University of Ferrara
PhD in Physics – XXV cycle

Calibration and performances of in-situ gamma ray spectrometer

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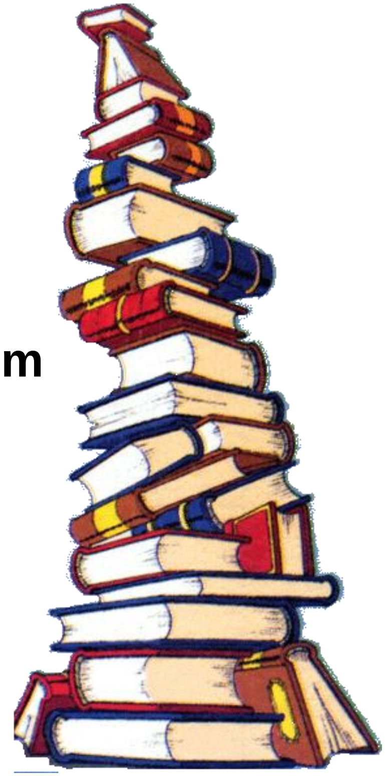
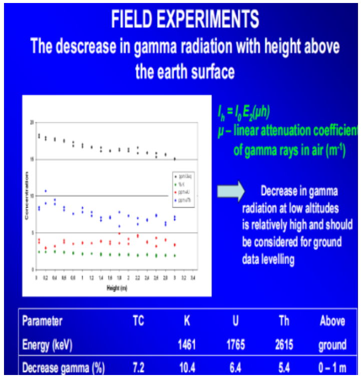
Summary

- Scientific motivation of this study
- Gamma – ray spectroscopy in situ. The ZaNaI_1.0 instrument
- Gamma – ray spectroscopy in laboratory. The MCA_Rad system
- Investigation of different parameters in-situ measurements
- Analysis of data
- Conclusions
- Publications



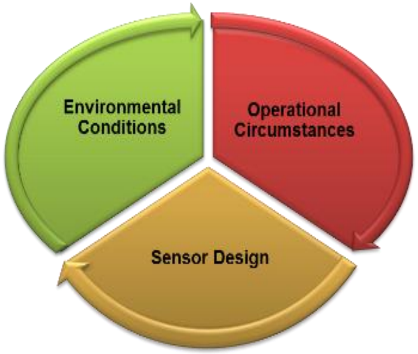
- The use of portable spectrometer for gamma-ray spectroscopy in situ is a task required for: geological, environmental and mining explorations.

- Not only ^{40}K, ^{238}U, and ^{232}Th have to be measured, but also an important key is determination of anthropic elements abundances like ^{137}Cs and ^{131}I, which are used to monitor the effect of nuclear accidents or other human activities.



- In γ-ray spectrometry surveys, it is essential to apply accurate altitude correction. The height of the detector above the ground is an important parameter for an appropriate characterization of the site[1234].

- Several factors can effect the measured concentrations of radioactive nuclides in situ gamma ray spectroscopy[5].



[1] R. L. Grasty., 1975 Atmospheric Absorption of 2.62 MeV Gamma ray Photons emitted from the ground. Geophysics vol 40, 1058-1065.

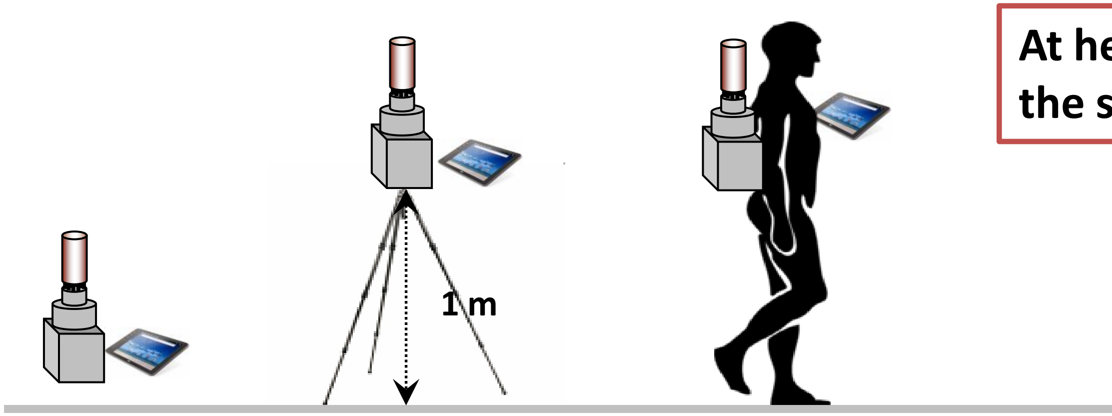
[2] Grasty et al., 1979 Fields of view of airborne gamma ray detectors. Geophysics, vol 44, 1447 – 1157.

[3] Guidelines for radioelement mapping using gamma ray spectrometry data, IAEA-TECDC-1363, July 2003

[4] Mantolin and Minty., 2009 Levelling Airborne and ground gamma ray spectrometric data to assist uranium exploration. International Symposium on Uranium Raw Material for Nuclear Fuel Cycle. IAEA 2009.

[5] E.H. Loonstra1 and F.M. van Egmond Factors influencing in situ gamma-ray measurements, EGU 2009-9247

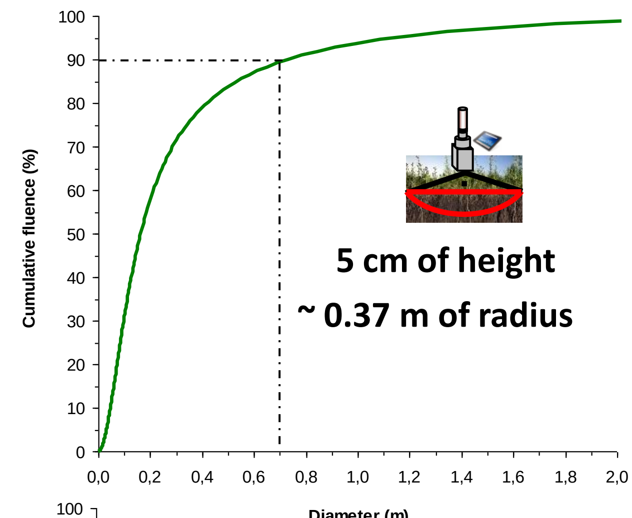
The ZaNaI_1.0L instrument: design and features

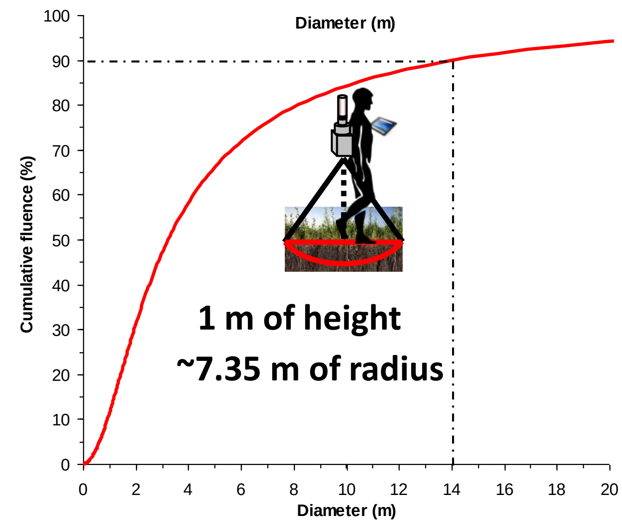


At height h, the detector receive 90% of the signal from a circle of radius R

h (m)	0.05	0.5	1.0
R (m)	0.37	3.7	7.35

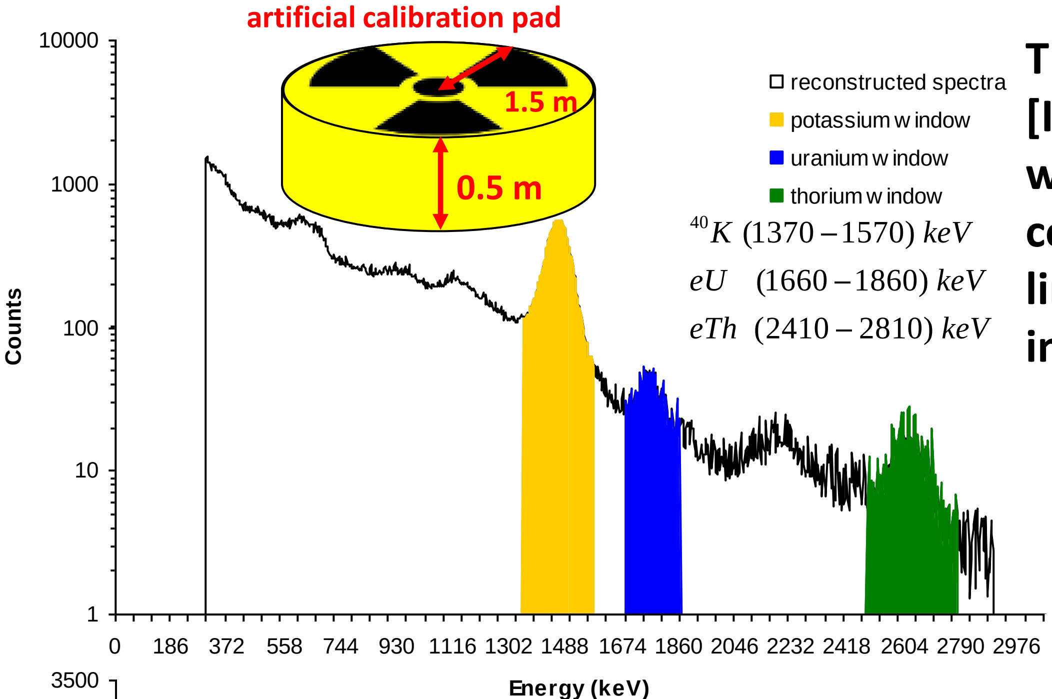
NaI(Tl) detector	**1 Liter (102 x 102 x 102 mm)**
Energetic resolution	**7.3% at 662 keV (^{137}Cs)**
Real-time feedback	**notebook (smartphone & tablet)**
Power autonomy	**6 hours**
Weight (total)	**~ 4.5 kg**
Acquisition time	**5 -10 min (static mode)** **10 – 30 sec (dynamic mode)**
Auxiliary sensors	**Pressure & Temperature**





R. L. Grasty., 1975 Atmospheric Absorption of 2.62 MeV Gamma ray Photons emitted from the ground. Geophysics vol 40, 1058-1065.

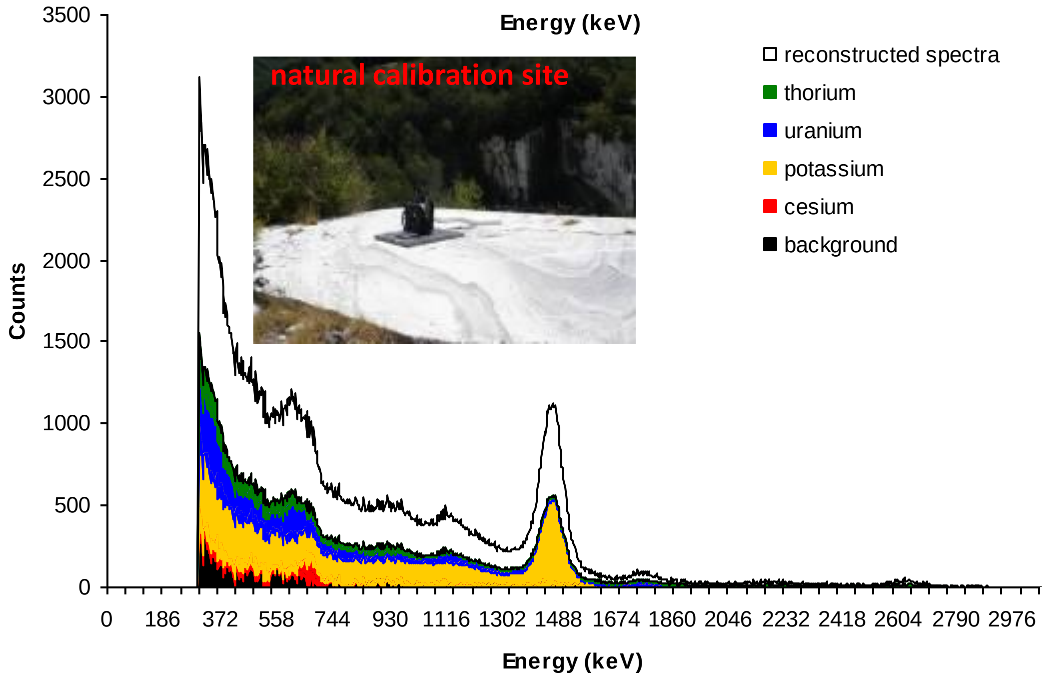
Portable γ–ray spectrometer: calibration methods



The conventional "stripping method" [IAEA 2003] consider the K, eU, eTh window count rates [N] (background corrected) obtained over the pads are linearly related to the concentrations [C] in the pads.

$$[N] = [S] \times [C]$$

[S] – 3 x 3 matrix of sensitivities.



The "full spectrum analysis" method consider the spectra composed by a number of standard spectra as the linear combination.

$$[N]^i = \sum_{j=1}^{m} [C]_j [S]^i_j$$

i (1 to n) channels and j (1 to m) standard spectra.

γ–ray spectroscopy in situ

The ZaNal_1.0L is used in different site and it is acquired a total number of 338 spectrums in-situ.

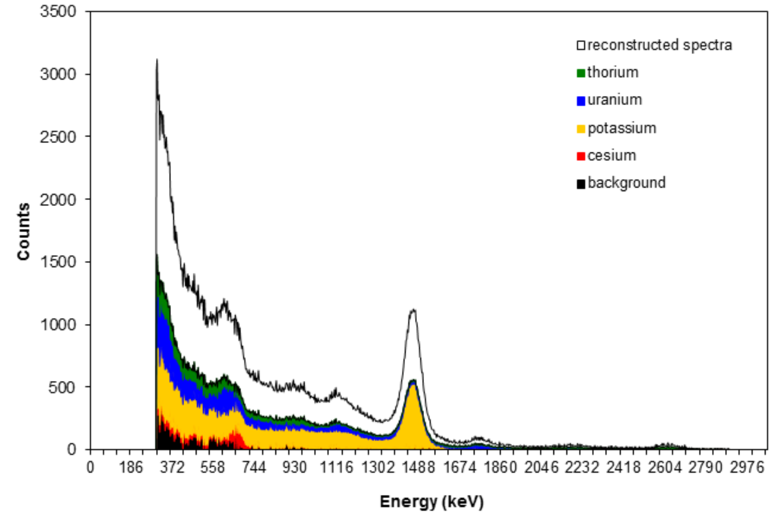
Time of acquisition for every single spectrum: 5 minutes

Placement of instrument	Number of sites
ZaNal_1.0L placed on ground	80 sites (Ombrone Basin)
ZaNal_1.0L placed on a tripod at 1m height	80 sites (Ombrone Basin)
ZaNal_1.0 placed on the shoulders of an operator	89 sites (Schio District)
ZaNal_1.0L placed on ground	89 sites (Schio District)

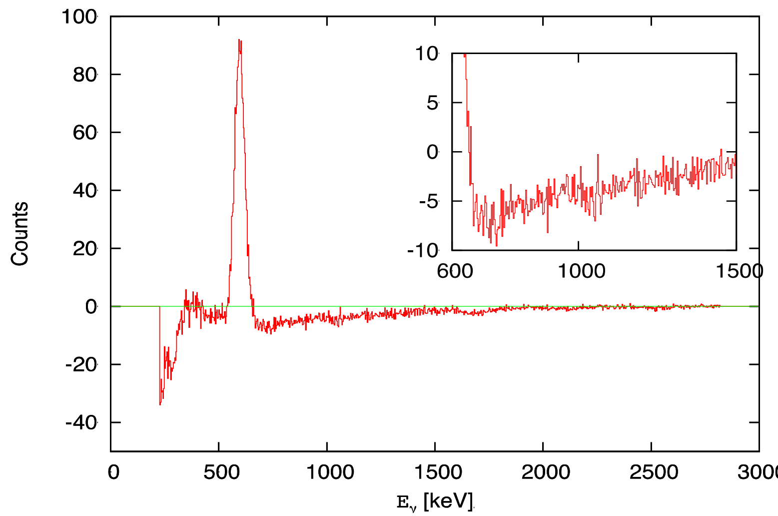
FSA with Non-negative least square constrain

In FSA method the shape of the total spectrum is taken into account and is 'unfolded' into the spectra for the individual radionuclides (*standard spectra*) and a background spectrum.

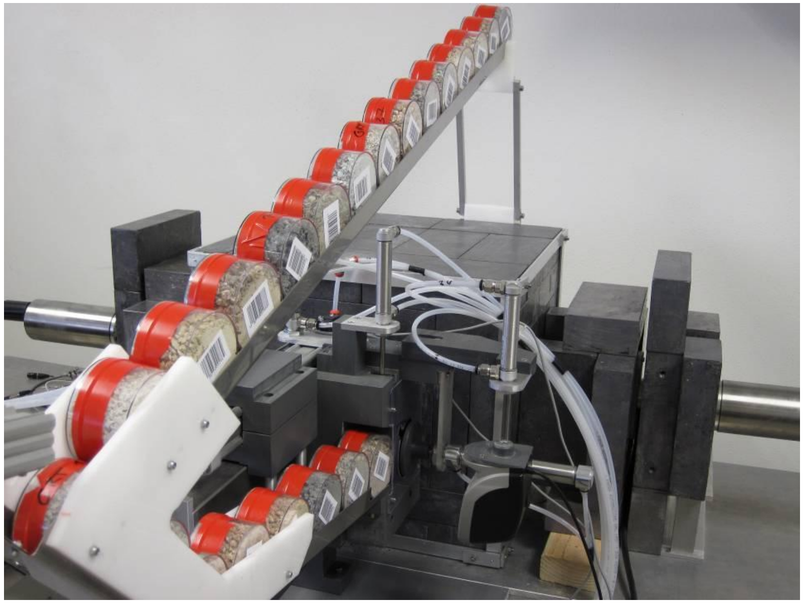
Obtained the *standard spectra* from the calibration, a Non Negative Least-Square (NNLS) procedure is used to find the optimal activity concentrations



- ✓ **It is studied the full range of energy 450-2900 keV**
- ✓ **The structural features of the spectrum are included**
- ✓ **It is investigated the presence of additional radionuclides such as ^{137}Cs.**



The MCA_Rad system



J Radioanal Nucl Chem
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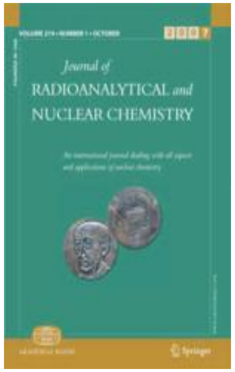
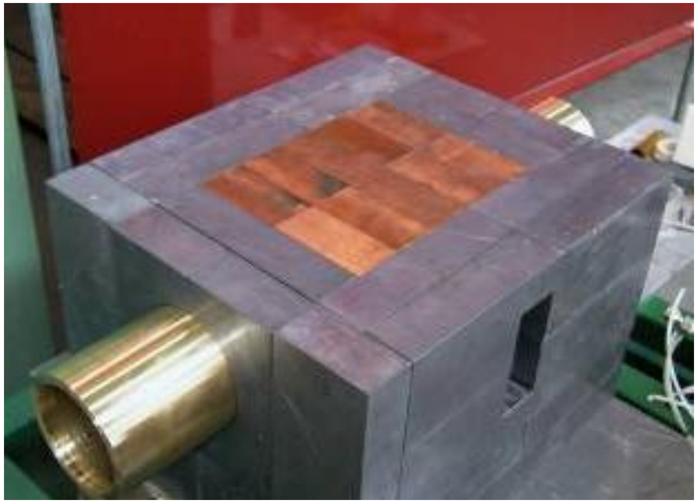
The worldwide NORM production and a fully automated gamma-ray spectrometer for their characterization

G. Xhixha · G. P. Bezzon · C. Broggini · G. P. Buso · A. Caciolli · I. Callegari · S. De Bianchi · G. Fiorentini · E. Guastaldi · M. Kaçeli Xhixha · F. Mantovani · G. Massa · R. Menegazzo · L. Mou · A. Pasquini · C. Rossi Alvarez · M. Shyti





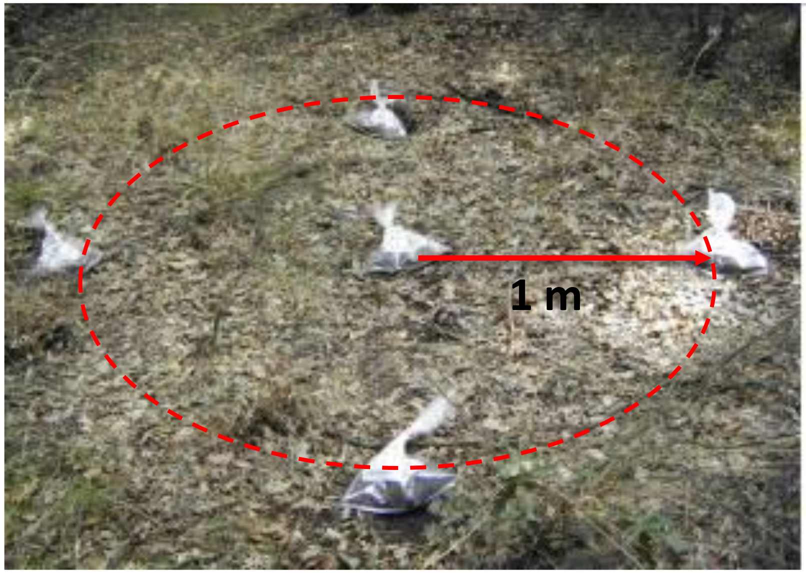
HPGe detectors	Coaxial p-type, 60% of rel. eff.
Energetic resolution	1.9 keV at 1.33 MeV (^{60}Co)
Cooling technology	Electromechanical (~ -190°C)
Shielding composition	10 cm Pb and 5 cm of Cu
Standard acquisition time	1 hour (180 cc sample volume)
Automatic sample manage	24 samples

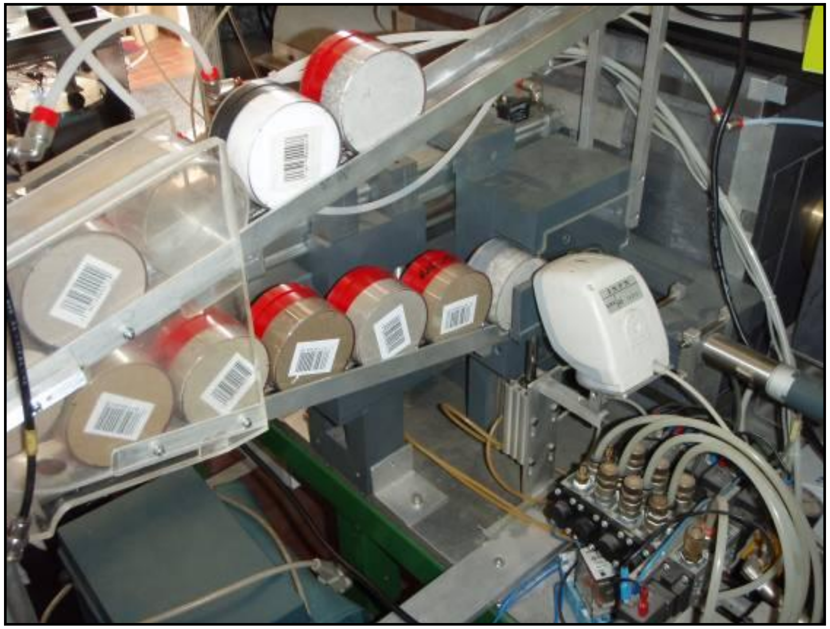
γ–ray spectroscopy in laboratory

Five soil samples on each selected place

- ✓ One central point is ZaNaI_1.0L position
- ✓ The other 4 samples 1m apart

A total of **400 soil samples from 80 places** were measured and analysed using a HPGe setup in the University of Ferrara





DataBase Rad_Nat.mdb

Environmental parameters to define in the campaign:

- temperature
- humidity
- climatic conditions
- density of vegetative cover
- tipology of vegetative cover

Microsoft Access - [Temp_Hum_Tuscany : Tabella]

File Modifica Visualizza Inserisci Formato

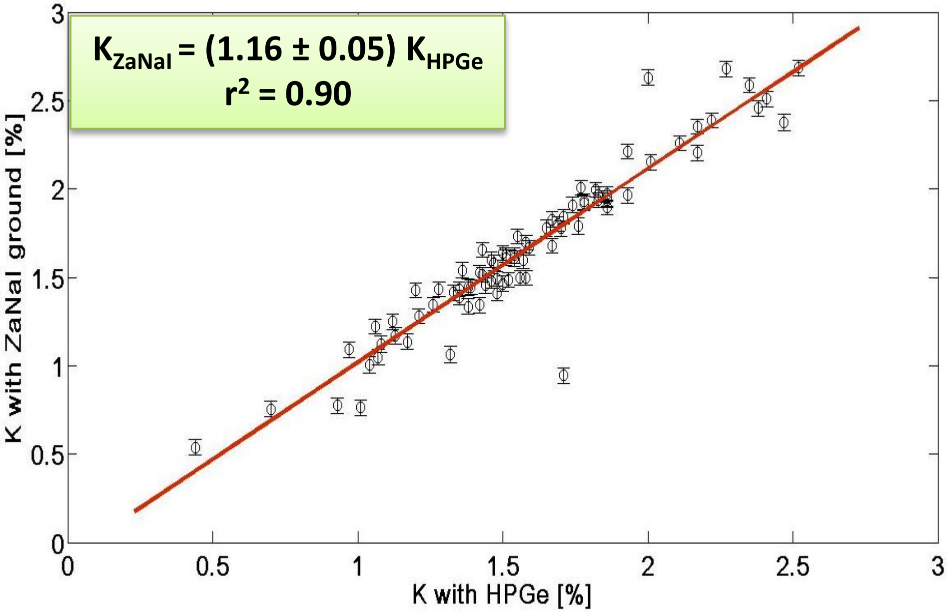
ID_GPS	Temp (°C)	Hum (%)
GPS_1200	40,96	35,8
GPS_1205	31,01	48,58
GPS_1210	37,27	34,52
GPS_1215	39,1	51,84
GPS_1220	43,23	34,43
GPS_1225	43,44	26,46
GPS_1230	28,3	48,53
GPS_1235	42,47	24,7
GPS_1240	26,37	54,95
GPS_1245	24,94	60,38
GPS_1250	29,66	54,5
GPS_1255	28,48	56,77
GPS_1260	36,05	26,95
GPS_1265	43,51	21,42
GPS_1270	39,23	21,68

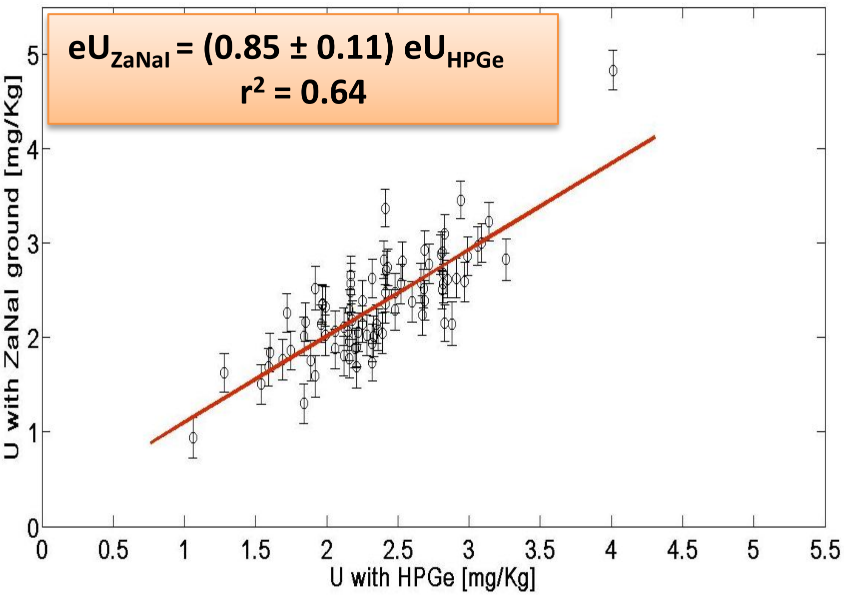
Pedological parameters to define in the campaign:

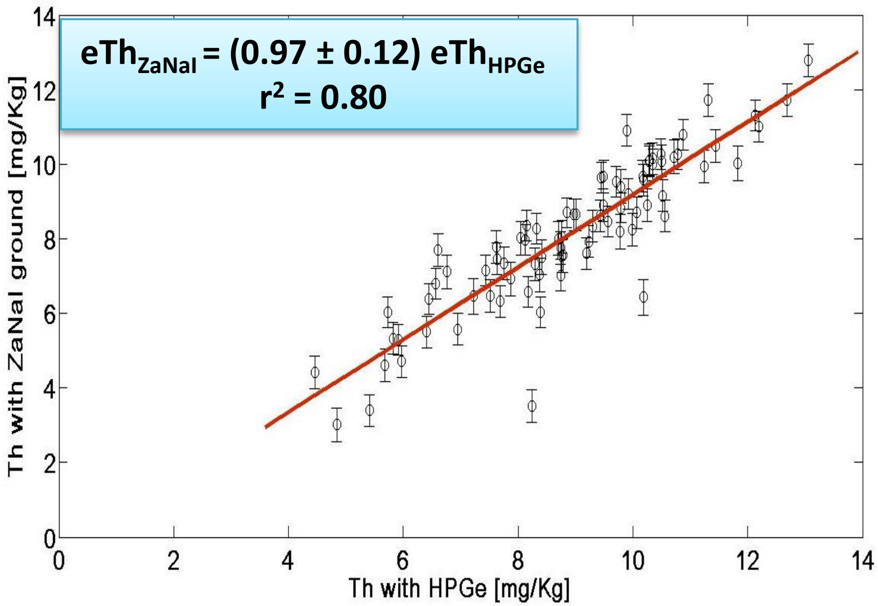
- litology
- granulometry
- rockness
- soil_use
- PH of soil
- colours
- water content

1	ID_Soil	ID_GPS	Soil_Colour	Speckled	Weaving	Lithology	Soil_Use	PH	Depth_Max_(cm)
2	Soil_1200	GPS_1200	3	1	12	SEDIMENTi MARINI	110	8	10
3	Soil_1201	GPS_1201	3	1	12	SEDIMENTi MARINI	110	8	10
4	Soil_1202	GPS_1202	3	1	12	SEDIMENTi MARINI	110	8	10
5	Soil_1203	GPS_1203	3	1	12	SEDIMENTi MARINI	110	8	10
6	Soil_1204	GPS_1204	3	1	12	SEDIMENTi MARINI	110	8	10
7	Soil_1205	GPS_1205	3	1	15	SEDIMENTi MARINI	200	8	10
8	Soil_1206	GPS_1206	3	1	15	SEDIMENTi MARINI	200	8	10

Correlation between in-situ acquisition on ground and laboratory measurements





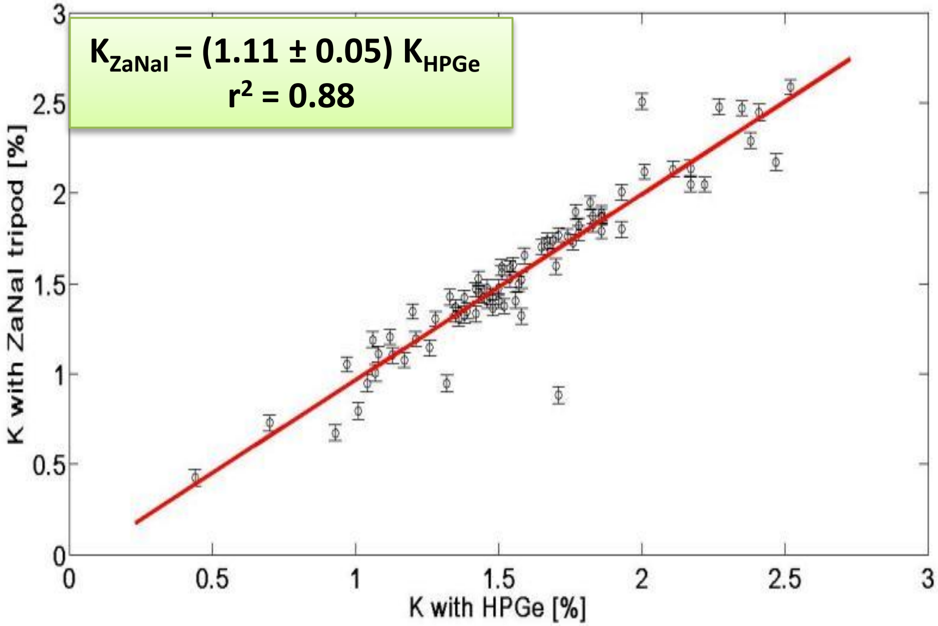


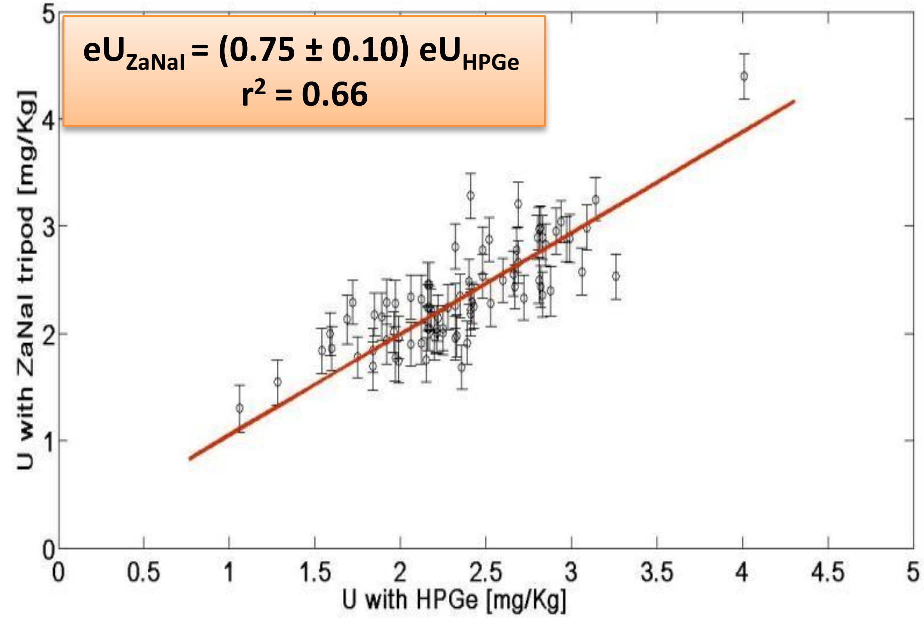
The correlation parameters obtained for in-situ measurements on ground and laboratory measurements.

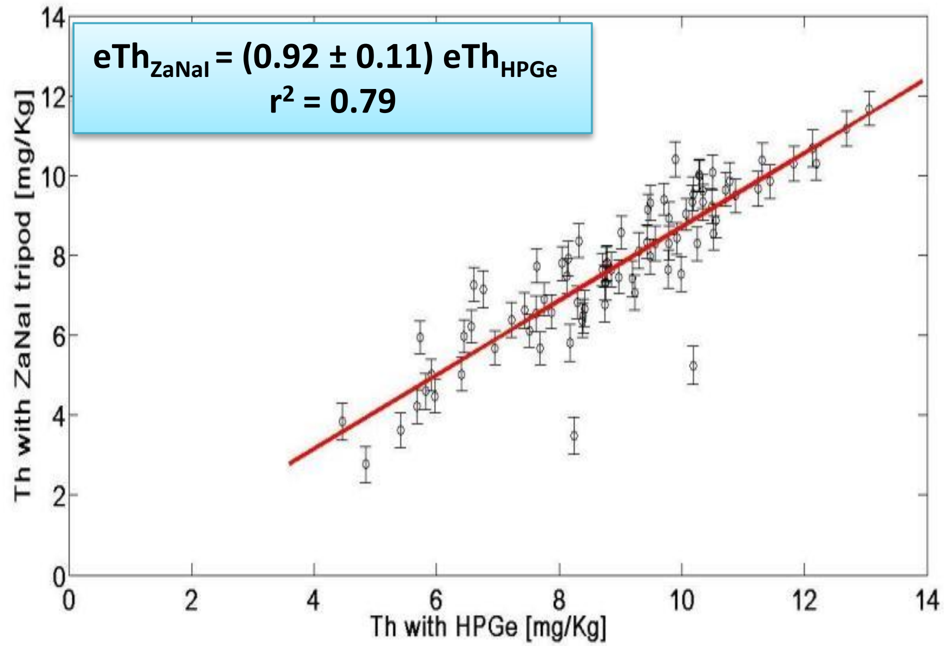
Isotopes	$a \pm \sigma_a$	R^2
K [%]	1.16 ± 0.05	0.90
U [mg/Kg]	0.85 ± 0.11	0.64
Th [mg/Kg]	0.97 ± 0.12	0.80

- **A good correlation between in-situ and laboratory measurements.**
- **The linear regression coefficient r^2 obtained for K and Th is very close to unity, while for U the data are more dispersed due to the effect of atmospheric radon.**
- **For Th the data are comparable within 1σ, while for U within 1.5σ and for K for more than 3σ.**
- **The final relative uncertainties for K, U and Th are less than about 20%, respectively 13%, 19% and 12%.**

Correlation between in-situ acquisition on tripod and laboratory measurements





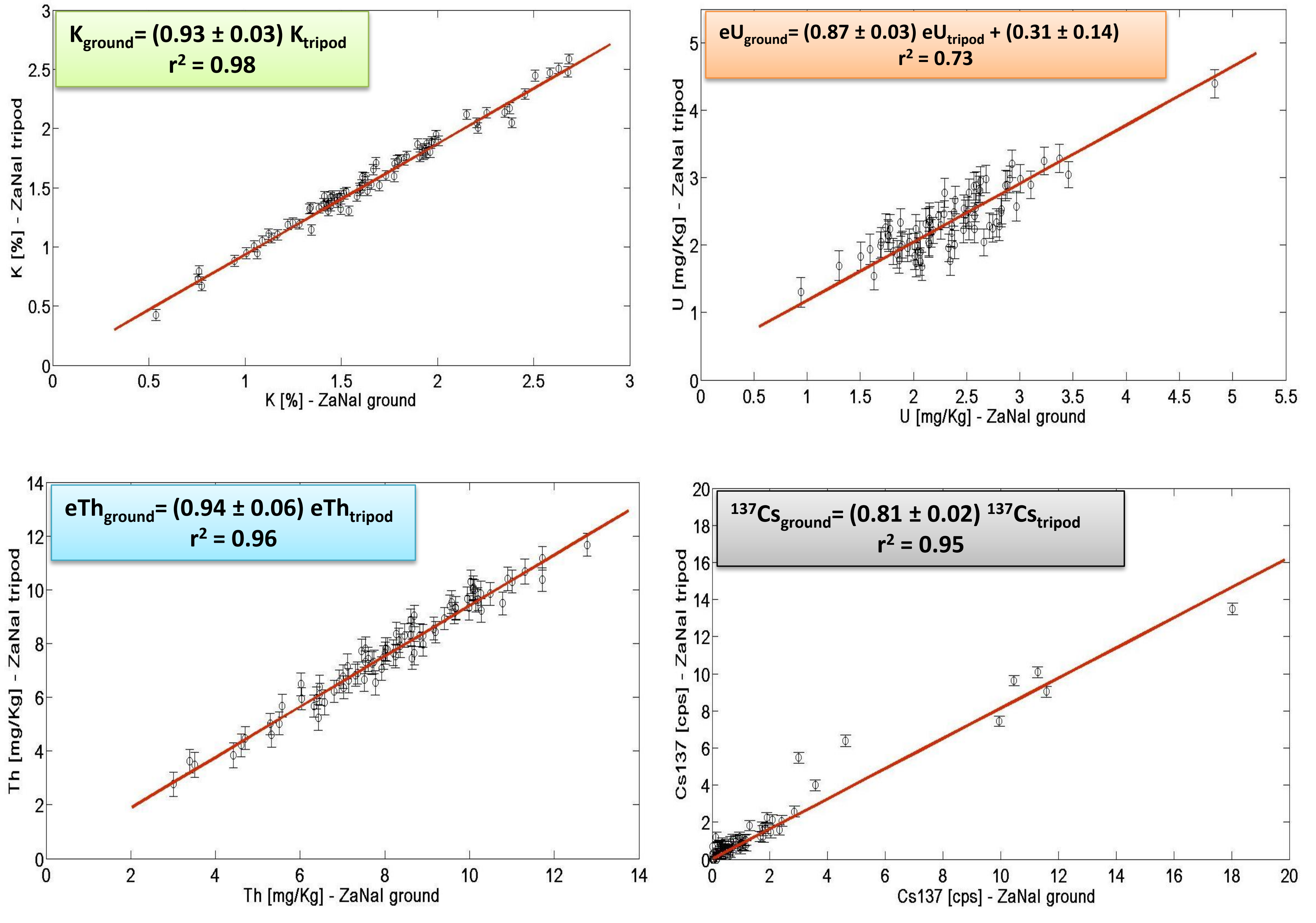


The correlation parameters between measurements in-situ acquisition on tripod and in the laboratory.

Isotopes	$a \pm \sigma_a$	R^2
K [%]	1.11 ± 0.05	0.88
U [mg/Kg]	0.75 ± 0.10	0.66
Th [mg/Kg]	0.92 ± 0.11	0.79

- **Good correlation between in-situ and laboratory measurements.**
- **The linear regression coefficient shows similar results as those obtained for in-situ measurements placing the detector on ground. For Th the data are comparable within 1σ, while for U within 2.5σ and for K for more than 2σ.**
- **The final relative uncertainties for K, U and Th are less than about 35%, respectively 10%, 33% and 12%.**
- **The increase of discrepancy between the two data sets can be attributed to the attenuation of 1m air for in-situ measurement performed by placing the detector on tripod.**
- **The attenuation due to 1 m air can be calculated as the difference between ground and tripod in-situ measurements compared with laboratory measurements, and are 5 ± 0.3 %, 10 ± 1.9 % and 5 ± 0.9 % respectively for K, U and Th.**

Correlation between in-situ acquisition on ground and on tripod



Correlation parameters between in-situ measurements on ground and on tripod

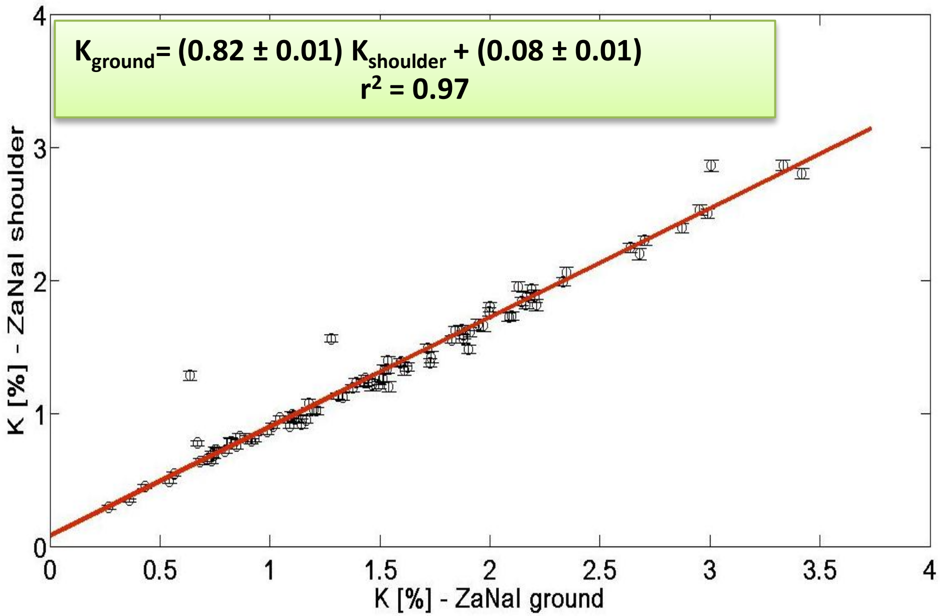
Isotopes	$a \pm \sigma_a$	$b \pm \sigma_b$	r^2
K [%]	0.93 ± 0.03	-	0.98
U [mg/Kg]	0.87 ± 0.06	0.31 ± 0.14	0.73
Th [mg/Kg]	0.94 ± 0.06	-	0.96
^{137}Cs [cps]	0.81 ± 0.02	-	0.95

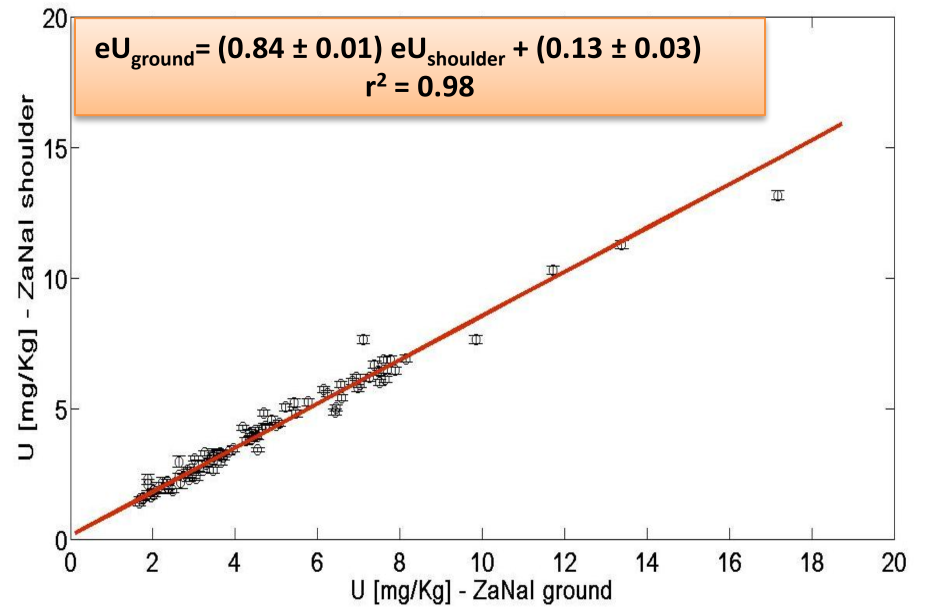
- **There is a very good correlation between in-situ measurements on ground and on tripod. Linear regression coefficient close to unity are an evidence of the homogeneity of the selected sites.**

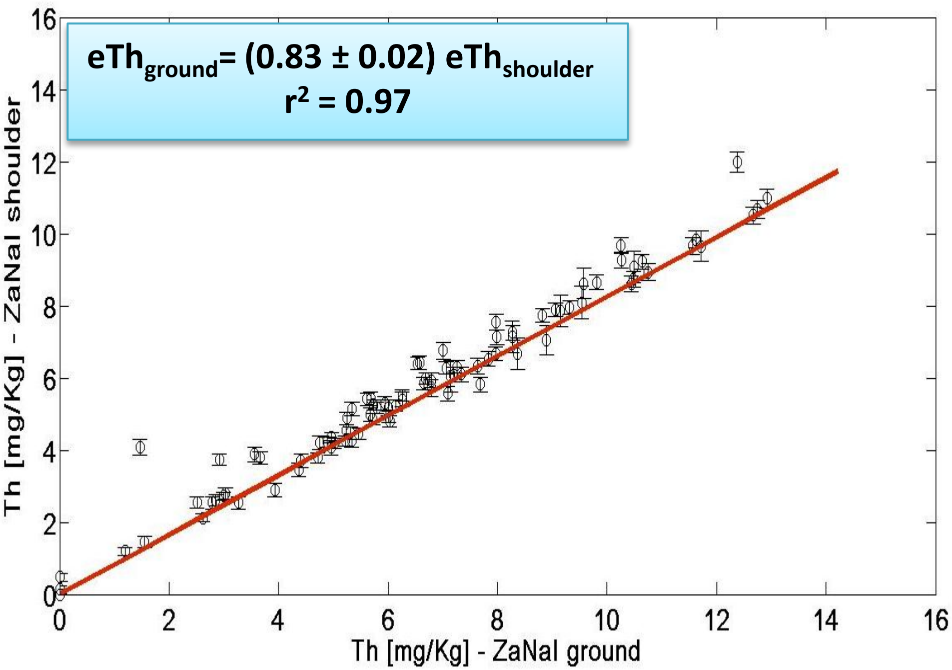
- **The deviation between the angular coefficients and the unity value quantifies the correction of the signal due to the attenuation effect of 1 m air, obtaining for ^{40}K, ^{238}U, ^{232}Th and ^{137}Cs respectively 7 ± 0.3%, 13 ± 0.9 %, 6 ± 0.4% and 19 ± 0.5%.**

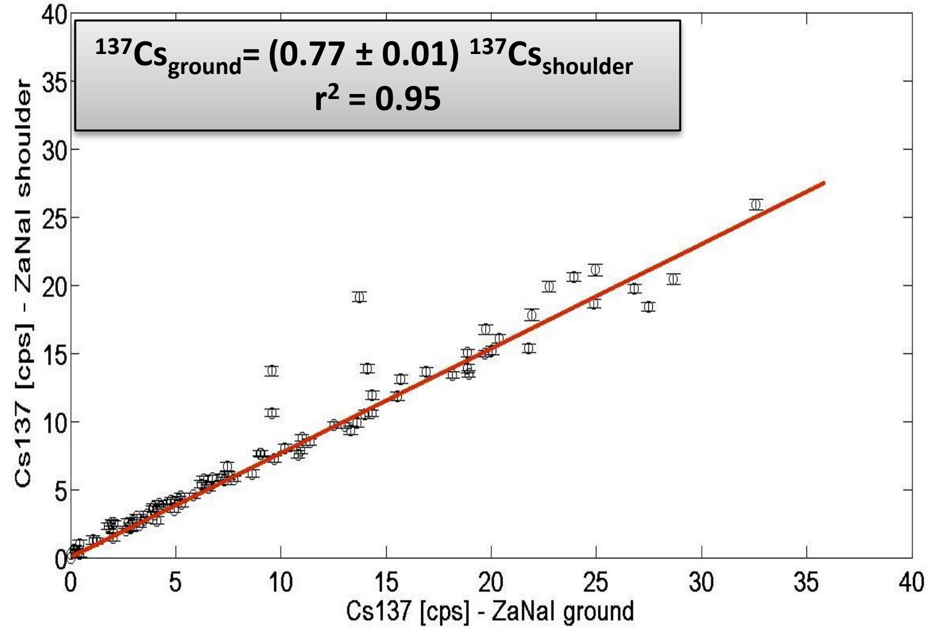
- **For ^{137}Cs the attenuation is higher due to the fact that it emits a gamma ray with relative lower energy (662 keV).**

Correlation between in-situ acquisition on ground and on operator shoulder









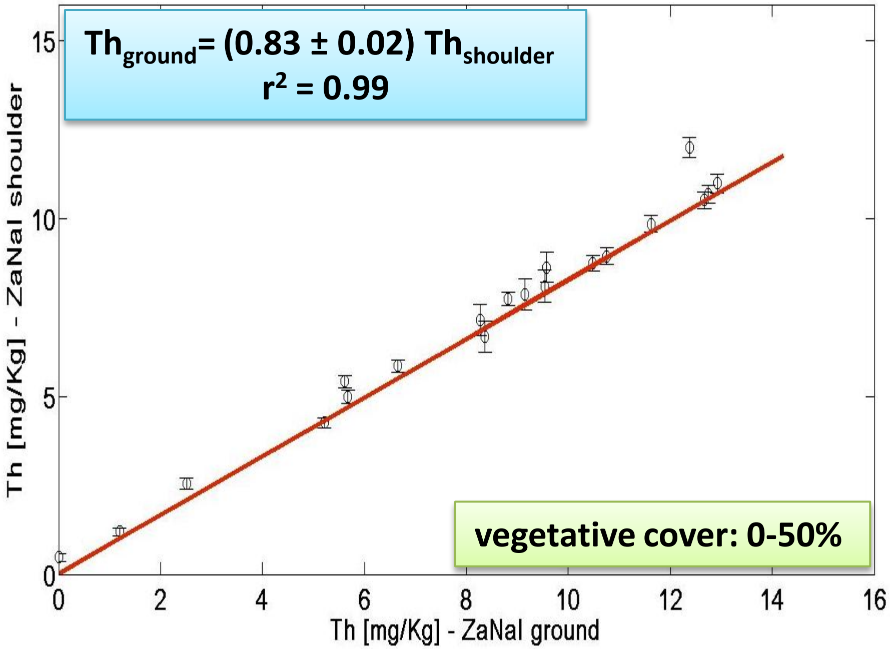
Correlation parameters between in-situ measurements on ground and on shoulder

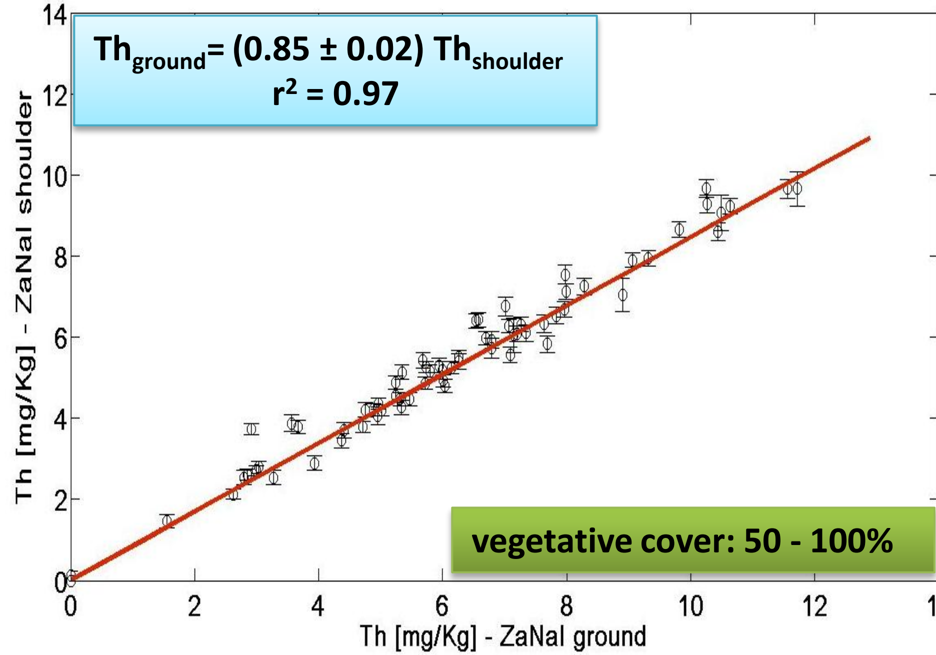
Isotopes	$a \pm \sigma_a$	$b \pm \sigma_b$	r^2
K [%]	0.82 ± 0.01	0.08 ± 0.01	0.97
U [mg/Kg]	0.84 ± 0.01	0.13 ± 0.03	0.98
Th [mg/Kg]	0.83 ± 0.02	-	0.97
^{137}Cs [cps]	0.77 ± 0.01	-	0.95

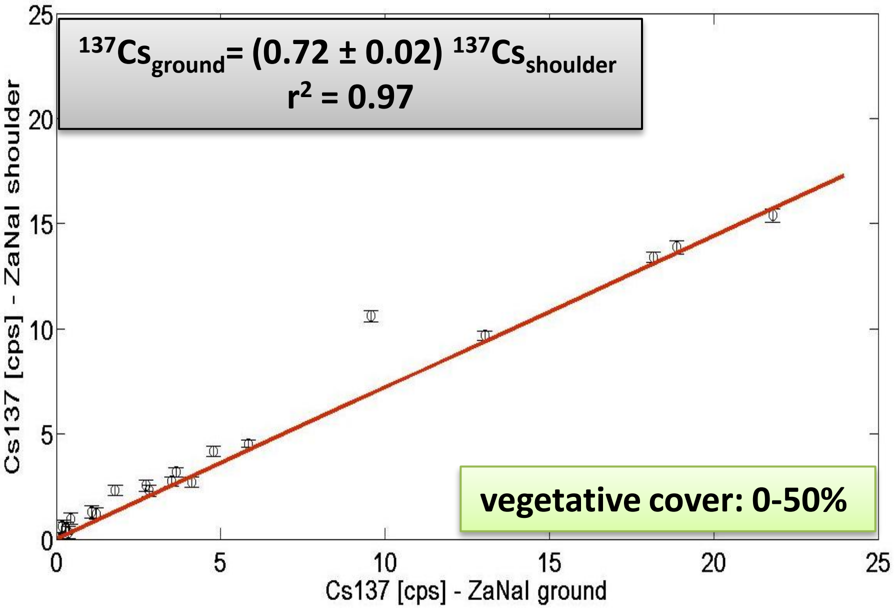
- **A very good correlation between in-situ measurements on ground and on shoulder.**

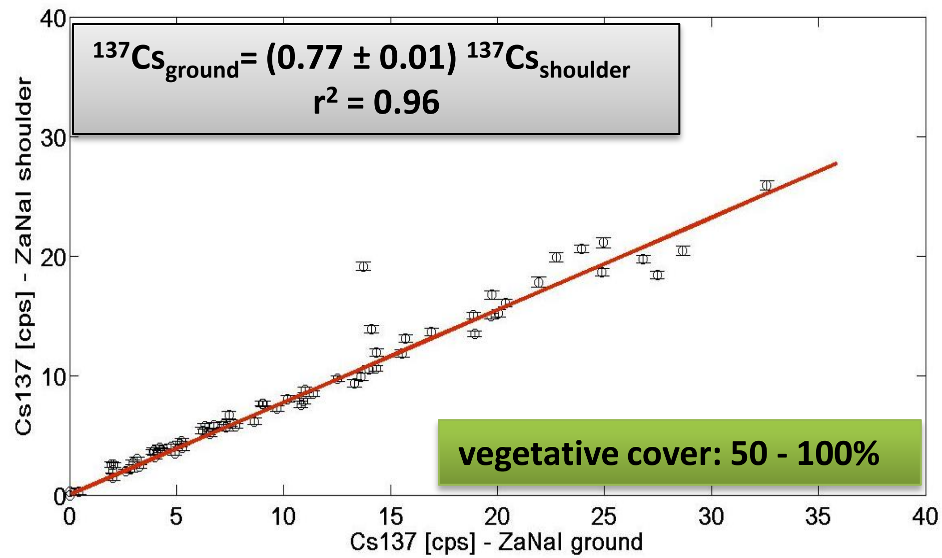
- **The deviation between the angular coefficients and the unity value quantifies the correction of the signal due to the presence of an operator, obtaining for ^{40}K, ^{238}U, ^{232}Th and ^{137}Cs respectively 18 ± 0.2%, 16 ± 0.2 %, 17 ± 0.4% and 23 ± 0.3%.**

Interference of vegetative cover for-situ acquisition on ground and on operator shoulder









Correlation parameters between in-situ measurements on ground and on shoulder for two classes of vegetative coverage

Isotopes	$a \pm \sigma_a$	
	Vegetative coverage 0-50 %	Vegetative coverage 50-100 %
Th [mg/kg]	0.83 ± 0.02	0.85 ± 0.02
^{137} Cs [cps]	0.72 ± 0.02	0.77 ± 0.01

- **In the case of ^{232}Th it is observed a minor degree of influence due to the vegetative cover from 50-100%, but comparable within 1σ with the case of 0-50% of vegetative coverage.**
- **In the case of ^{137}Cs it is seen clearly within the 1σ the influence due to the presence of the vegetative cover.**
- **As it is expected, the presence of vegetative cover is more visible in the case of relatively lower energies.**

Conclusions

- Realization of extensive measurements (80 sites) investigated both in-situ using ZaNaI_1.0L (FSA-NNLS method) and in laboratory using MCA_Rad showing a very good correlation between them.

Isotope	$[ZaNaI]_{ground} = (a \pm \sigma_a)$ [HPGe]	$[ZaNaI]_{tripod} = (a \pm \sigma_a)$ [HPGe]
^{40}K (%)	1.16 ± 0.05	1.11 ± 0.05
eTh (mg/kg)	0.85 ± 0.11	0.75 ± 0.10
eU (mg/kg)	0.97 ± 0.12	0.92 ± 0.11

- The final relative uncertainties for K, U and Th are found to be less than about 20% for ZaNaI_1.0L on ground versus HPGe measurements and about 35 % ZaNaI_1.0L on tripod versus HPGe measurements.

- In the case of ^{238}U, correction is much more complex during the measurement in situ because the presence of Radon in air distorts our signal.

Conclusions

- By using a ZaNaI_1.0L detector in situ measurements it is evaluated experimentally the corrections between different configurations (on the ground, at 1m height and on the shoulders)

Isotopes	Max. Energy [keV]	Correction at 1 m height [%]	Air + operator attenuation correction [%]
^{40}K [%]	1460	7 ± 0.3	18 ± 0.2
eTh [mg/kg]	2615	6 ± 0.4	17 ± 0.4
eU [mg/kg]	1764	13 ± 0.9	16 ± 0.2
^{137}Cs [cps]	662	19 ± 0.5	23 ± 0.3

- As expected from theoretical models, the corrections for different configurations are lower for gamma rays with high energy.

- In the obtained concentrations of radioactive nuclides are studied several parameters as environmental conditions, operational circumstances and vegetative cover for their interference. In several parameters that are taken into account it is seen a light influence of them, but inside the errors 1 sigma we can give a clear result only for the influence of vegetative cover in the case of ^{137}Cs.

Peer-reviewed scientific papers

1. Xhixha G. et al (2013). *First Characterization Of Natural Radioactivity In Building Materials Manufactured In Albania*. Journal of Radiation Protection Dosimetry. doi: 10.1093/rpd/ncs334.

2. Xhixha G. et al. (2012). *Fully automated gamma-ray spectrometer for NORM characterization*. Journal of Radioanalitical and Nuclear Chemistry, 1-13. doi: 10.1007/s10967-012-1791-1.

3. Caciolli A. et al. (2012). A new FSA approach for in situ γ-ray spectroscopy. Science of the Total Environment 414 (2012) 639–645.

4. Cfarku F. et al. (2011). *Radioactivity Monitoring in Drinking Water of Albania*. J. Int, Environmental Protection & Ecology, ISSN 1311-5065, Vol. 12, Nr. 3 - p.1116.

5. Bode K. et al. (2010). Results Of The National Survey On Radon Indoors In Albania. doi: 10.1063/1.3322533, ISSN 0094-243, ISBN 978- 0-7354-0740-4. American Institute of Physics.

Conference proceedings and papers not peer-reviewed

1. 1. Mou L. et al. (2011). *Nuovo spettrometro gamma per il monitoraggio della radioattività in situ*. Mus. Civ. Rovereto, Atti del Workshop in geofisica, 59-72.

2. Bezzon G.P. et al. (2011). *Mapping of natural radioelements using gamma-ray spectrometry: Tuscany Region case of study.* ISSN 1828-8545, INFN-LNL Rep. 234.

3. Bezzon G.P. et al. (2011). *A γ-Spectroscopy System for Atmospheric Radon Detection*. ISSN 1828-8545, INFN-LNL Rep. 234.

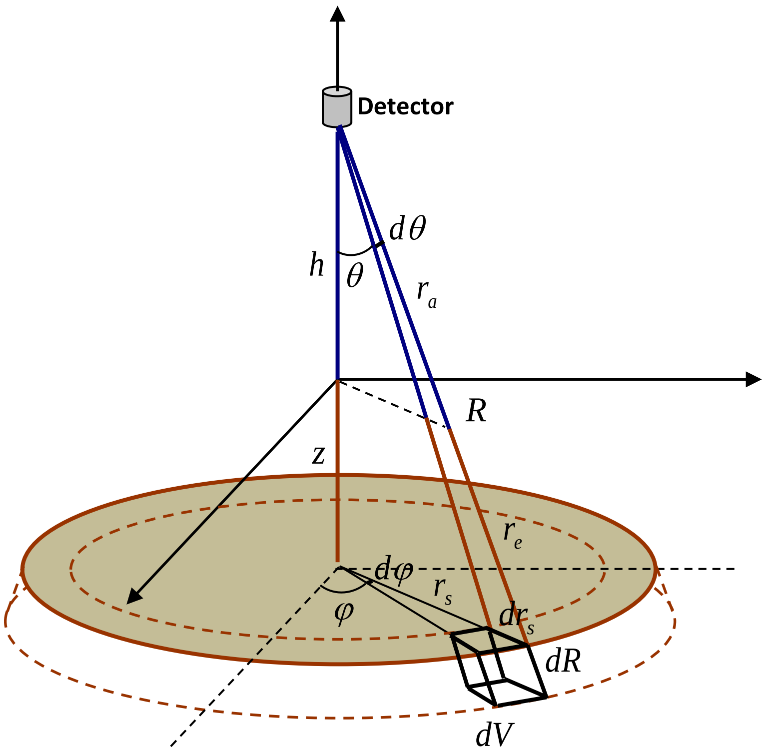
4. 4. Bezzon G.P. et al. (2010). *Preliminary results for the characterization of the radiological levels of rocks in Tuscany Region*. Atti 85° Congr. Soc. Geol. It., vol. 11, 513-514.

5. Cfarku F. et al. (2009). Determination of alpha and total beta radiation in water by the GPC method (gas proportional counters). Bulletin of Natural Science No. 7, 83-88.


Thank

Theoretical models

The number of photons detected per second in the thorium window is given by[1]:



$$N = \varphi A \varepsilon$$

where:

φ - flux of 2.62 MeV photons at the detector

A - cross-section area of the detector

ε - photopeak efficiency

$$N = \frac{A\varepsilon n}{2\lambda} \int_1^\infty \frac{e^{-\mu h \sec\theta} d(\sec\theta)}{(\sec\theta)^2} = N_0 E_2(\mu h)$$

μ and λ - linear absorption coefficients for air and soil

N_0 – thorium count rate at ground level

The theoretical models depend on the calculation of exponential integral of second kind:

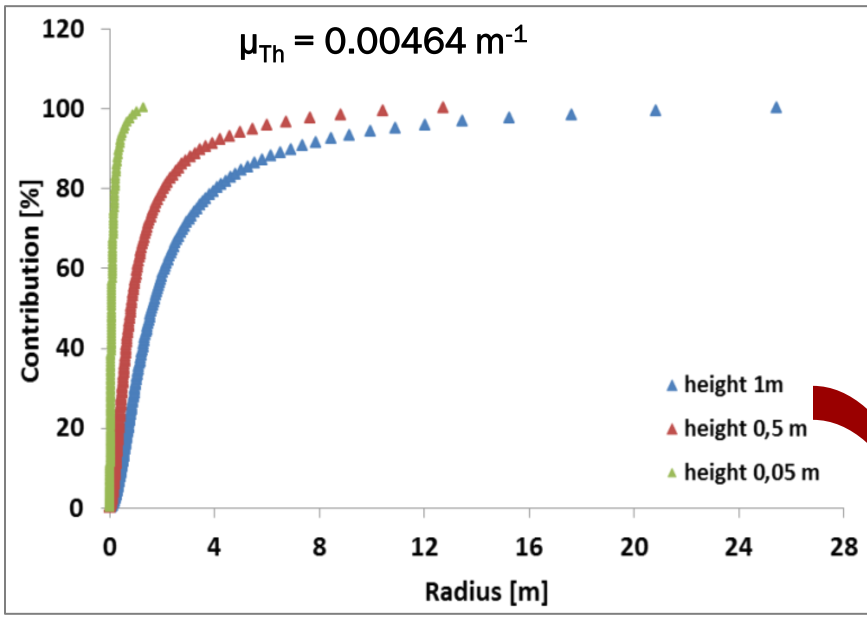
$$E_2(\mu h) = \int_1^\infty \frac{e^{-\mu h t} dt}{t^n}$$

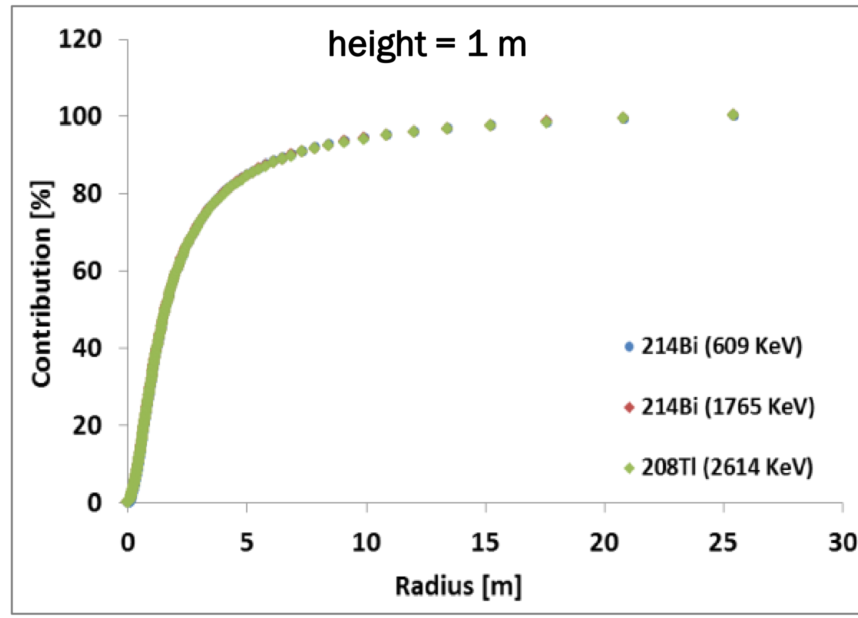
[1] R. L. Grasty., 1975 Atmospheric Absorption of 2.62 MeV Gamma ray Photons emitted from the ground. Geophysics vol 40, 1058-1065.

Variation in response of a spherical detector versus heights and energies

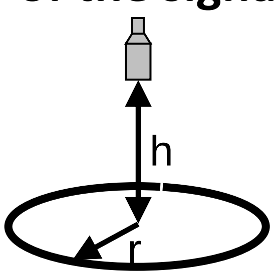
The number of photons N detected above a uniformly radioactive infinite source per unit time is:

$$N = \frac{n}{2\lambda}\int_{1}^{\sec\theta} \frac{A\varepsilon e^{-\mu h \sec\theta}}{\sec^2\theta} d(\sec\theta)$$





At height h, the detector receive 90% of the signal from a circle of radius R

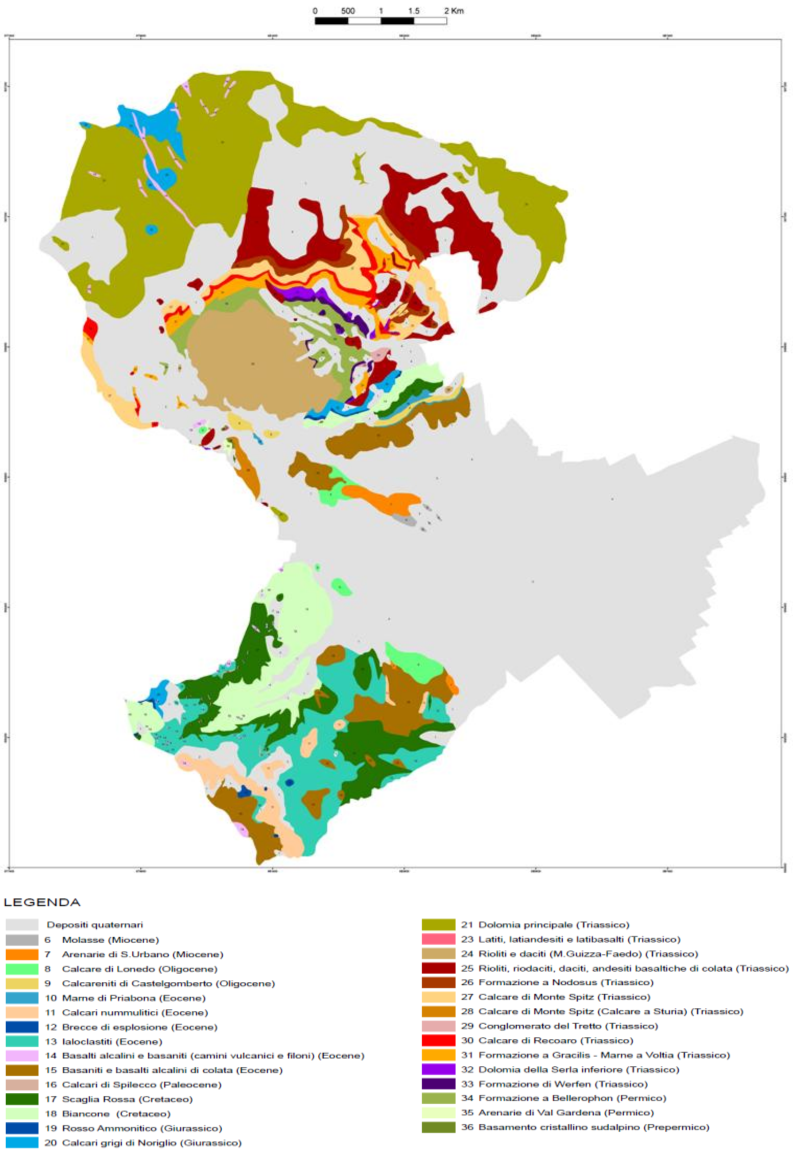


h (m)	0.05	0.5	1.0
R (m)	0.37	3.7	7.35

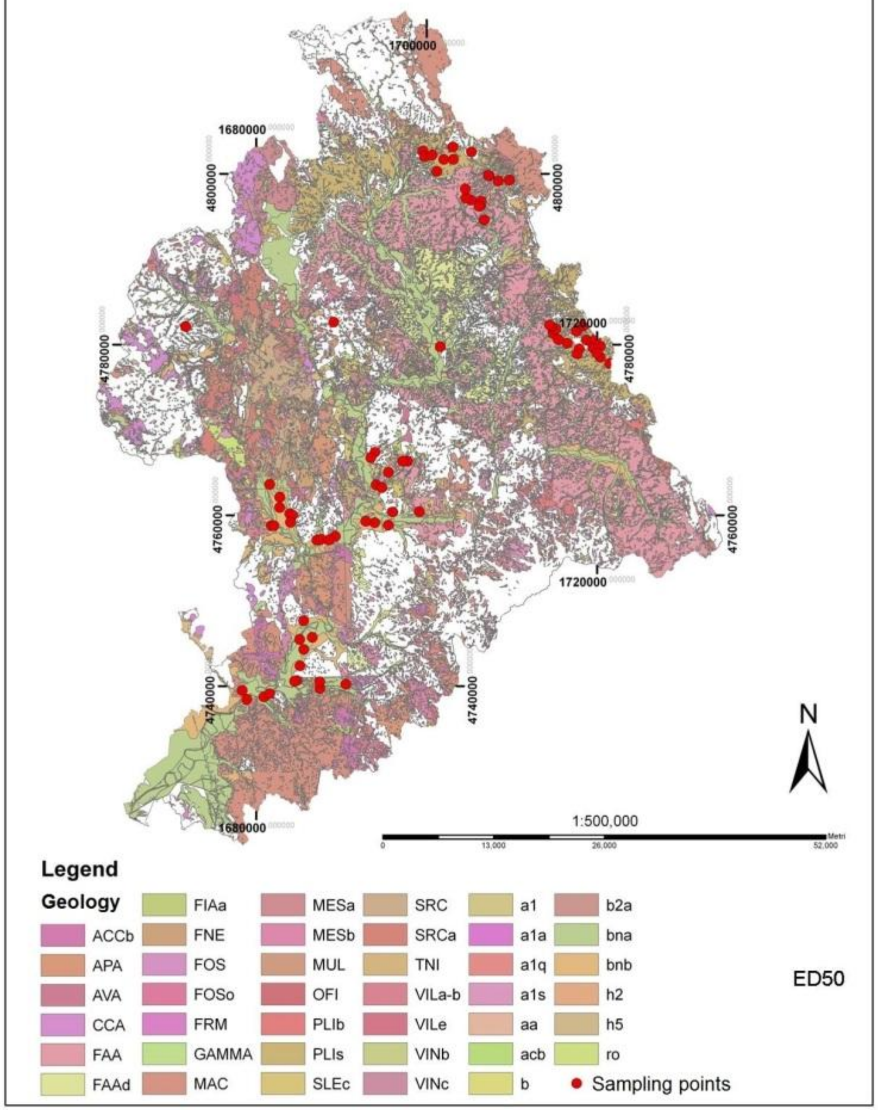
	Energy (keV)	μ_Linear absorption coeff (m^{-1})-air
^{214}Bi	609	0.00990
	1764	0.00558
^{208}Tl	2614	0.00464

Compaign activity

Geological map of the Commune of Schio



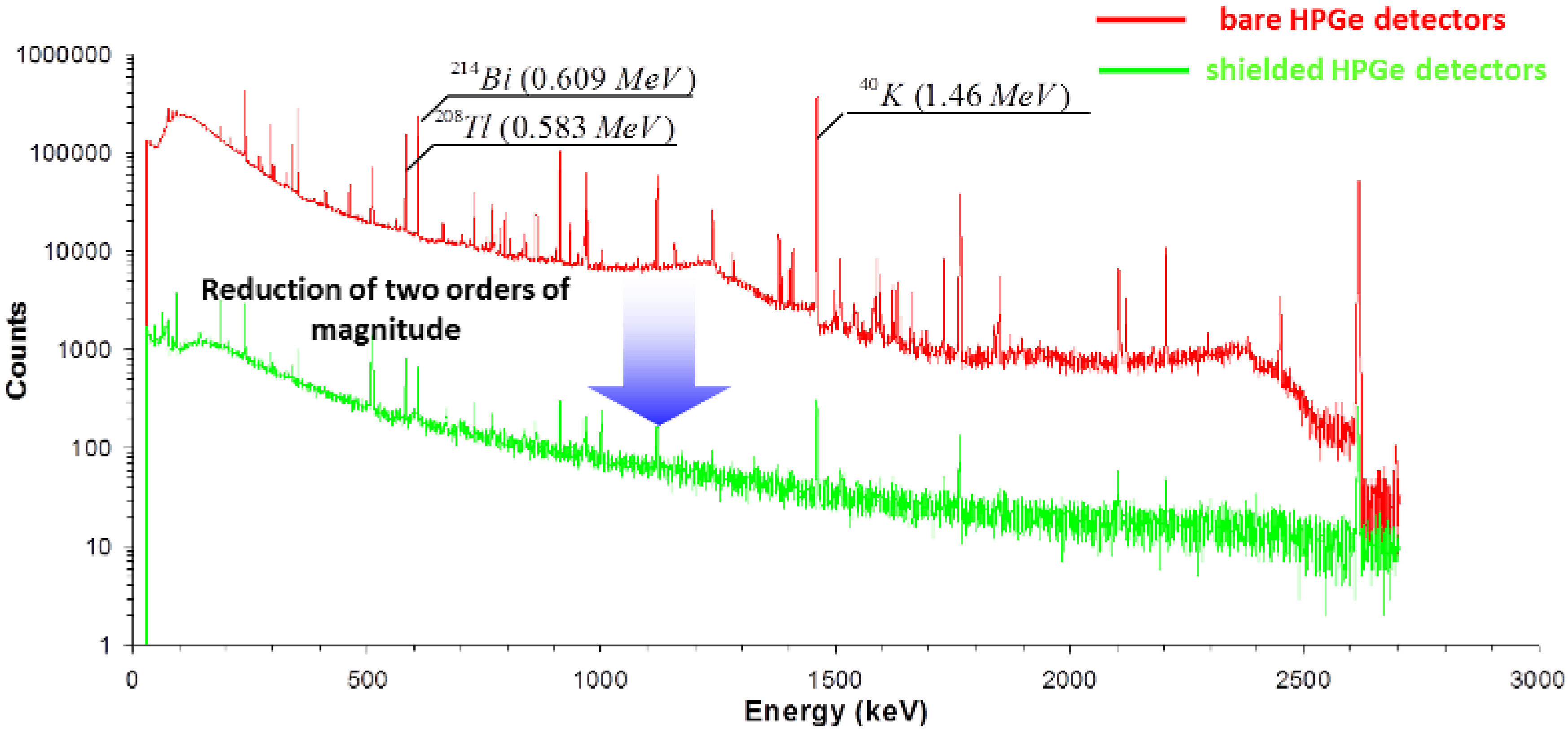
Geological map of Ombrone Basin with the location of sampling points



Background reduction of MCA_Rad system

Estimation of Minimum Detectable Activity (MDA) for blank test **[Curie 1986].**

Isotope	E (keV)	MDA (Bq)
^{40}K	1460	0,26
^{214}Bi	609	0,04
^{208}Tl	583	0,06



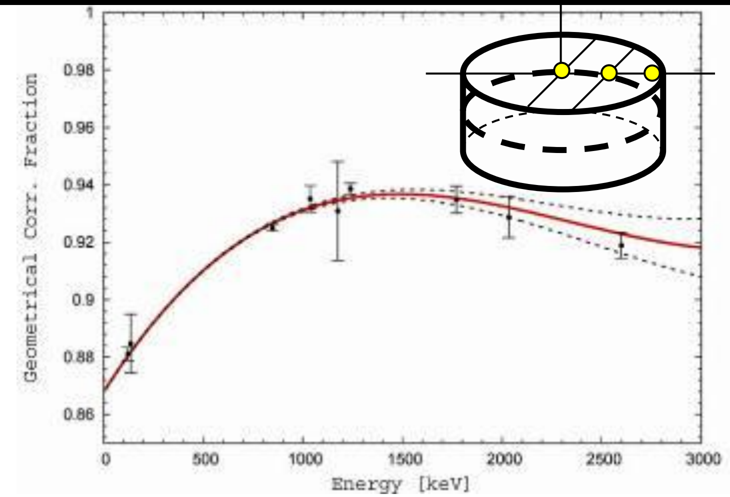
Currie L. A., 1986. Limits for Qualitative Detection and Quantitative Determination Application to Radiochemistry. Analytical Chemistry 40. 586–593.

Efficiency analysis: three main corrections

1- Geometrical correction (C_G): moving the standard point source in three positions (for three planes) It is calculated the C_G for different energies (E_i) fitting the expression.

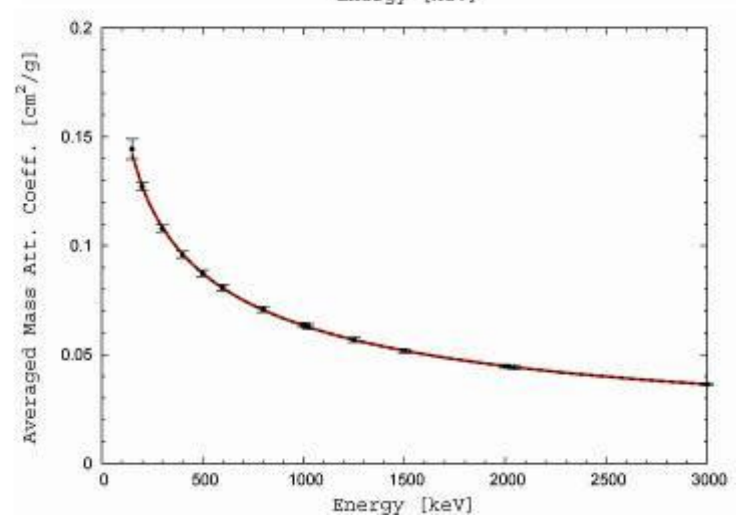
$$C_G = \sum_{i=0}^{3} a_i (E_i / E_0)^i$$

where E_0 = 1keV.



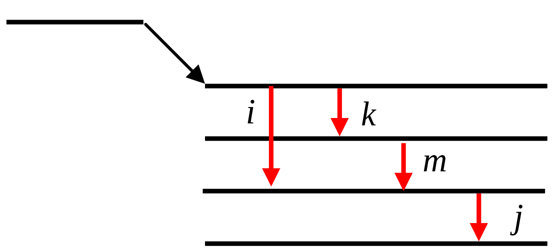
2- Self absorption correction (C_{SA}): averaging the mass attenuation coeff. μ for a "standard rock" with density ρ, It is calculated the C_{SA} for the sample thickness t = 4.5 cm using the simplified approach:

$$C_{SA} = \frac{1 - e^{-(\mu_s \rho_s - \mu_{ref} \rho_{ref})t}}{(\mu_s \rho_s - \mu_{ref} \rho_{ref})t}$$

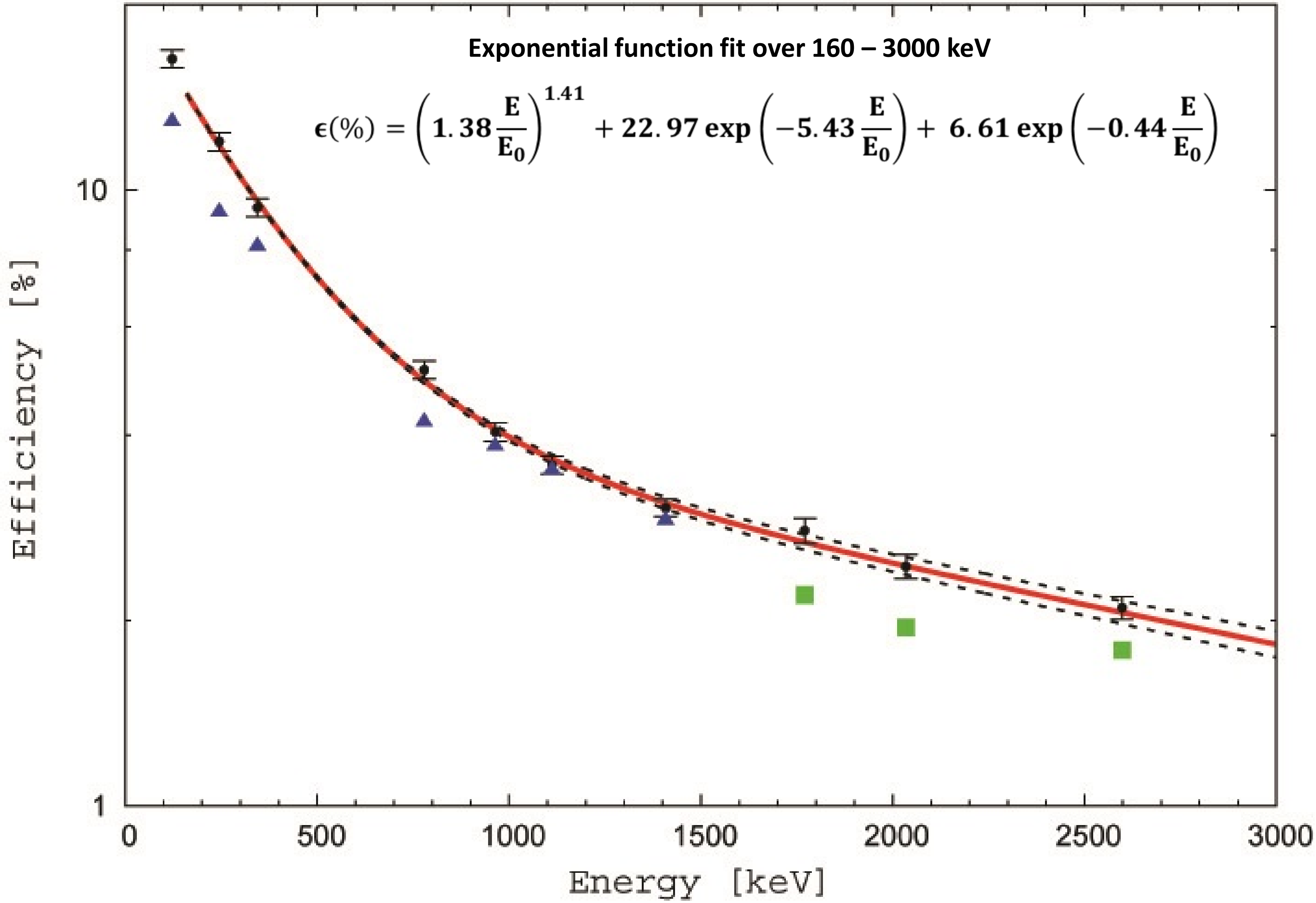


3- Coincidence summing correction (C_{CS}): the correction of (i) events takes into account the summing out (j) and summing in (k,m) and effects:

$$C_{CS(i)} = \left[1 - \frac{\sum_j P_{tij} P_i P_j \varepsilon_{tj}}{I_{\gamma i}}\right]\left[1 + \frac{\sum_{k,m} P_{tkm} P_k P_m \varepsilon_k^{app} \varepsilon_m^{app}}{I_{\gamma i} \varepsilon_i^{app}}\right]$$



Absolute full-peak energy efficiency for MCA_Rad system



$$\epsilon(\%) = \left(1.38\frac{E}{E_0}\right)^{1.41} + 22.97\exp\left(-5.43\frac{E}{E_0}\right) + 6.61\exp\left(-0.44\frac{E}{E_0}\right)$$

Knoll G.F., 1999. Radiation Detection and Measurements, Third Edition, John Wiley & Sons, 1999.