rocks(2021):

- DEMAND (FERTILIZER)$\sim 47 \cdot 10^9$ kg
- DEMAND (OTHER USES)$\sim 8 \cdot 10^9$ kg
- PROD. + RES.$\sim 71 \cdot 10^9$ kg

80-85% of ^{238}U soluble in H_2SO_4 ends up in the **phosphoric acid**. Present in phosphate rocks at average content of **30 ÷ 200 ppm.**

1 ton of PA -> 5 tons of phosphogypsum

80-90% of ^{226}Ra insoluble in H_2SO_4 ends up in the solid **phosphogypsum** byproduct.

NORM and TENORM

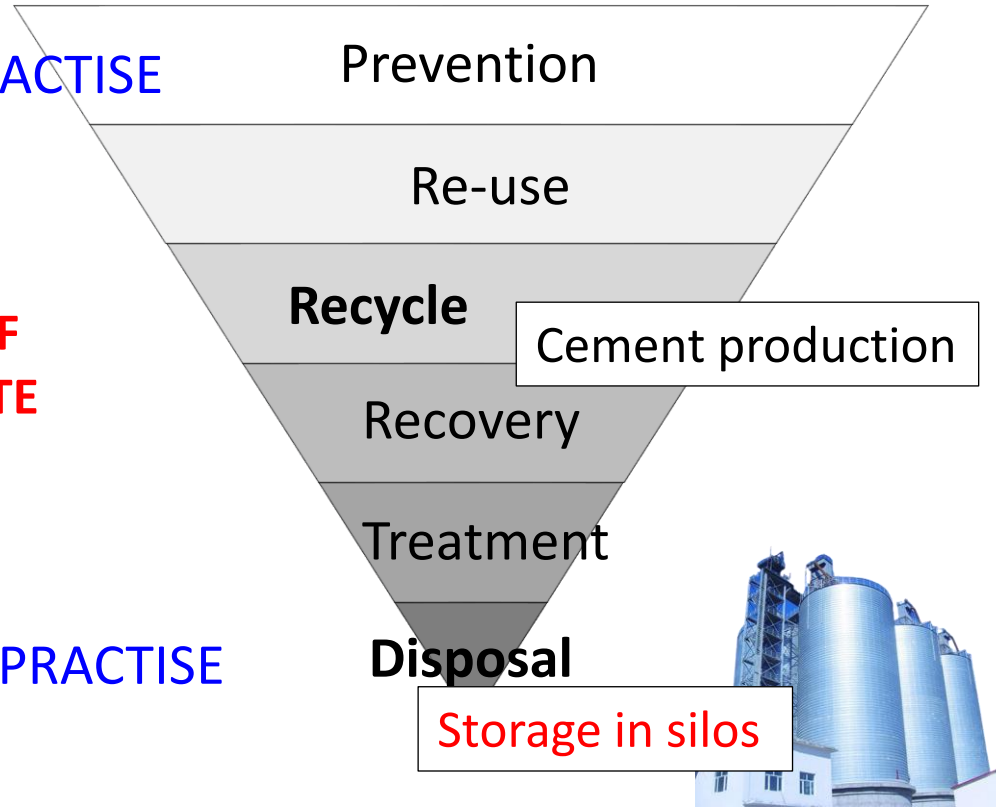
Radionuclides of natural origin contained in or released from processed materials that may pose a risk to workers, public or the environment.

- **NORM**: *Naturally Occurring Radioactive Materials* - Radioactive material containing no significant amounts of radionuclides other than naturally occurring radionuclides
- **TENORM**: *Technically Enhanced NORM* - NORM that have been concentrated or exposed to the accessible environment as a result of human activities

RADIOPROTECTION

Defined by AIEA as:
the **protection of people from** harmful effects of **exposure to ionizing radiation**, and the means for achieving this

MANAGEMENT OF HAZARDOUS WASTE

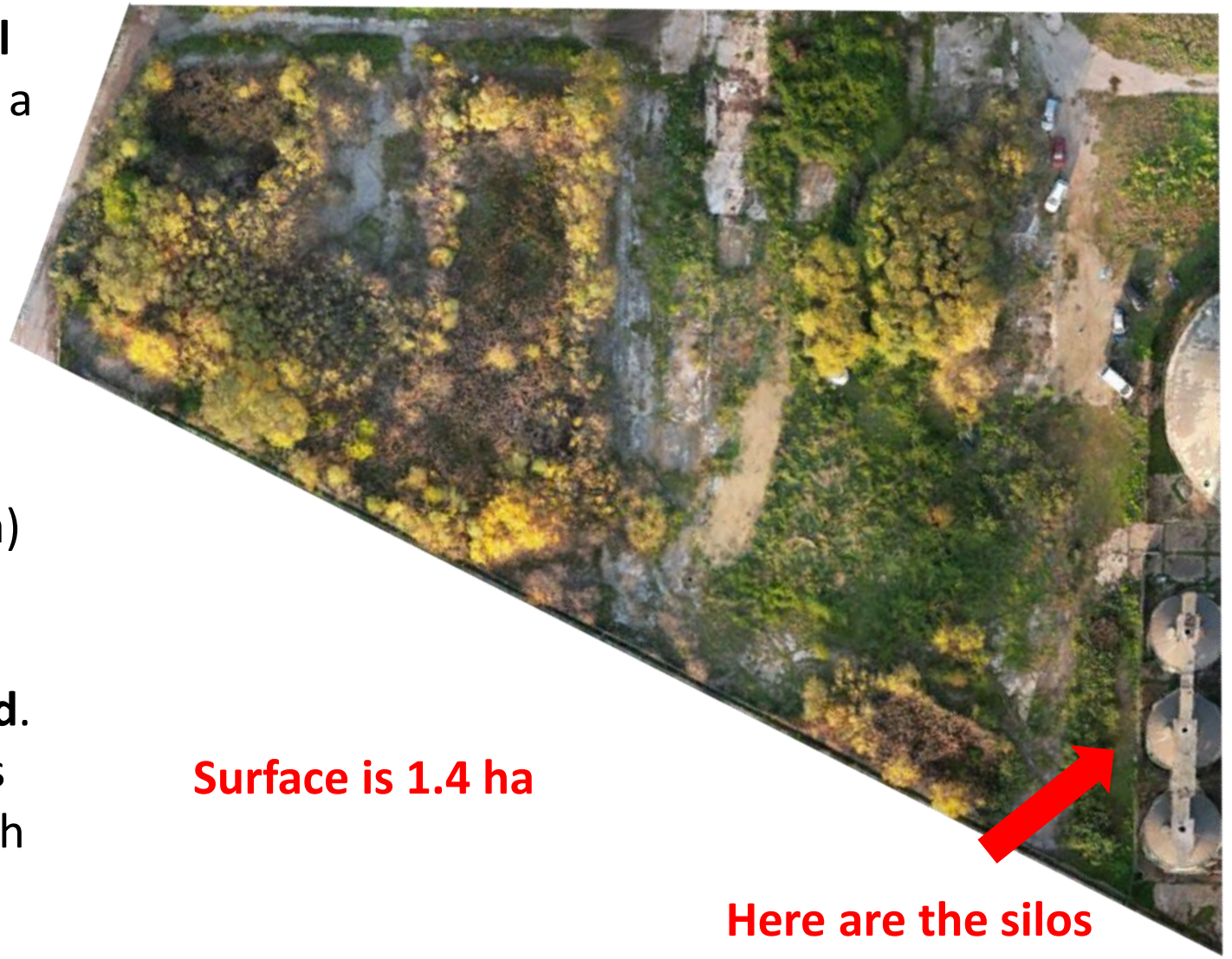


The potentially contaminated area to survey

- The survey was run on a **private industrial area**, whose **access** permit was **limited** to a few hours of a day.

- The area were suspected to be **NORM contaminated** because the terrain was once devoted to the storage of phosphogypsum. The area i'm showing today **is part of a bigger one** (surface 5 ha) characterized by **12 silos**.

- The overall area (5 ha) has been **reclaimed**. Was this done **properly or not**? How does the **radioactivity of the area** compare with the crustal median? Are **exposure values harmful** for operators?



The instruments used

ZaNaI in dynamic mode
to cover the entire accessible area on foot.

- NaI(Tl) 1L volume scintillator crystal
- PMT Hamamatsu R6231 (10 stage, 14 pin).
- MCA (0-2047 channels)

A **GNSS mobile device** for tracking the paths.

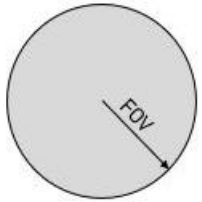
- Frequency of acquisition is 1 Hz.
- Geographic coordinates are stored in nmea format (*National Marine Electronics Association*), a standard of the satellite communications

DJI Air 2S for photogrammetric survey faithfully reporting the conditions of the day.

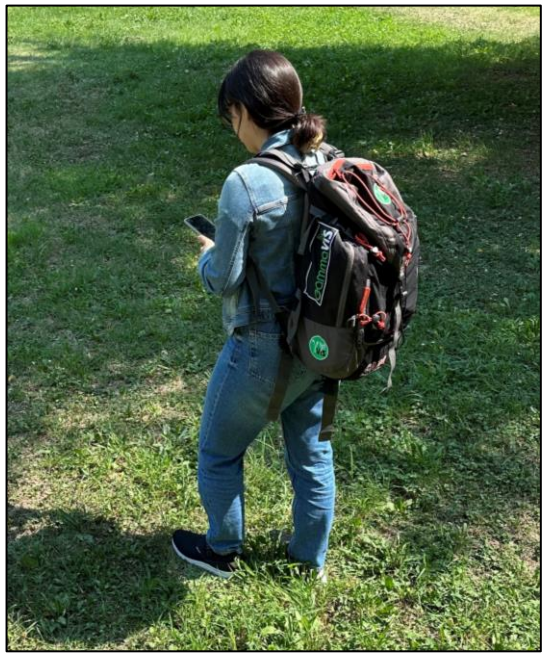
- 862 images were acquired at the rate of 2 every second, with resolution 1.5 cm/pixel

The ZaNaI detector

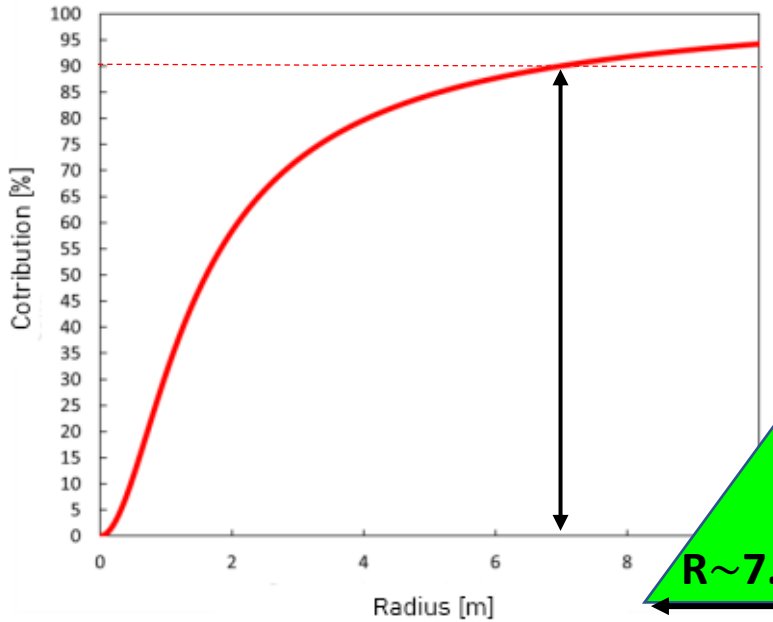
Fov in static mode

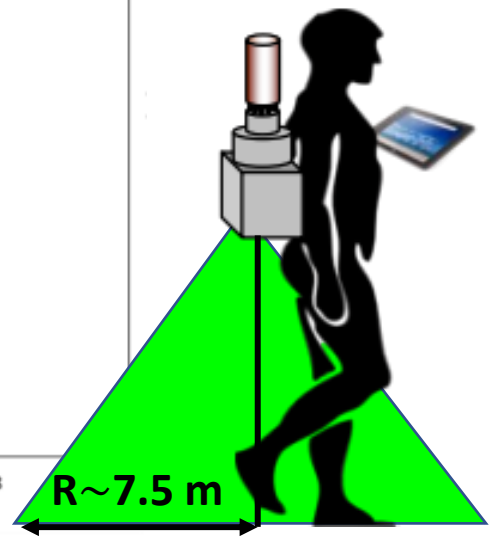


Fov in dynamic mode



Characteristics	NaI(Tl)
Volume	1 L (102x102x102 mm)
Density	3.67 g/cm^3
Energetic res.	7.3% at 662 keV (^{137}Cs) 5,2% at 1172 and 1332 keV (^{60}Co)
Absolute η	12%
Fov (1m height)	70% of the signal from a circle of 3 m radius 90% of the signal from a circle of 7.5 m radius



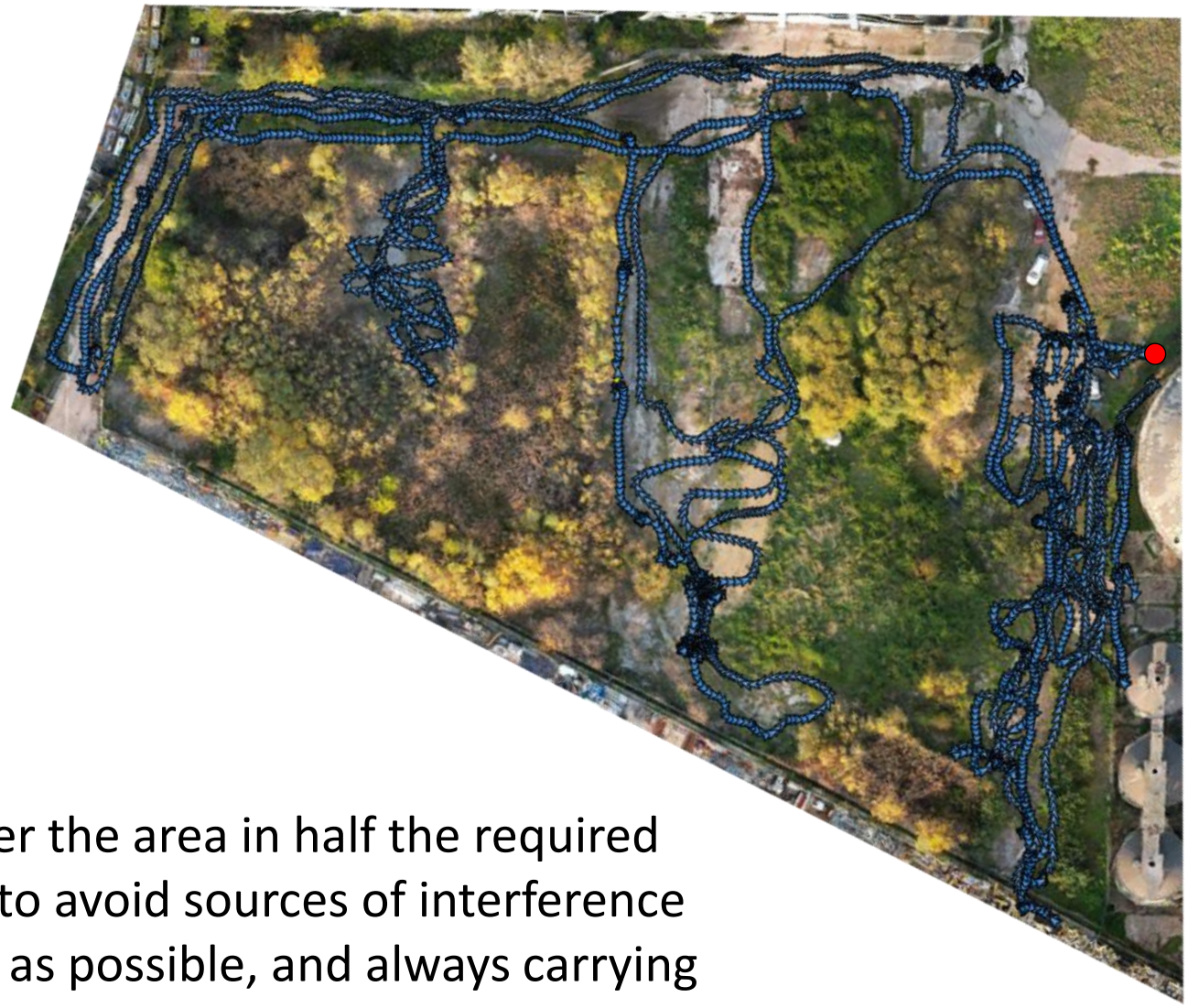


- Efficient
- Portable on the shoulders
- Doesn't need to be cooled

Survey planning and execution

Need to cover large area in small amount of time

18 November 2022	
Start of the survey	12:28:05
End of the survey	14:54:44
Total acquisition time	00:59:25
Total GNSS points	3570
Average speed [m/s]	0.78 ± 0.23



- The **employment of 2 operators** allow to cover the area in half the required time. We adopted a **standardized procedure** to avoid sources of interference for the GNSS, maintaining as constant speeds as possible, and always carrying the detectors at same heights to ensure measurement uniformity.

In red the base camp

Measures adopted to facing few challenges

Path	Start time	End time	Time range [m]	Spectra LT=10s
A	13:12:34	13:19:38	07:04	42
B	12:28:05	12:46:51	18:46	112
C	13:12:29	13:20:11	07:32	45
D	14:07:22	14:14:57	07:35	45
E	14:36:19	14:54:47	18:28	110
Total			59:25	354

- **Optimized spatial resolution** – Gamma spectra were populated with **live time of 10 s**.
- **Paths fractionalizations** – Used both during the acquisition for **better GNSS management** and during the analysis stage for regular calibration, **offsetting the instrument gain** changes due to T variations.
- **Human shielding correction** – The presence of the human body keeping the detector 1 m height was corrected by making use of **correlation parameters** calibrated for the ZaNaI on shoulder and the ZaNaI on the ground.

Procedures for data elaboration

INPUT

Gamma events + **GNSS data**

list-mode

#GammaStream 1.5 LIST	
#StartTime: 2022-11-18T12:10:31	
#Fields: Time	Channel
2259088	418
4342976	398
14443184	123
18184064	302
22971408	149

1 2 3

Post-set

Choice of live time

Energy calibration

Full spectrum analysis

Radiometric data

KMZ

GIS file

OUTPUT

1. SPECTRA CONSTRUCTION AND GEOLOCALIZATION

2. SPECTRAL ANALYSIS

3. RETURN OF GEOREFERENCED ACTIVITY CONCENTRATIONS AND DOSE

Spectra construction and geolocalization

1 Reading of ***list-mode*** and GNSS data

2 **Time alignement** of gamma events and corresponding spatial coordinates

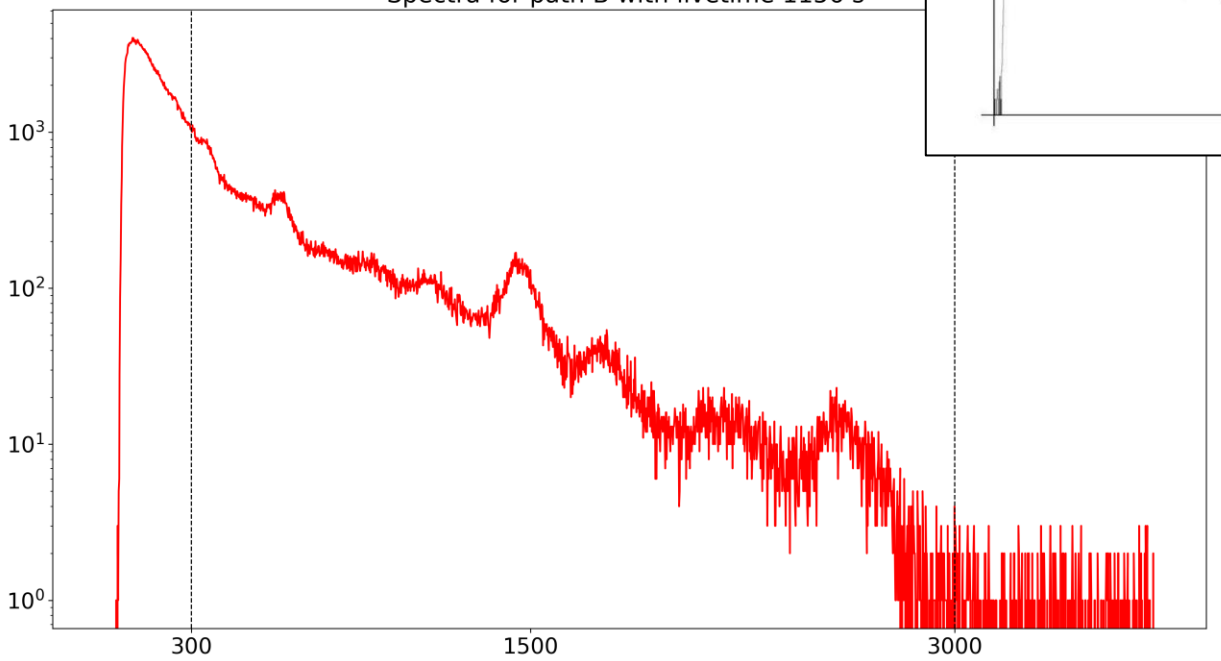
3 **Creation of 354 geospectra.** Coordinates correspond to the **temporal barycenter** of the path followed in 10s.

4 Energy calibration

3

Live time 10s

Spectra for path B with livetime 1156 s



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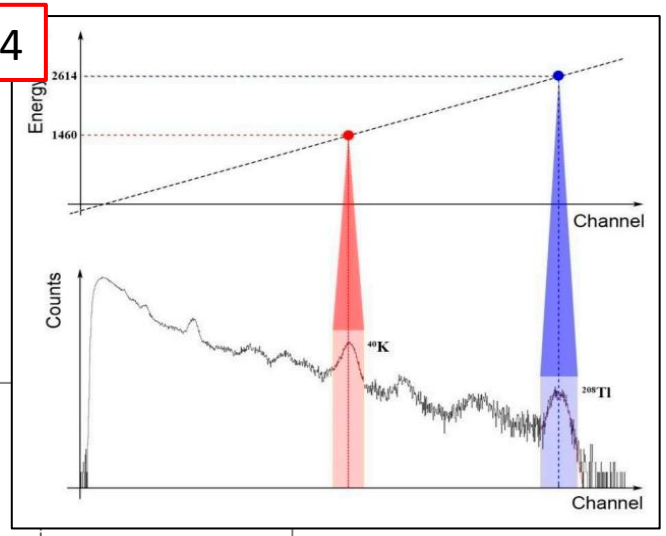
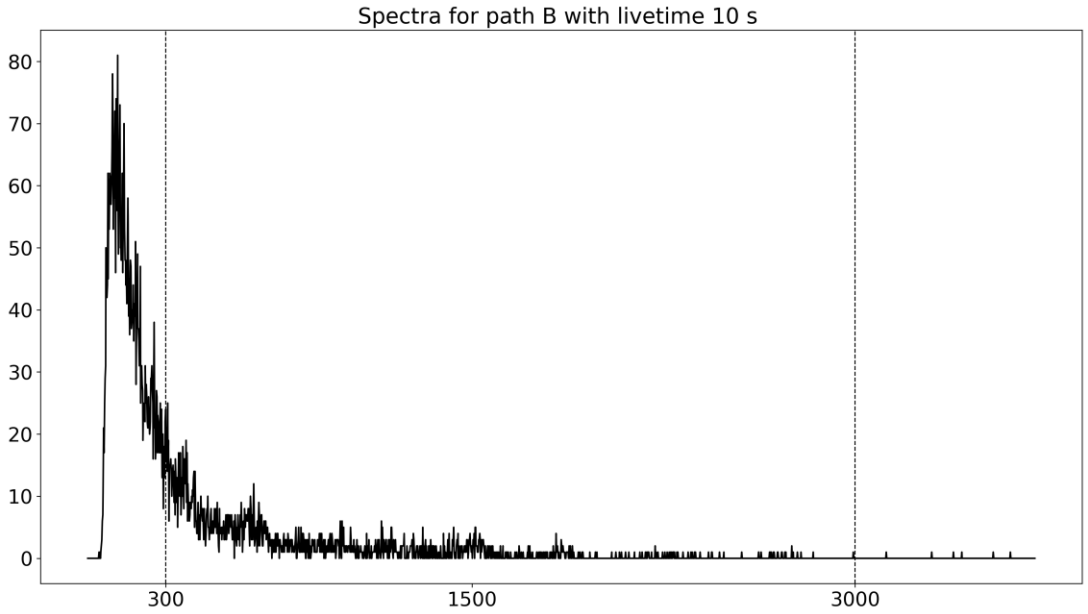
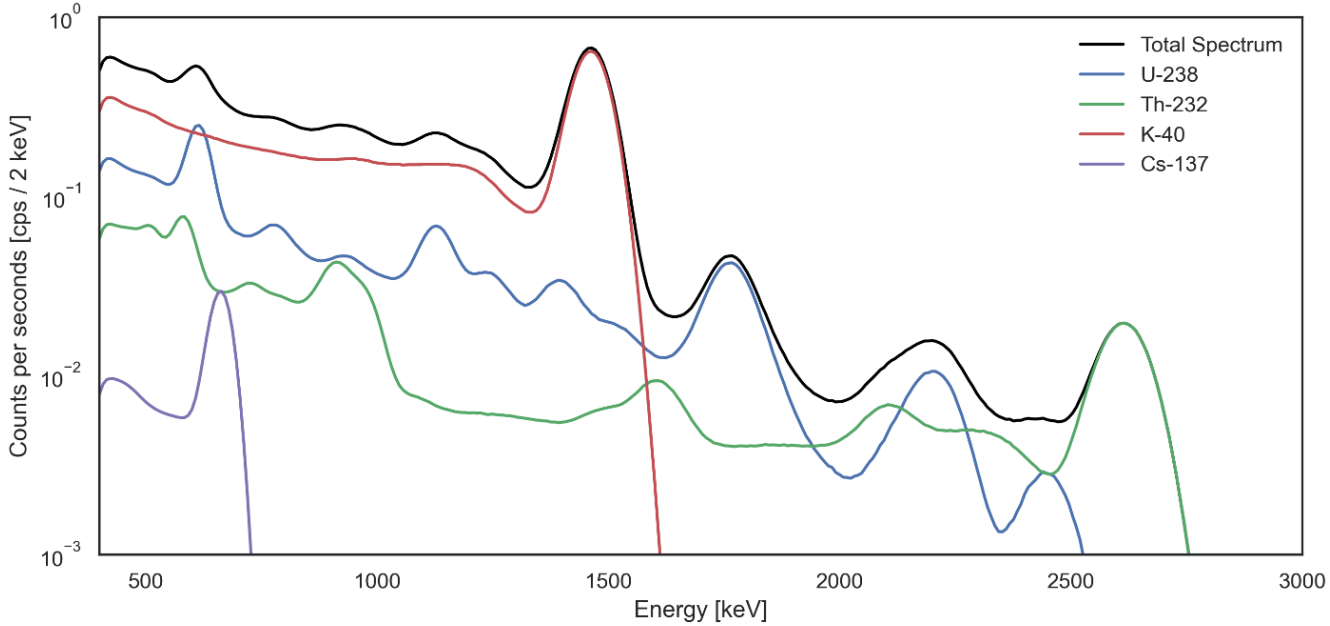


Spectral analysis

In the energy range 0.3 – 3.0 [MeV] the **Full Spectrum Analysis** method algorithm is applied so that the detected experimental gamma spectra can be seen as a combination of the fundamental spectra from ^{40}K, ^{238}U, ^{232}Th, and ^{137}Cs.





$$N(i) = \sum_{k=1}^{4} C_k \cdot S_k(i)$$

ELEMENTS CONCENTRATION

COUNT RATE for CHANNEL i

COUNTS of the FUNDAMENTAL SPECTRUM of the ELEMENT k

Overall area results 1/2

Overall radiometric data of the area (path A+B+C+D+E):

- Mean and median very close
- Highest σ/μ value is for ^{226}Ra~25%
- Max value in ^{226}Ra concentration is > mean+3σ
- Only ^{226}Ra values are above the references

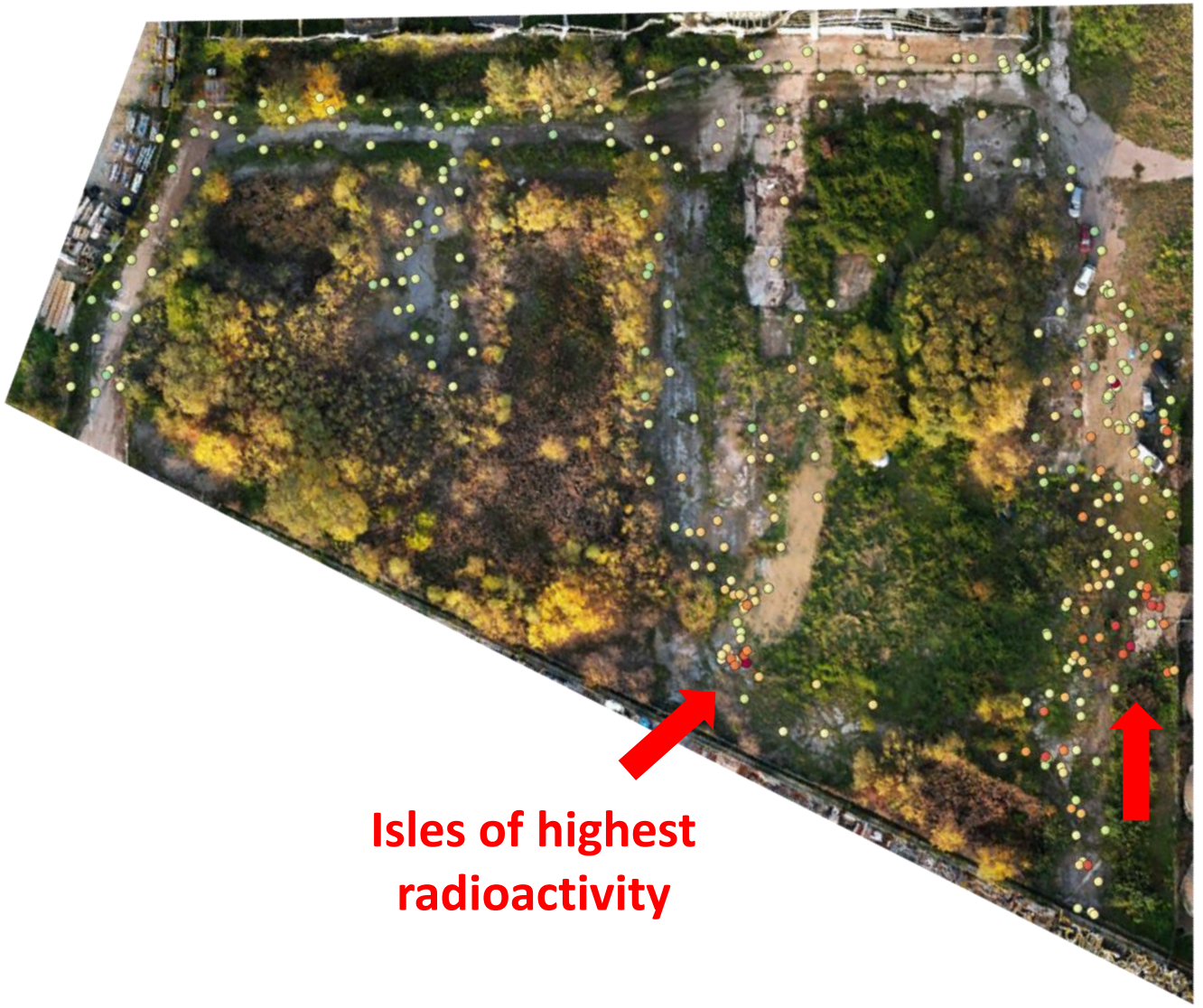
	Median	Mean ± 1σ	Min	Max	Median earth's crust	Building constr. materials
^{40}K [Bq/kg]	276	277 ± 60	128	514	410	400
^{226}Ra [Bq/kg]	50	53 ± 13	23	106	33	40
^{232}Th [Bq/kg]	22	22 ± 4	12	40	45	30
A_{tot} [Bq/kg]	348	352 ± 66	191	604	488	470

Overall area results 2/2

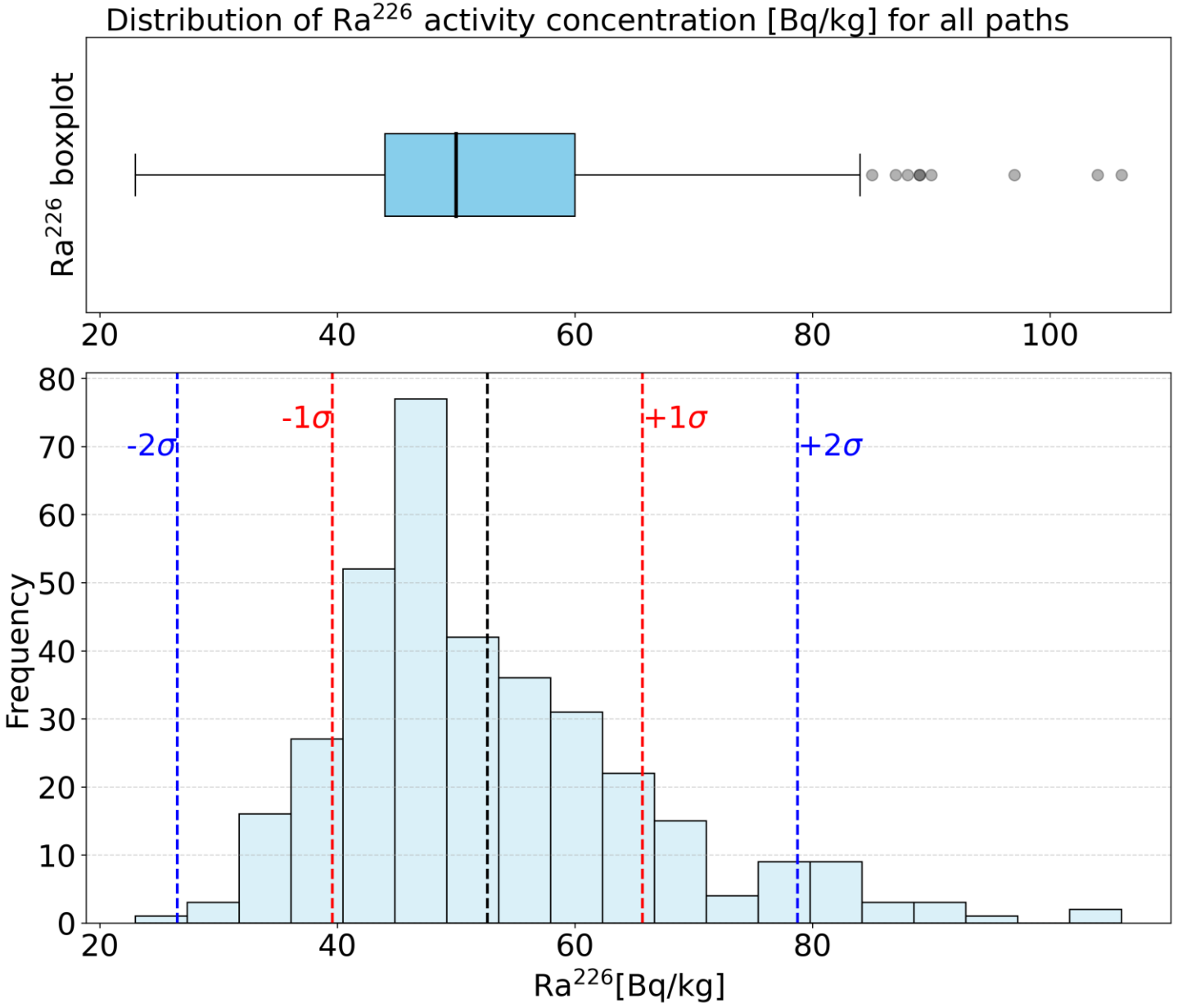
- In this map, each **radiometric measurement** is **georeferenced** and represented graphically through a **colored dot** according to a graduated scale that tends toward the red at higher activity concentration.

- Most of the dots have concentration values that are **mid-scale**

- The **hottest** extremes are **well located in specific areas**.



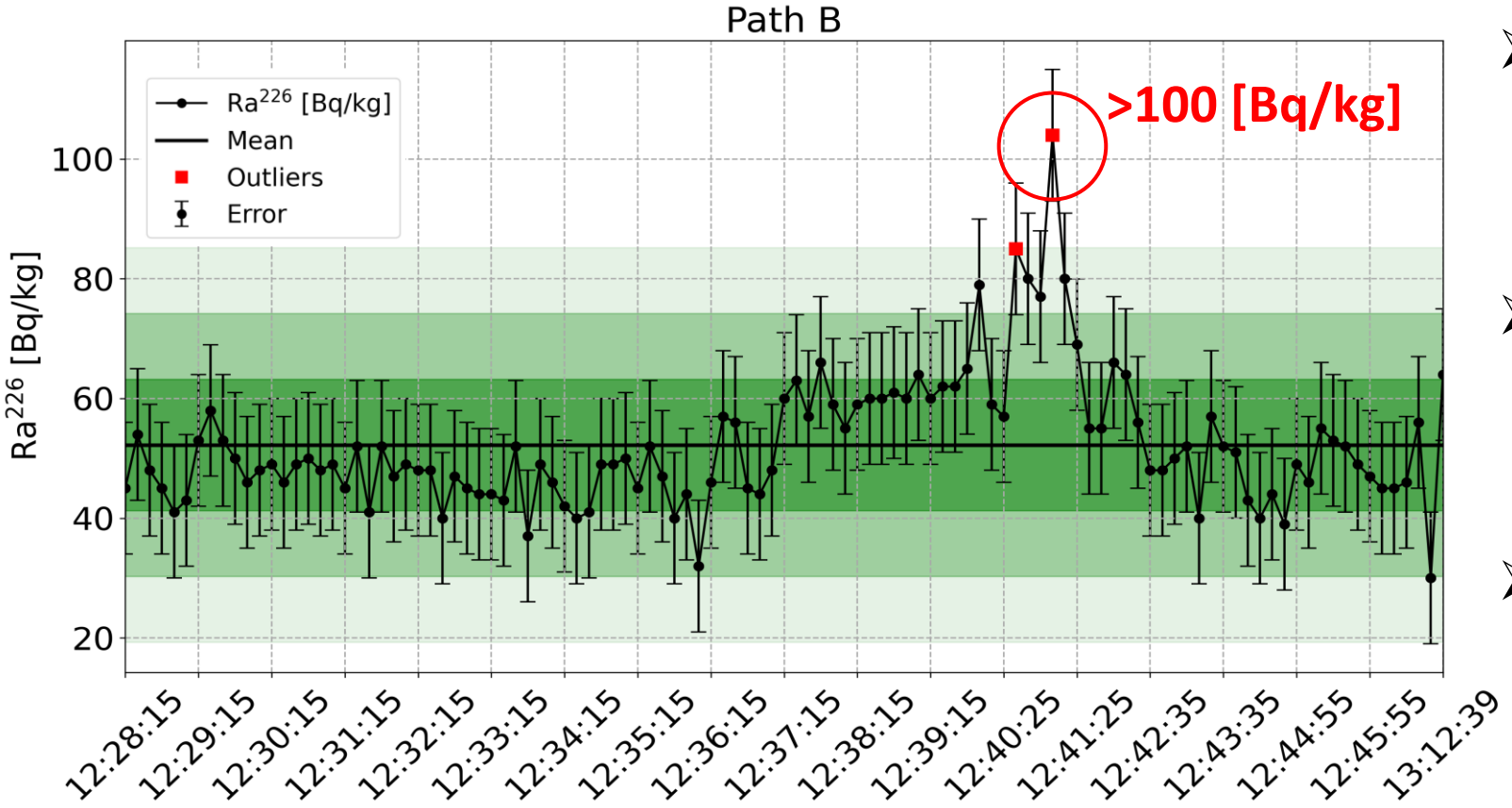
Outliers identification



The boxplot is the instrument used to identify outliers in ^{226}Ra [Bq/kg] overall distribution.

^{226}Ra [Bq/kg]	Path	Time
88 ± 14	A	13:18:04
85 ± 14	B	12:40:35
104 ± 16	B	12:41:05
97 ± 15	C	13:14:49
89 ± 14	C	13:14:59
89 ± 14	D	14:10:42
90 ± 14	D	14:11:32
106 ± 16	D	14:13:42
87 ± 14	D	14:14:22

Radium-226 outliers 2/2

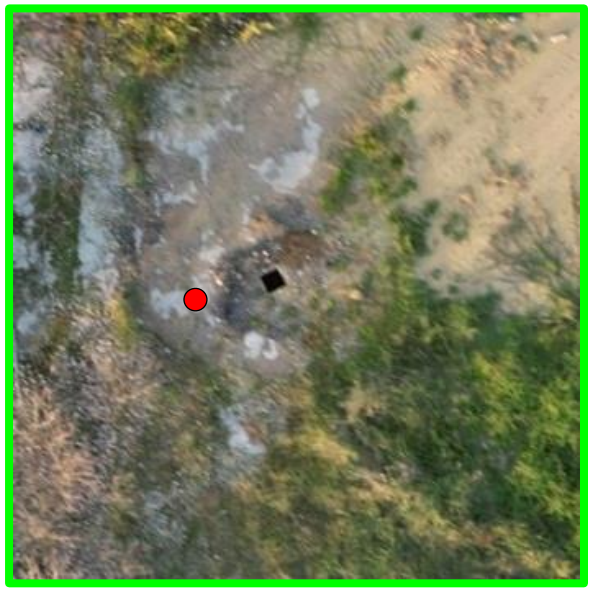


- Increasing and decreasing trend toward the two outliers
- Green areas represent distances at 1σ, 2σ and 3σ from the mean
- The same has been done for all the paths

Area of suspected NORM contamination

Path A

- No visual element could suggest NORM contamination, however, the area shown is cemented, this indicates previous human activity



Path B

Path C

Path D

Is the area harmful for operators?

Absorbed dose [Gy]

Energy deposited per unit of mass

Equivalent dose [Sv]

Absorbed dose weighted for different radiation (W_R)

Effective dose [Sv]

Equivalent dose weighted for different tissue of human body (W_T)

$$D_{abs}\left[\frac{nGy}{h}\right] = {}^{*}0.0417 \cdot A_{K-40} + {}^{*}0.462 \cdot A_{Th-232} + {}^{*}0.604 \cdot A_{Ra-226}$$

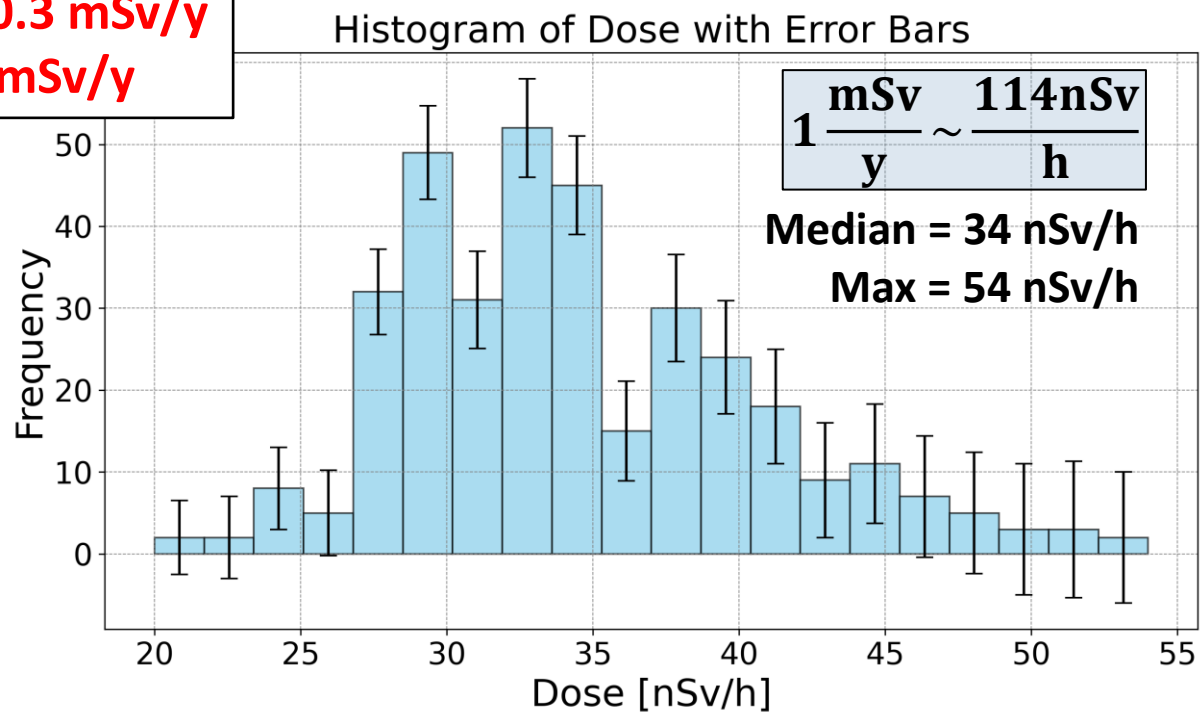
$$D_{eff}\left[\frac{nSv}{h}\right] = {}^{*}0.7\left[\frac{nSv}{nGy}\right] \cdot D_{abs}\left[\frac{nGy}{h}\right]$$

Non exposed workers
D_{eff} < 1 mSv/y = 114nSv/h

VS

Exposed workers	
D_{eff} > 1 mSv/y	
Category A	**Category B**
D_{eff} > 6 mSv/y	D_{eff} < 6 mSv/y
Special	
Exceptionally: D_{eff} >> 6 mSv/y	

Median = 0.3 mSv/y
Max = 0.5 mSv/y



*AIEA

Conclusions

Hotspot detection due to ^{226}**Ra**

- Overall, the method adopted successfully investigated an **area of 1.4 ha with an effective acquisition time of 1 hour.**
- 9 outliers for ^{226}Ra activity concentrations distribution were found, with maximum values touching the 100 [Bq/kg].
- They correspond to **5 areas of interest of possible NORM contamination**.
- These areas were located at mainly manholes, in agreement with a contamination caused by the leaking of PP byproduct into the area with rain and surface water having caused them to run into manholes.
- From a dosimetric point of view the maximum value found for the dose is **54 nSv/h** in line with the effective dose found on Italian territory of **52 nSv/h**. For the area investigated the personnel falls within the category of **unexposed workers**.

And that’s all folks!

Thank You for your kindly attention.