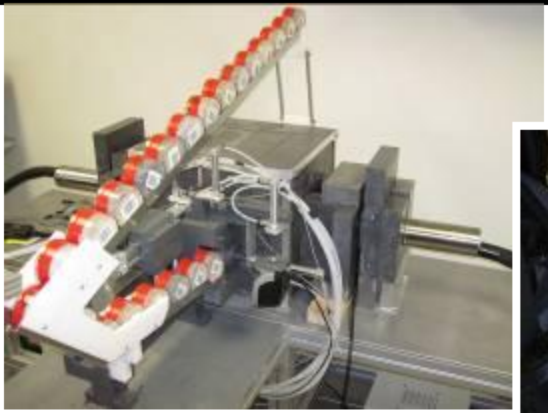
Advances γ-ray spectrometry for environmental radioactivity monitoring



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University of Ferrara
PhD in Physics – XXIV cycle

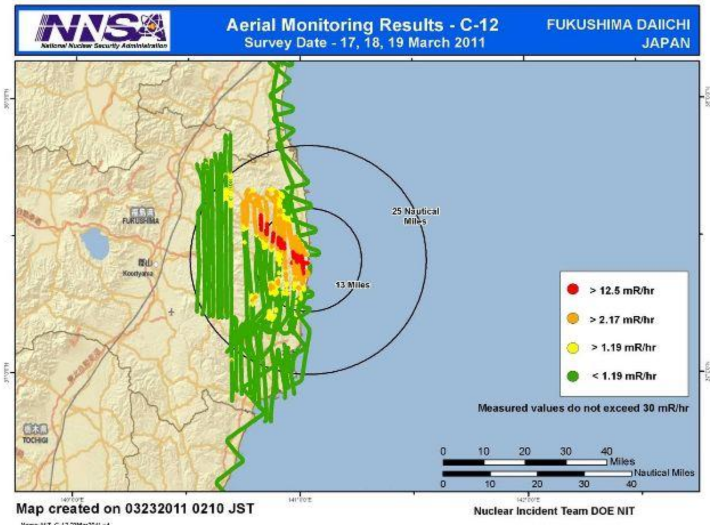
March, 29 2012

Centro di GeoTecnologie CGT — università di ferrara — INFN ISTITUTO NAZIONALE DI FISICA NUCLEARE

Applied Geophysics Laboratory | **Physics Department** | **Legnaro National Laboratory**

Summary

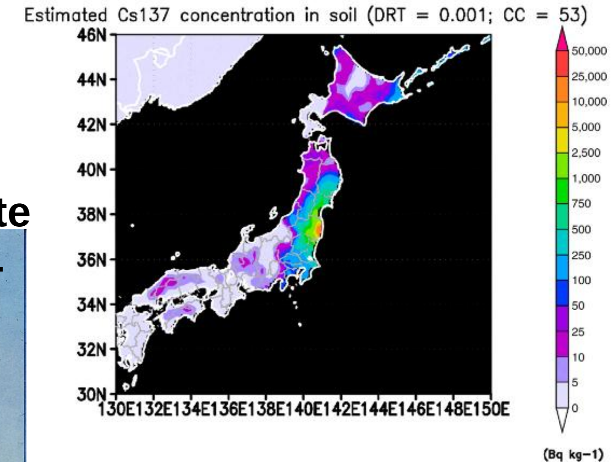
- **Environmental radioactivity monitoring: social, scientific and technological motivations**



- **γ-ray spectrometry techniques:**
  - **Laboratory γ-ray spectrometry**
  - **In-situ γ-ray spectrometry**
  - **Airborne γ-ray spectrometry**

- **Conclusions and prospects**
- **Publications**

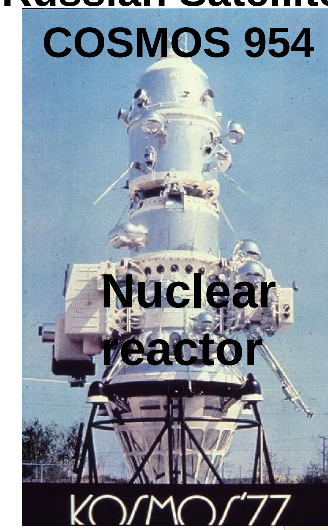
Environmental radioactivity monitoring: social, scientific and technological motivations



Radiation protection monitoring
[Council Directive 96/29/EURATOM 13 May 1996]

Detection of orphan sources and illegal dump deposits [Bristow 1978]

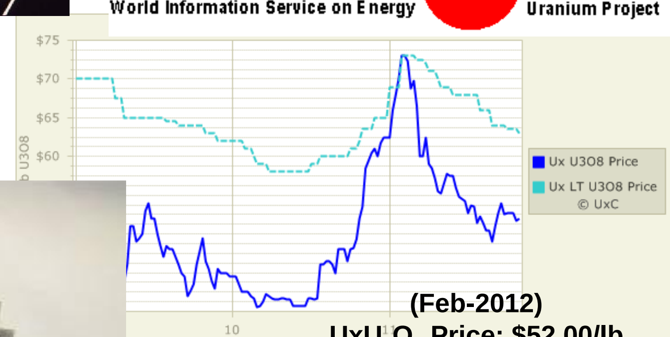
Russian Satellite



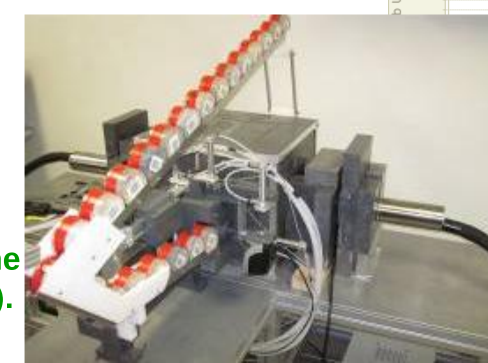
Improvements on γ-ray spectrum analysis [Caccioli, Xhixha et al. 2012]

Improvements on spatial data analysis for geological exploration [Gustaldi, Xhixha et al. 2012]

wise
World Information Service on Energy
Uranium Project



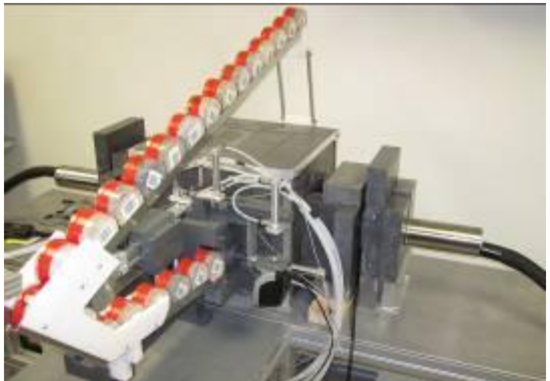
Nuclear techniques for mineral exploration and geophysics application [Xhixha et al. 2012]



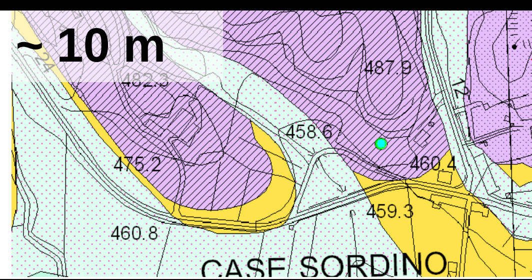
Bristow, Q. 1978. Application of airborne gamma-ray spectrometry in the search for radioactive debris from the russian satellite cosmos 954 (operation "morning light"). Geol Surv Can Pap Issue 78 -1B, 1978, Pages 151-162

My PhD research contribution to the nuclear techniques applied for environmental radioactivity monitoring

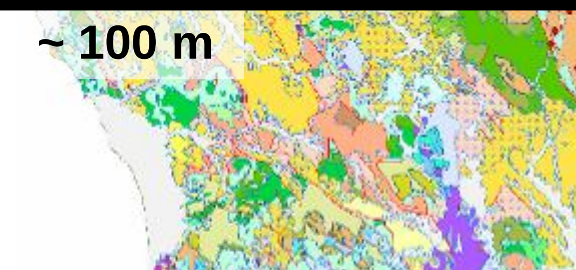
…in lab



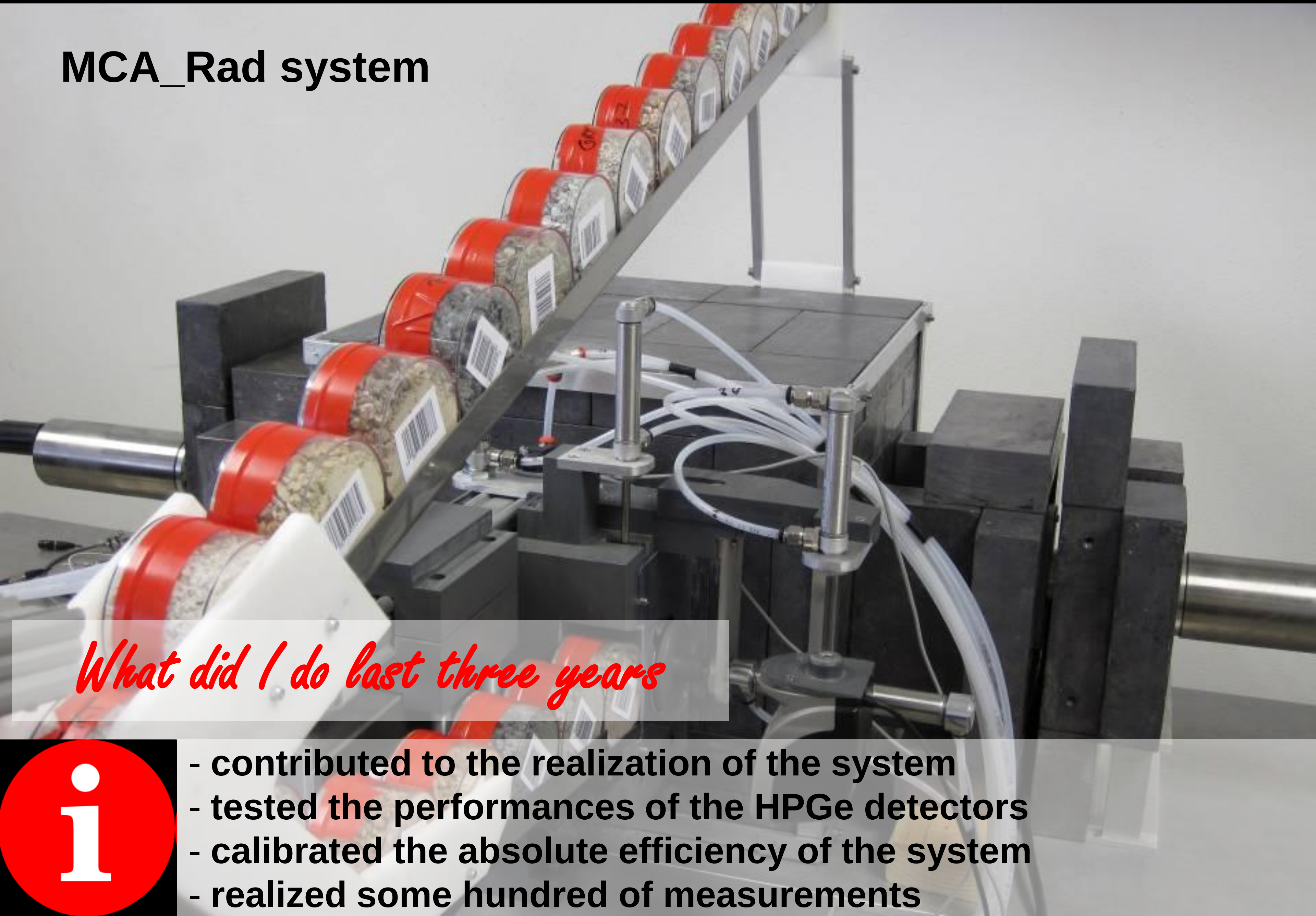
…in situ



…in airborne



A new instrument at Physics Department (316C)



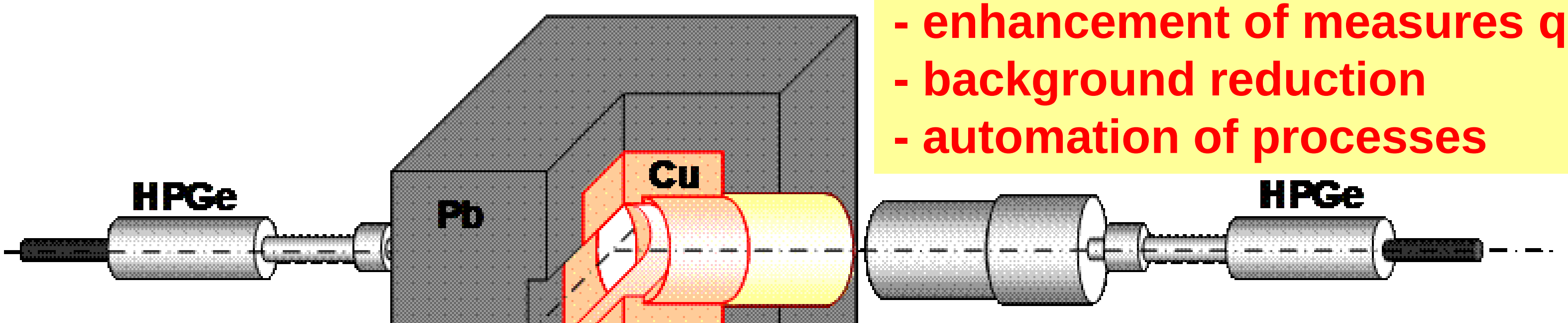
MCA_Rad system

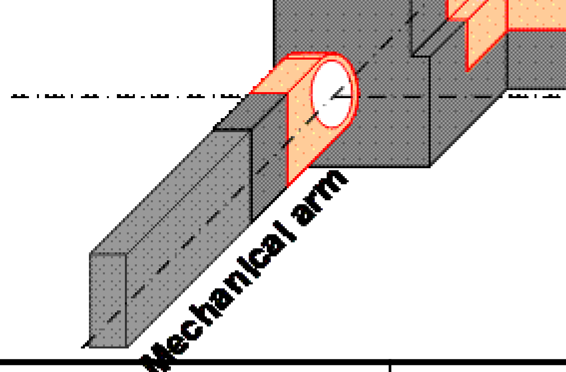
What did I do last three years

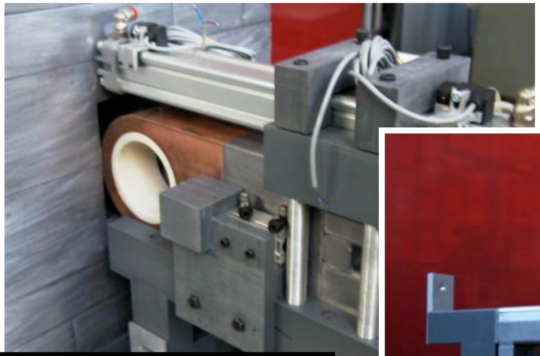
- contributed to the realization of the system
- tested the performances of the HPGe detectors
- calibrated the absolute efficiency of the system
- realized some hundred of measurements

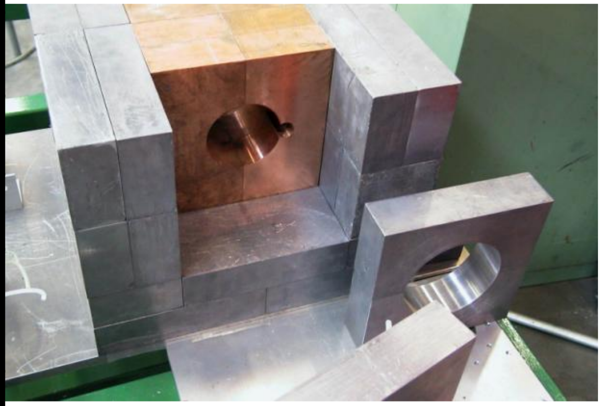
Design and features of MCA_Rad system

Improvements:
- enhancement of measures quality
- background reduction
- automation of processes







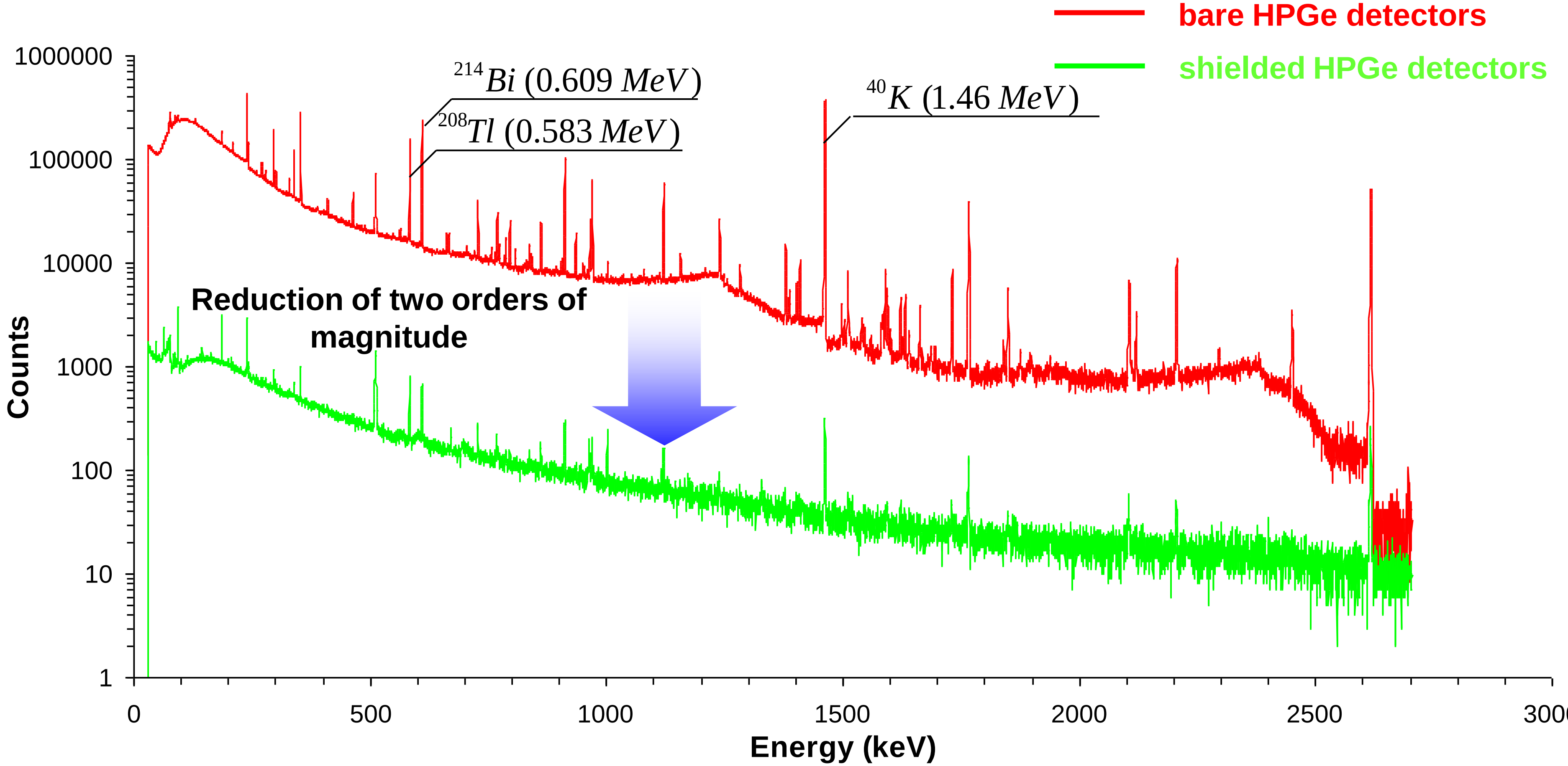


HPGe detectors	Coaxial n-type, 60% of rel. eff.
Energetic resolution	1.9% at 1.33 MeV (^{60}Co)
Cooling technology	Electromechanical (~ -190°C)
Shielding composition	10 cm Pb and 5 cm of Cu
Standard acquisition time	1 hour (180 cc sample volume)
Automatic sample manage	24 samples

Background reduction of MCA_Rad system

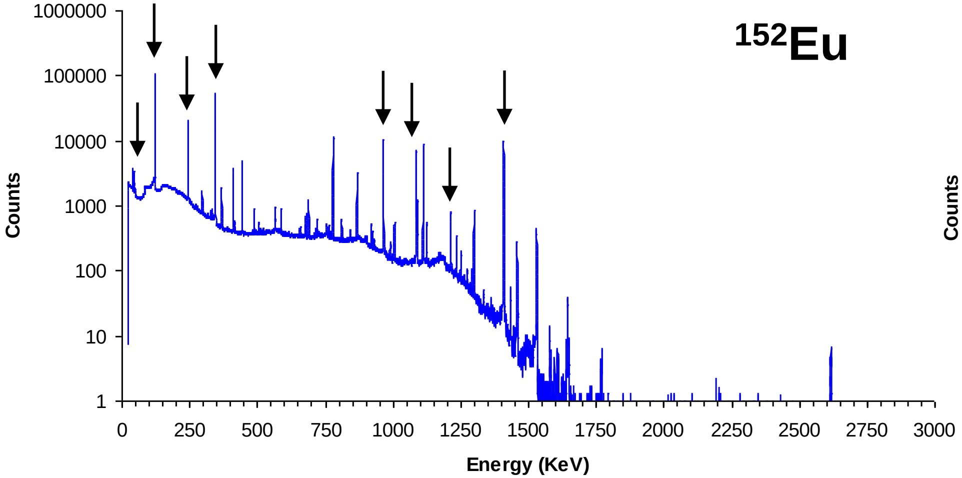
Estimation of Minimum Detectable Activity (MDA) for blank test [Curie 1986].

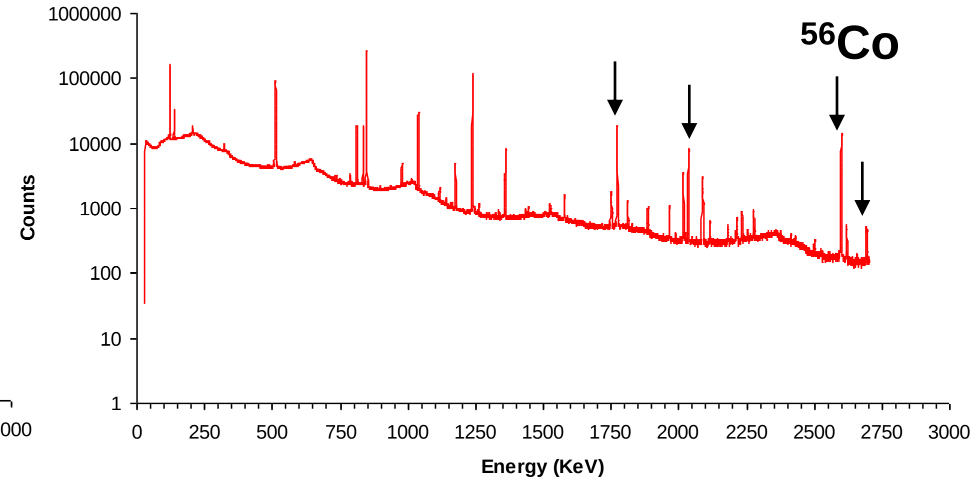
Isotope	E (keV)	MDA (Bq)
^{40}K	1460	0,26
^{214}Bi	609	0,04
^{208}Tl	583	0,06

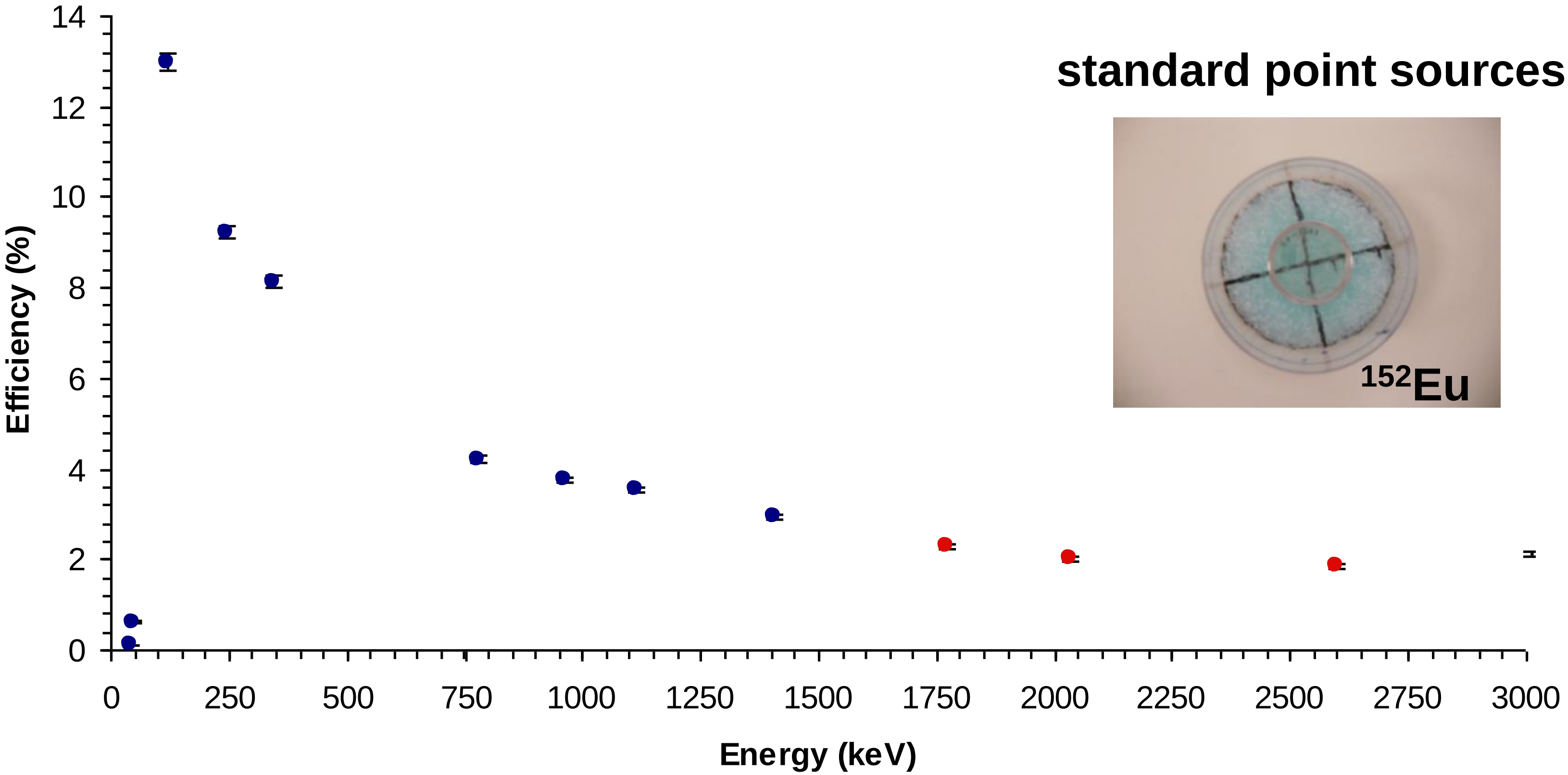


Currie L. A., 1986. Limits for Qualitative Detection and Quantitative Determination Application to Radiochemistry. Analytical Chemistry 40. 586–593.

Efficiency measurements of MCA_Rad system







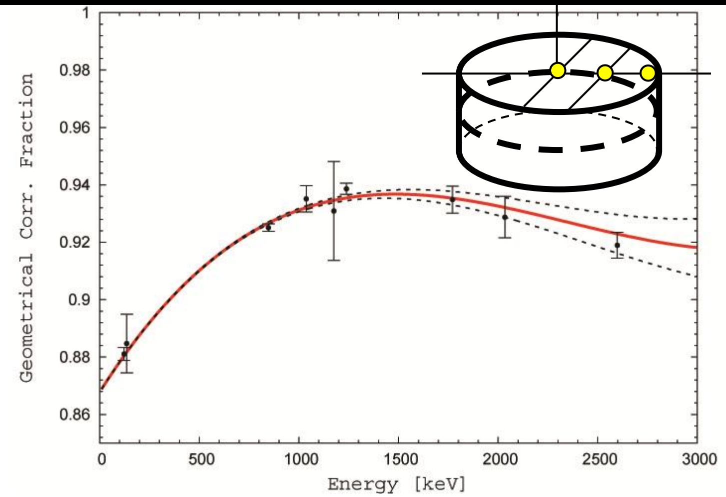
standard point sources

Efficiency analysis: three main corrections

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

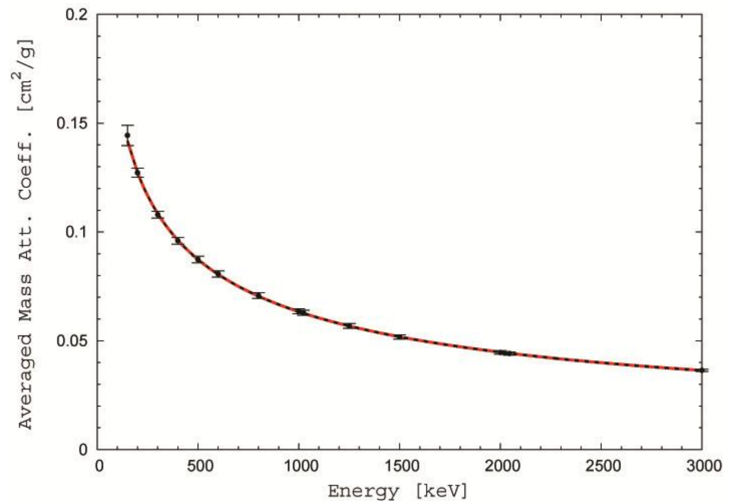
$$C_G = \sum_{i=0}^{3} a_i (E_i / E_0)^i$$

where E_0 = 1keV.



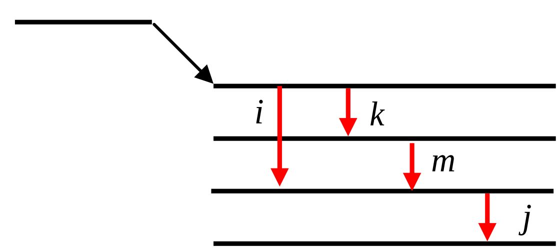
2- Self absorption correction (C_{SA}): averaging the mass attenuation coeff. μ for a "standard rock" with density ρ, I calculated the C_{SA} for the sample thickness t = 4.5 cm using the simplified approach:

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

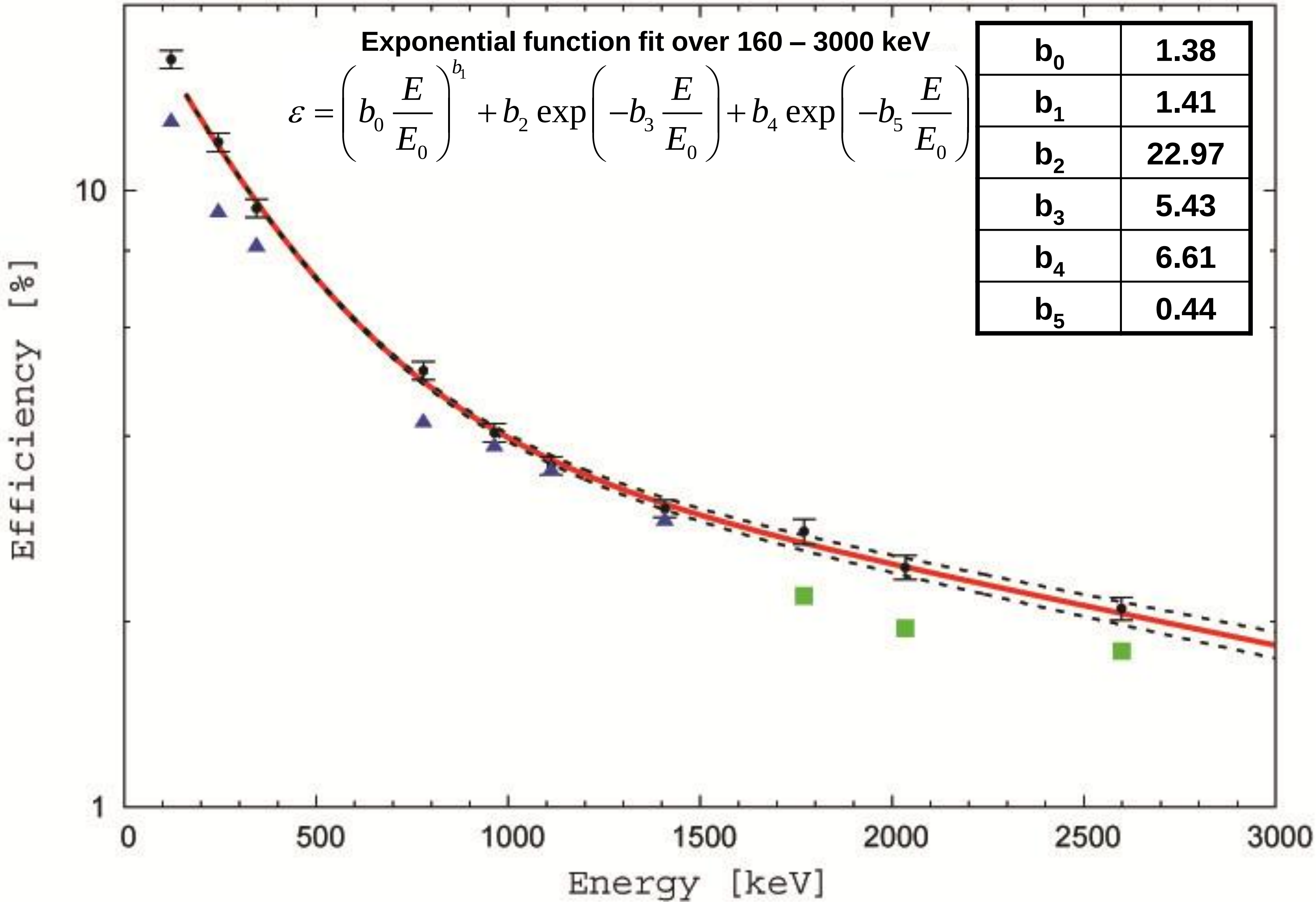


3- Coincidence summing correction (C_{CS}): the correction of (i) events takes into account the summing in (k,m) and summing out (j) effects:

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



Absolute full-peak energy efficiency for MCA_Rad system



Exponential function fit over 160 – 3000 keV

$$\varepsilon = \left(b_0 \frac{E}{E_0} \right)^{b_1} + b_2 \exp\left(-b_3 \frac{E}{E_0} \right) + b_4 \exp\left(-b_5 \frac{E}{E_0} \right)$$

b_0	1.38
b_1	1.41
b_2	22.97
b_3	5.43
b_4	6.61
b_5	0.44

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

Validation test using certified IAEA ref. materials

^{228}Ac
^{212}Pb
^{212}Bi
^{208}Tl

^{234m}Pa
^{214}Bi
^{214}Pb

	Data certified by IAEA		MCA_Rad system results	
Ref. material	A (Bq/kg)	1σ	A (Bq/kg)	1σ (dev.st)
IAEA_RGK_1	14000	200 (1.4 %)	14274	241 (1.7%)
IAEA_RGU_1	4940	20 (0.3 %)	4881	101 (2.0%)
IAEA_RGTh_1	3250	45 (1.4 %)	3205	113 (3.5 %)

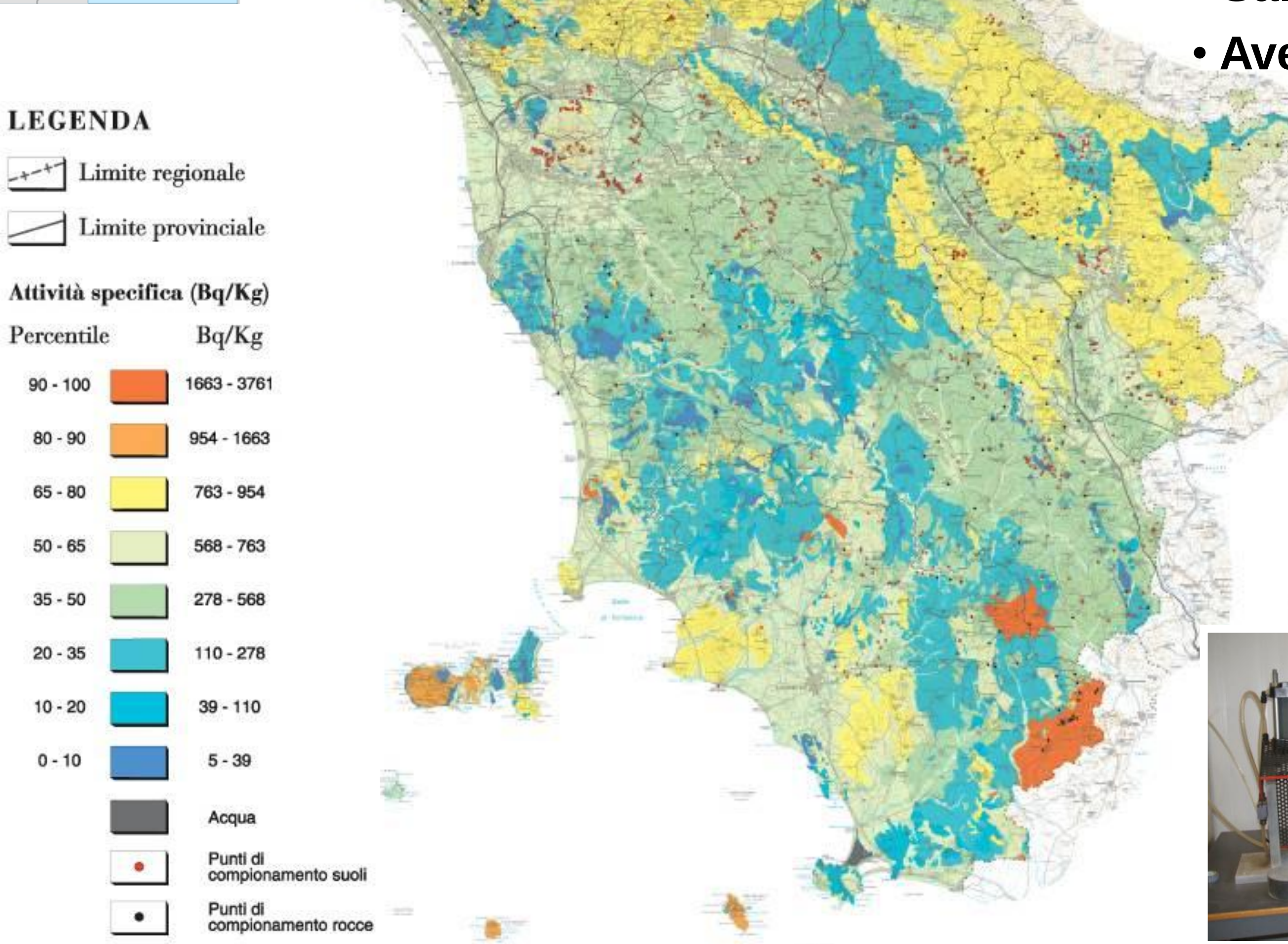
The average compositions of the terrestrial crust are: 720 Bq/kg (^{40}K); 30 Bq/kg (^{238}U) and 40 Bq/kg (^{232}Th).

Spectrum acquisition live time of 1h			
Stat. unc.	^{40}K (Bq/kg)	^{238}U (Bq/kg)	^{232}Th (Bq/kg)
10%	60	20	30
5%	220	50	70
1%	4400	440	450

Map of radioactivity content of Tuscany territory

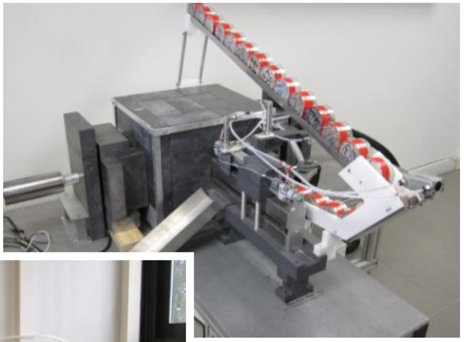
Tuscany Region, Italy

1:250.000



Sampling summary:

- **Sample total: 1913**
- **Rock samples: 677**
- **Soil samples: 1236**
- **Sampling days: 92**
- **Average distribution ~12 km^2**



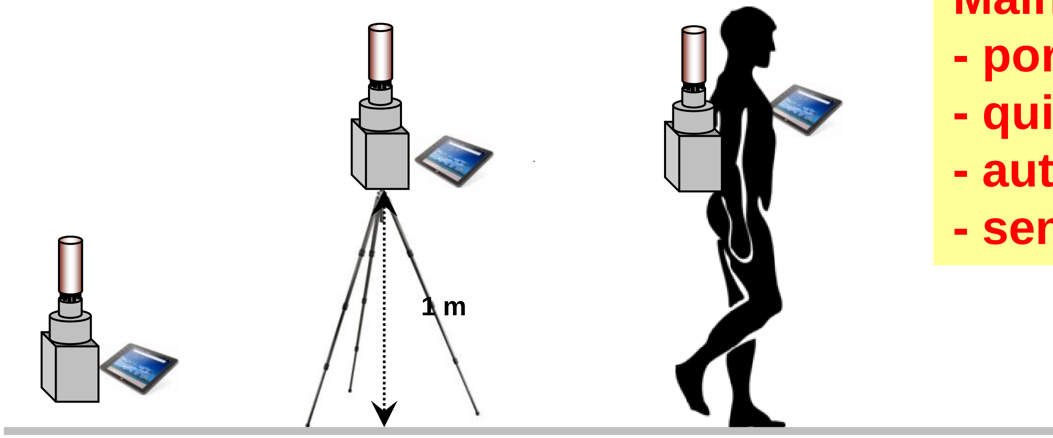
New portable scintillation γ-ray spectrometer

ZaNaI_1.0L system

What did I do last three years

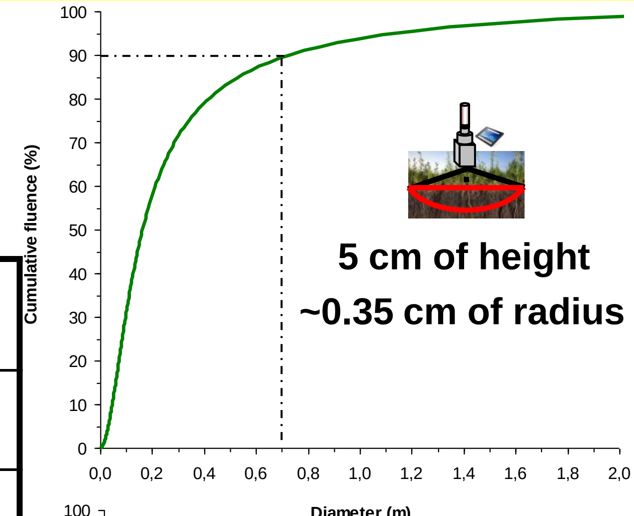
- contributed to the development of a new approach on spectrum analysis using FSA with NNLS constrain
- realized the sensitivity calibration of the system
- realized some tens of measurements

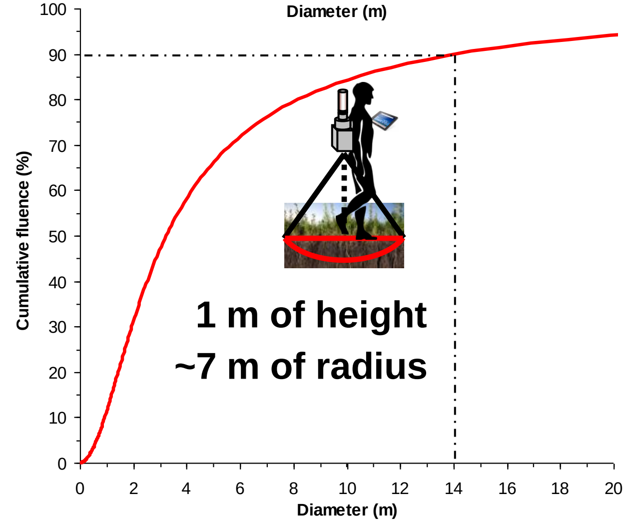
ZaNaI design and features



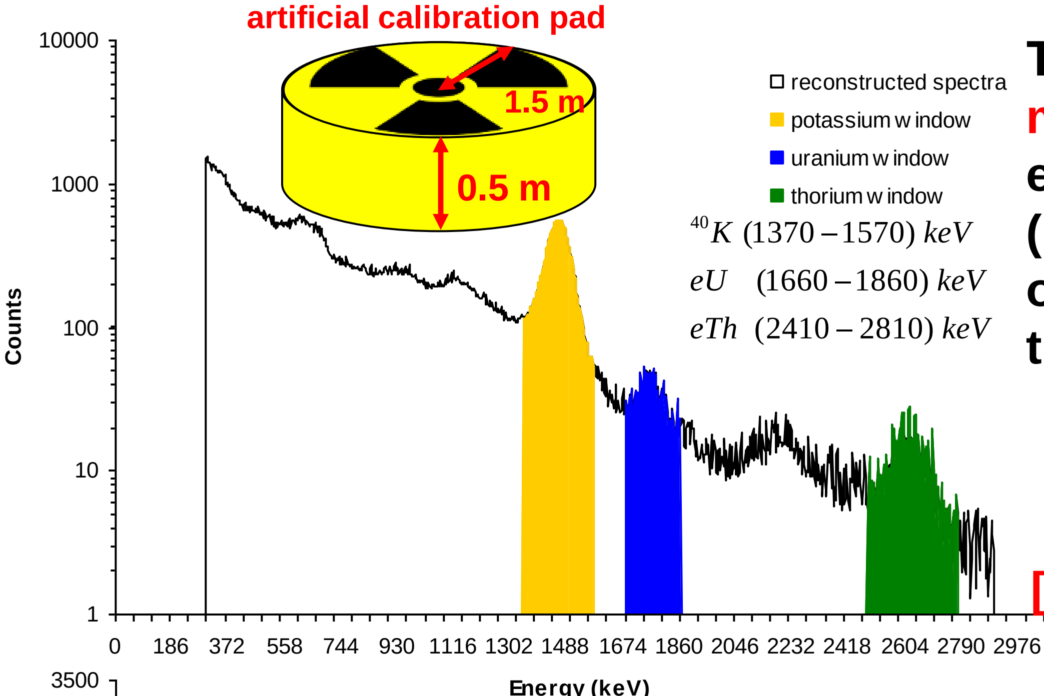
Main characteristics:
- portable
- quick feedback
- autonomy
- sensibility calibration

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 min (static mode) 10 – 30 sec (dynamic mode)**
Auxiliary sensors	**Pressure & Temperature**





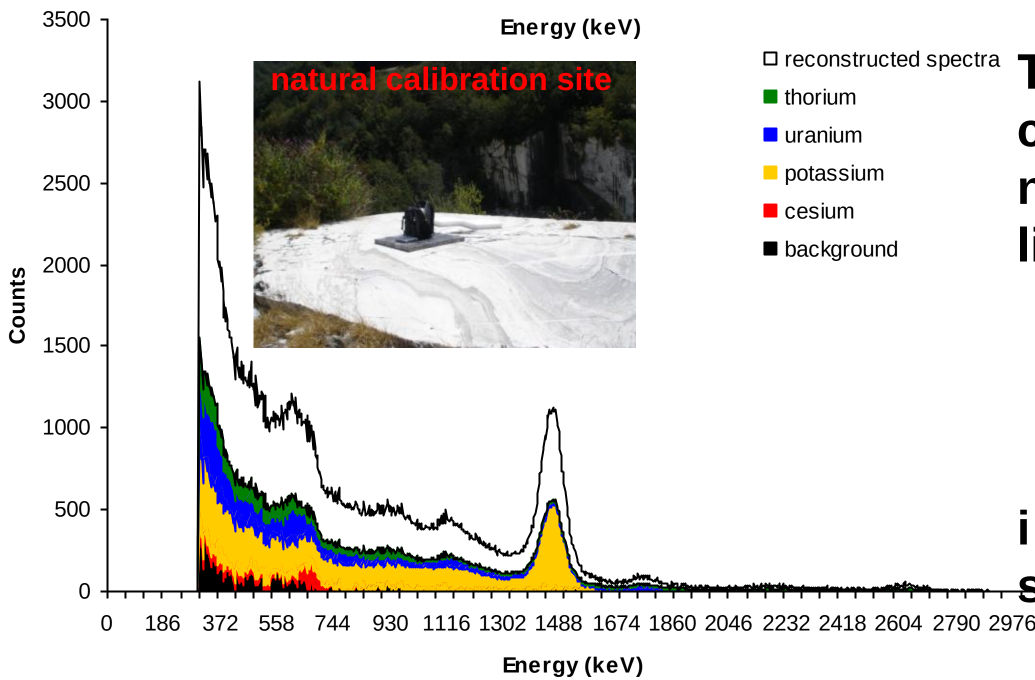
Portable γ–ray spectrometer: calibration methods



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.

Using natural sites as calibration pads

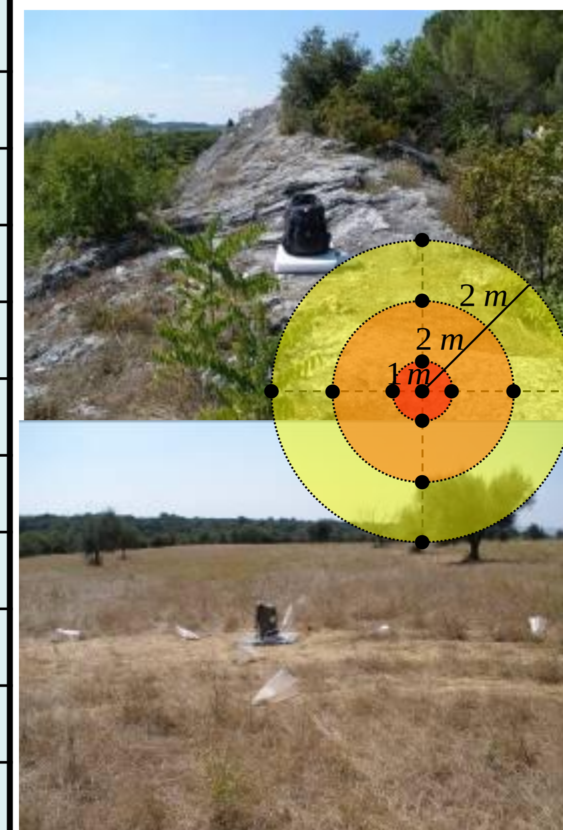
We have identified and characterized up to now 11 natural which are used as calibration sites.

Site criteria:

- undisturbed and flat area
- enriched prevalently in one natural radioelement
- relatively homogeneous radioelement distributions

135 samples

Nr.	Site ID	Samples	Feature
1	Sorano T1	12	all high
2	Sorano P1	18	eTh 44.2 ppm
3	Pratomagno 1	10	^{40}K 2.8%
4	Campo Cecina	31	~ blank + ^{137}Cs
5	Spiaggia Ronchi	5	all low
6	Gobbie Cava	6	~ blank
7	Rapolano Terme	7	eU 6.8 ppm
8	Sorano P2	10	eTh 39.2 ppm
9	Pratomagno 2	10	^{40}K 2.9%
10	San Michele	13	all low
11	Gavorrano	13	^{40}K



FSA with Non-negative least square constrain

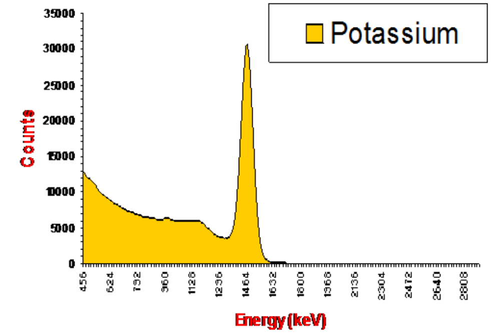
We reconstruct the measured spectra as a linear sum of standard sensitivity spectra referred to ^{40}K, ^{238}U, ^{232}Th and ^{137}Cs.

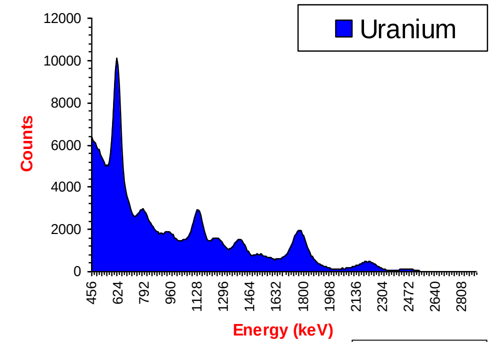
$$N_{(i)} = S_{K(i)}C_K + S_{U(i)}C_U + S_{Th(i)}C_{Th} + S_{Cs(i)}C_{Cs} + Bckg_{(i)}$$

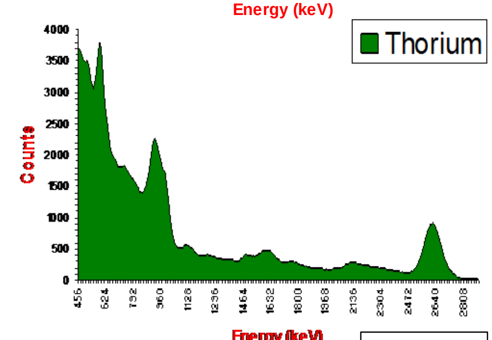
$N_{(i)}$ – measured spectra,
$S_{(j)}$ – standard sensitivity spectra,
C_K, C_U, C_{Th}, C_{Cs} – activity concentrations,
$Bckg_{(i)}$ – background spectra,
$1 \leq i \leq 867$ (300 keV $\leq E \leq 2900$ keV),
j referred to ^{40}K, ^{238}U, ^{232}Th, ^{137}Cs and Background.

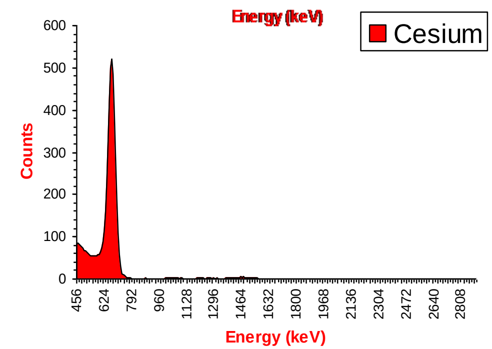
The optimal activity concentration is estimated minimizing | [C][S] – [N] | using the constrain [S] ≥ 0 (non-negative least square) according to the formula:

$$\chi^2_\nu = \frac{1}{n-m}\sum_{i=1}^{N}\frac{\left(\sum_j [C]_j [S]^i_j - [N]^i\right)^2}{\sigma^2_{[N]^i}}$$



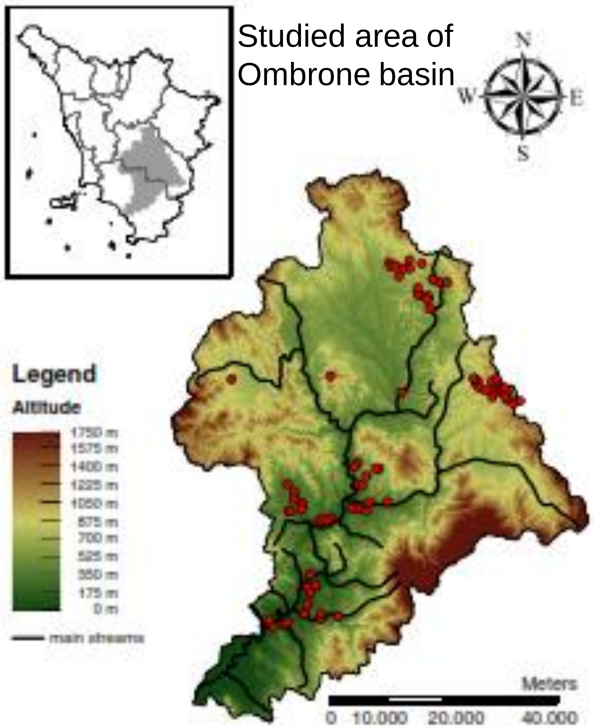


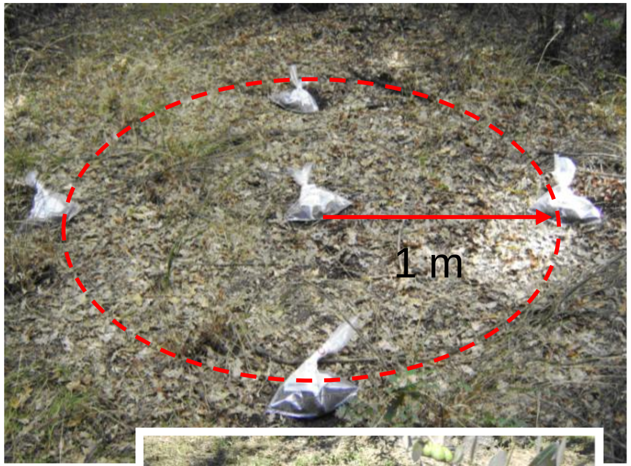


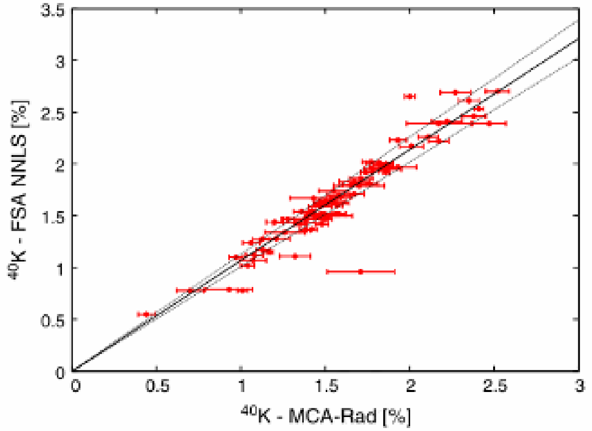


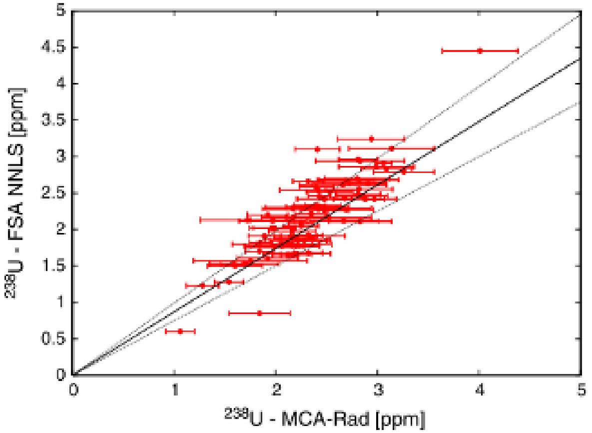
Validation test of the method FSA with NNLS constrain

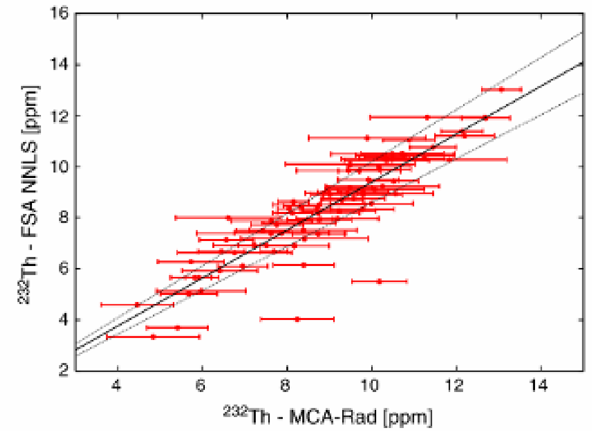
For 80 different sites we measured 5 samples of soil in laboratory using the MCA_Rad system and compare the results with the data obtained in-situ by ZaNal.









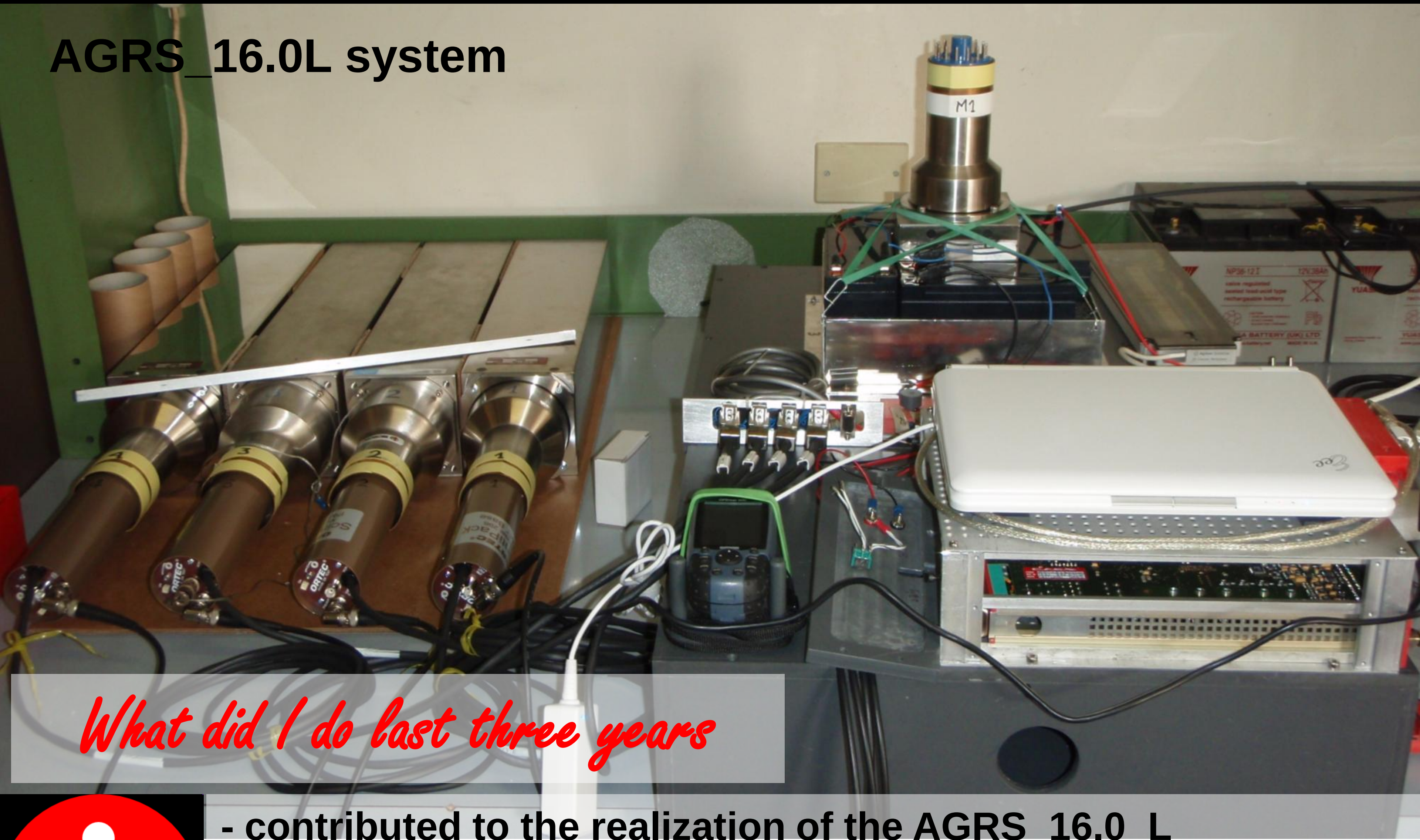


Regression coefficients between two methods (in lab vs. in situ) using different spectra analysis

	K	U	Th	X^2
WAM	1.12 ± 0.07	1.11 ± 0.10	1.00 ± 0.09	-
FSA	0.99 ± 0.06	0.78 ± 0.14	0.86 ± 0.07	1.22 ± 0.08
FSA-NNLS	1.06 ± 0.06	0.87 ± 0.12	0.94 ± 0.07	1.06 ± 0.05

An airborne γ-ray spectrometer

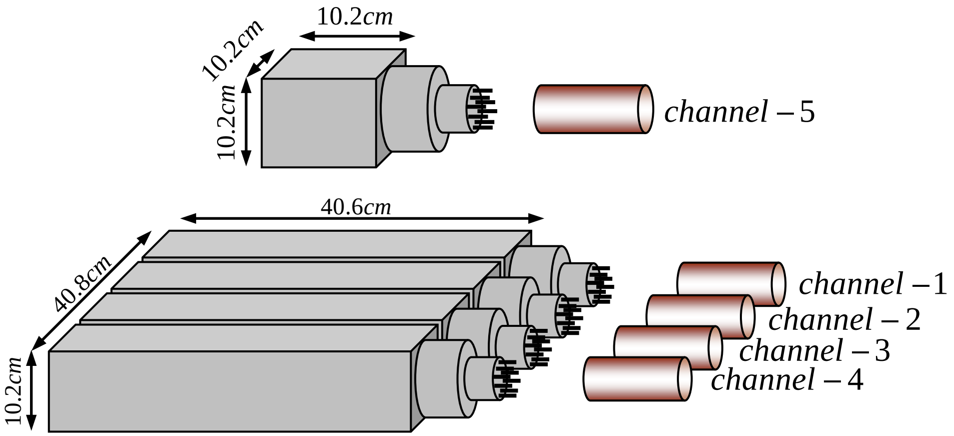
AGRS_16.0L system



What did I do last three years

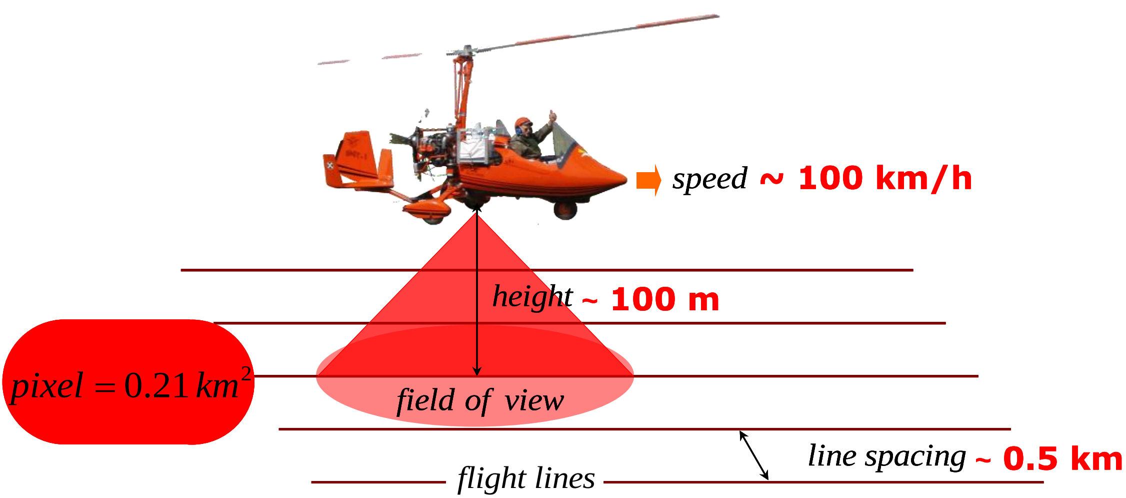
- contributed to the realization of the AGRS_16.0_L
- realized the sensitivity calibration of the system implementing the FSA using NNLS constrain
- realized some tens of hours of airborne measurements

AGRS design and features



4 NaI(Tl) detector	**4 Lit. (102 x 102 x 406 mm)**
1 NaI(Tl) detector	**1 Lit. (102 x 102 x 102 mm)**
Energetic resolution	**8.5% at 662 keV (^{137}Cs)**
Channels	**1024 (512, 256)**
Real-time feedback	**notebook (smartphone & tablet)**
Power autonomy	**3 hours**
Weight (total)	**~ 115 kg**
Output	**List mode events (individual & composite spectra)**
Spectrum analysis	**FSA with NNLS constrain (stripping ratio method)**
Auxiliary sensors	**Pressure & Temperature**

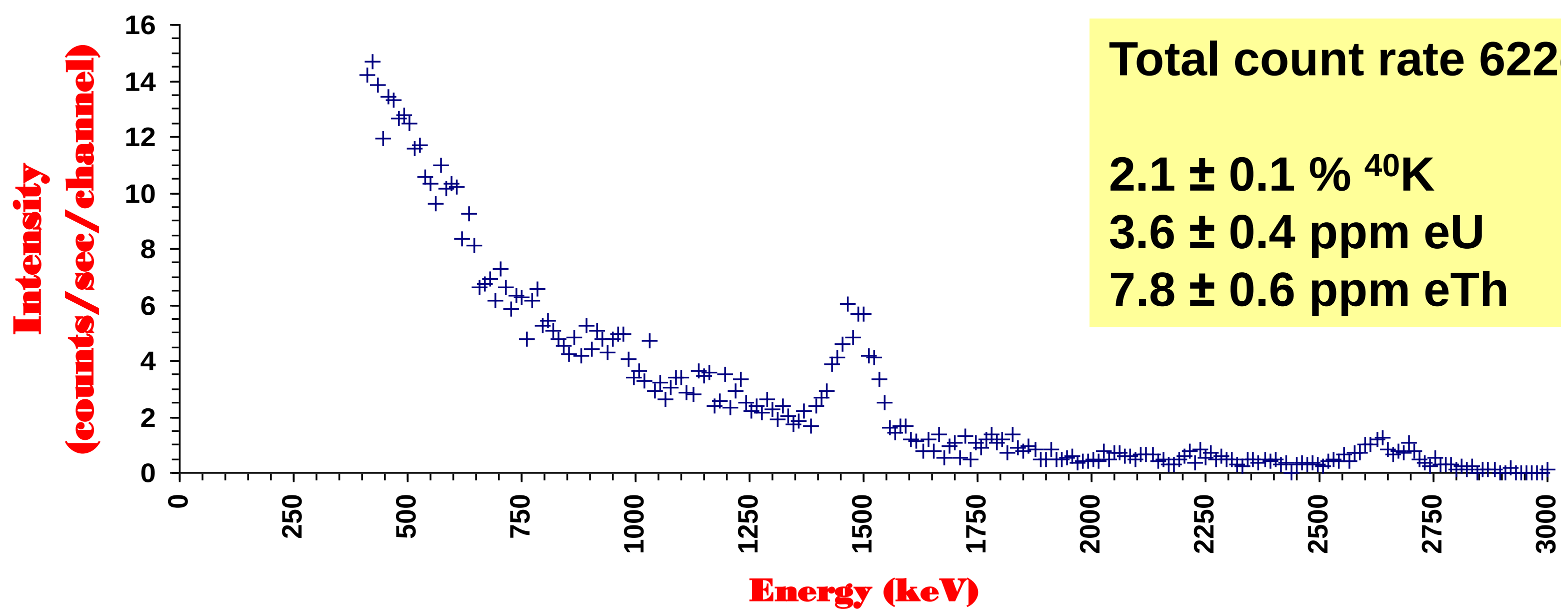
A typical AGRS_16.0L measurement*



main corrections

- Flying height
- Topography
- Atmospheric radon gas

A typical 1s spectrum acquisition with AGRS_16.0L at 100 m of height.
The spectra is recorded in 256 channels in the energy reange 0-3 MeV.

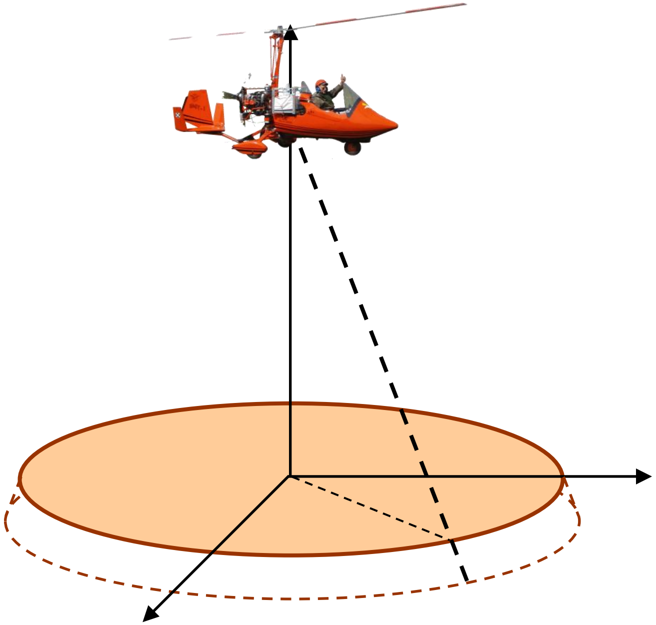


Total count rate 622cps

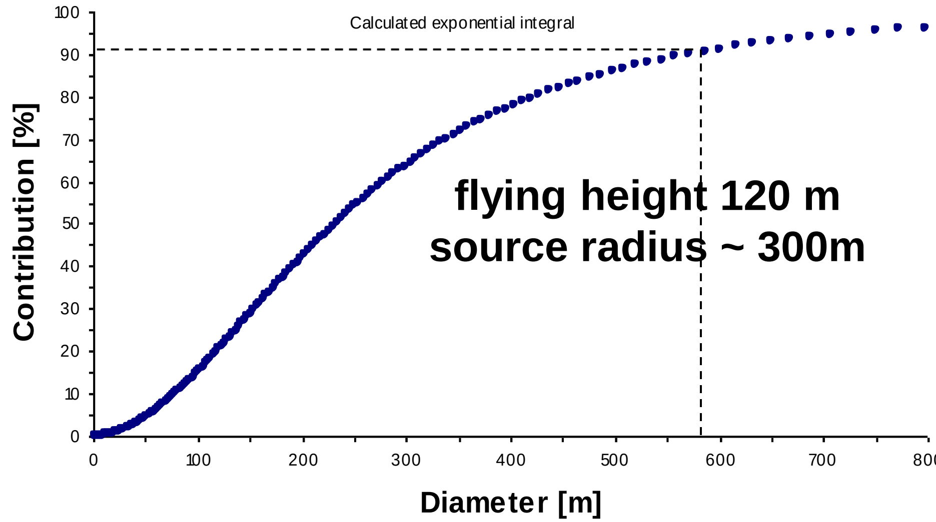
2.1 ± 0.1 % ^{40}K
3.6 ± 0.4 ppm eU
7.8 ± 0.6 ppm eTh

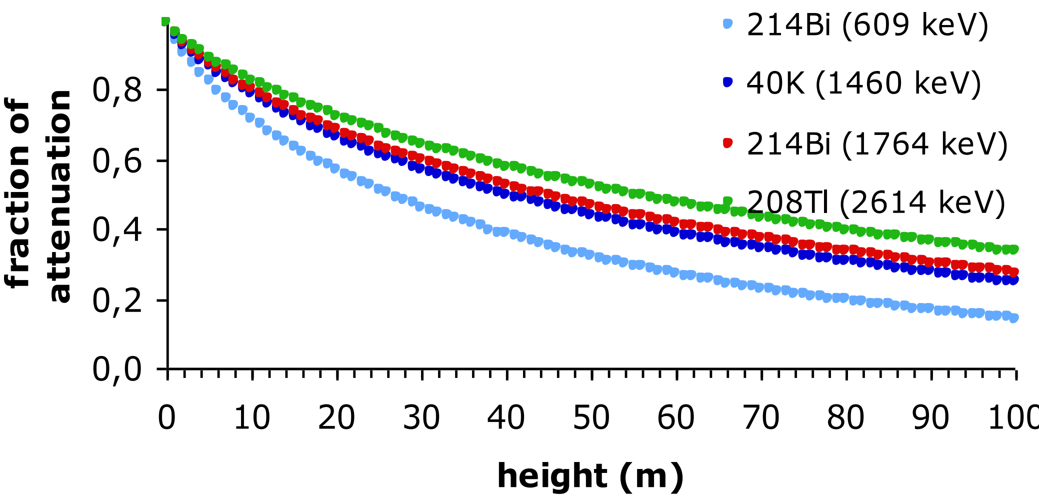
* International Atomic Energy Agency. Guidelines for radioelement mapping using gamma-ray spectrometry data. IAEA-TECDOC-1363, Vienna; 2003.

Flying height correction



Monoenergetic unscattered photons detected above a uniformly radioactive infinite source per unit time





	Energy (keV)	Linear attenuation coeff. in air (m^{-1})
^{40}K	1460	0.0104
^{214}Bi	609	0.0068
	1765	0.00679
^{208}Tl	2614	0.00506

$$C_{height} = 0.25h^{1/3} + 0.75e^{-(2h/\mu)}$$

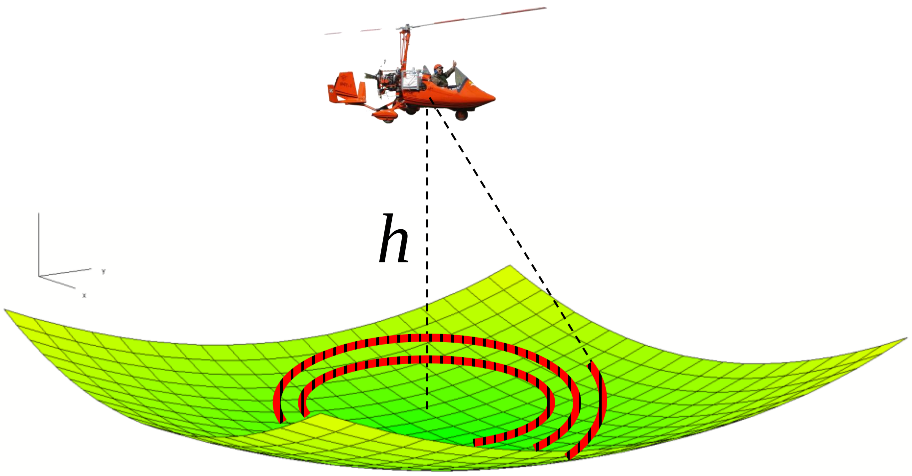
Topographic correction

Using the DEM (digital elevation model) with 10 x 10 m spatial resolution we calculate:

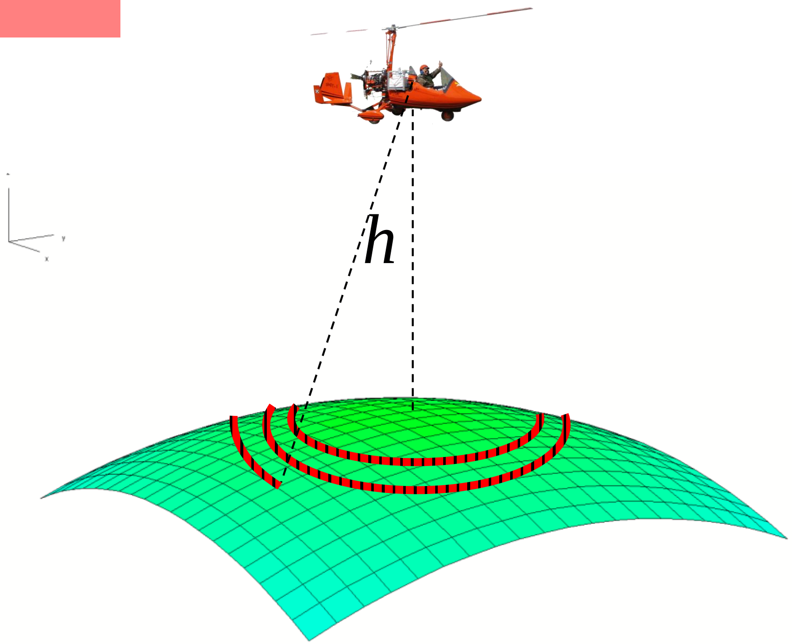
N_h^{DEM} – the flux coming from the "real" topography,

N_h^{PLANE} – the flux coming from the "ideal flat" topography (for h = 100m).

$$C_{topography} = \frac{N_h^{DEM}}{N_h^{PLANE}}$$



$$C_{topography} > 1$$



$$C_{topography} < 1$$

Atmospheric radon correction

The “upward looking detector” method consist on finding the relationship between measured count rates in the ^{222}Rn monitor $U_{monitor}$ window to those in the system U_{system} window for radiation due to U in the ground.

$$eU_{system}(ppm) = \Gamma C^{corr}_{monitor}(cps)$$

dove:

ground contribution in upward looking detector

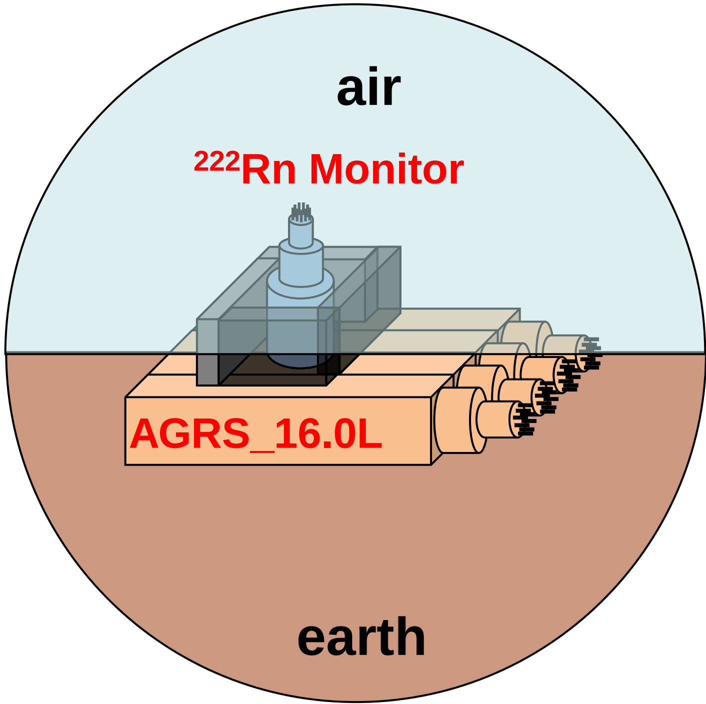
$$C^{corr}_{monitor}(cps) = C_{monitor}(cps) - \alpha C^{system}_{Cs} - \beta C^{system}_{K} - \gamma C^{system}_{U} - \delta C^{system}_{Th}$$

These coefficients can be determined by flying a large body of water

$C^{system}_{Rn} = \Gamma C_{monitor}(cps)$	$\Gamma = 44.98$

and flying over a calibrated line

$C_{monitor}(cps) = \alpha C^{system}_{Cs}$	$\alpha = 0.15$
$C_{monitor}(cps) = \beta C^{system}_{K}$	$\beta = 11.47$
$C_{monitor}(cps) = \gamma C^{system}_{U}$	$\gamma = 3.67$
$C_{monitor}(cps) = \delta C^{system}_{Th}$	$\delta = 1.17$



Summary of airborne surveys

Region	Tuscany
Area	23 10^3 km^2
Period	April-June '10
Eff. flights	33
Total hours	~ 100
Survey area	~ 20%
Data amount	~ 30GB

Elba island survey:

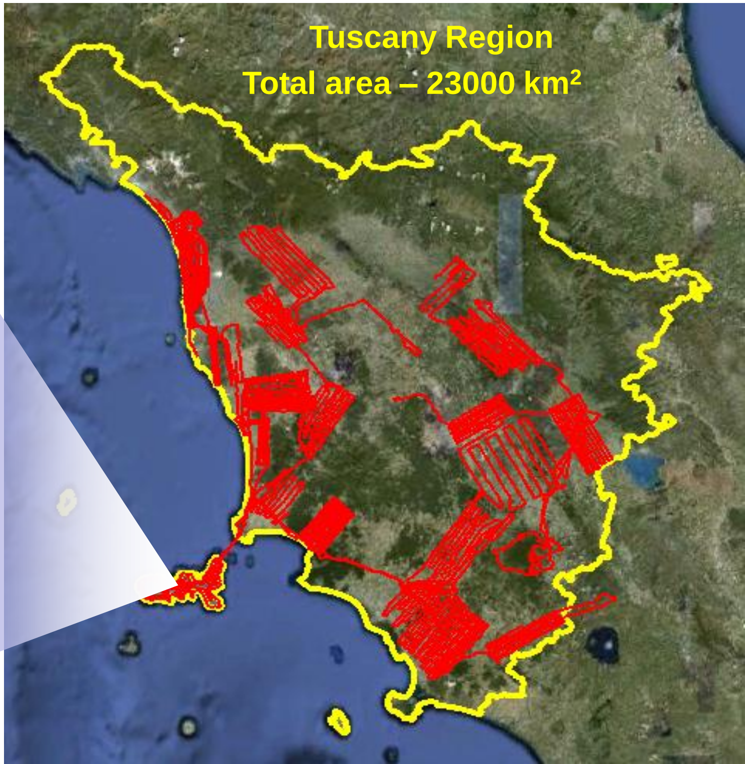
Flight realized at June, 3 2010

Flight duration: ~2.2 h

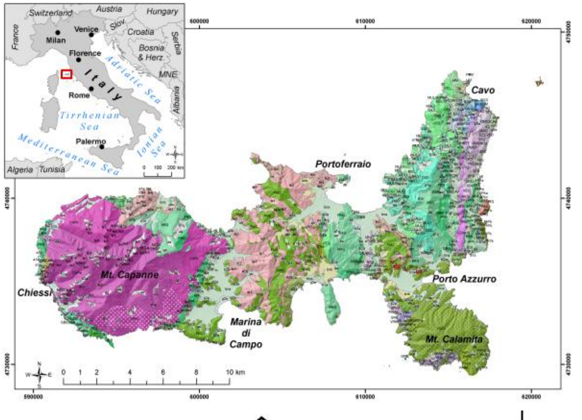
Surveyed area: ~225 km^2

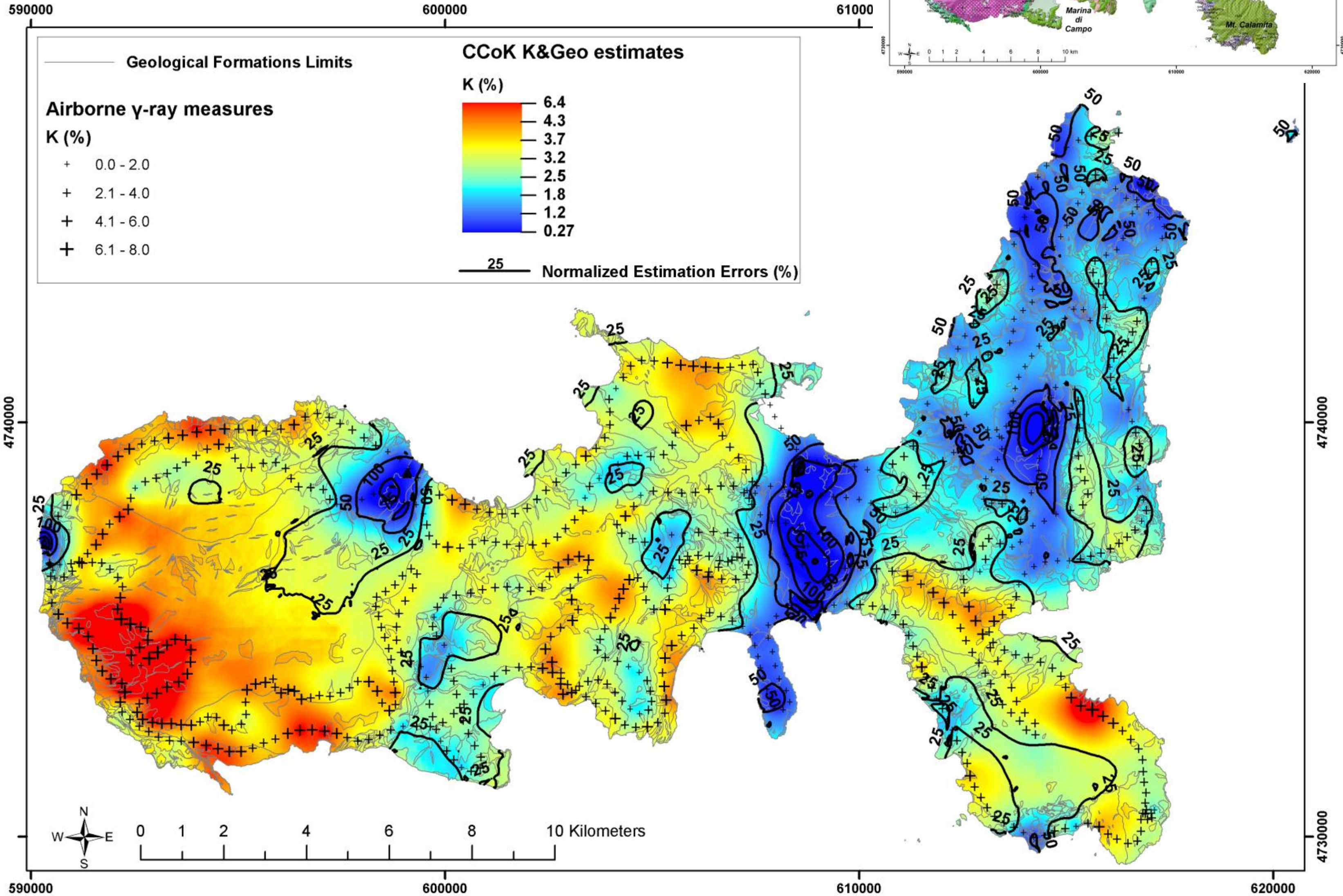
Weather conditions: cloudy

Data acquisition: ~ 800 data (10 sec)

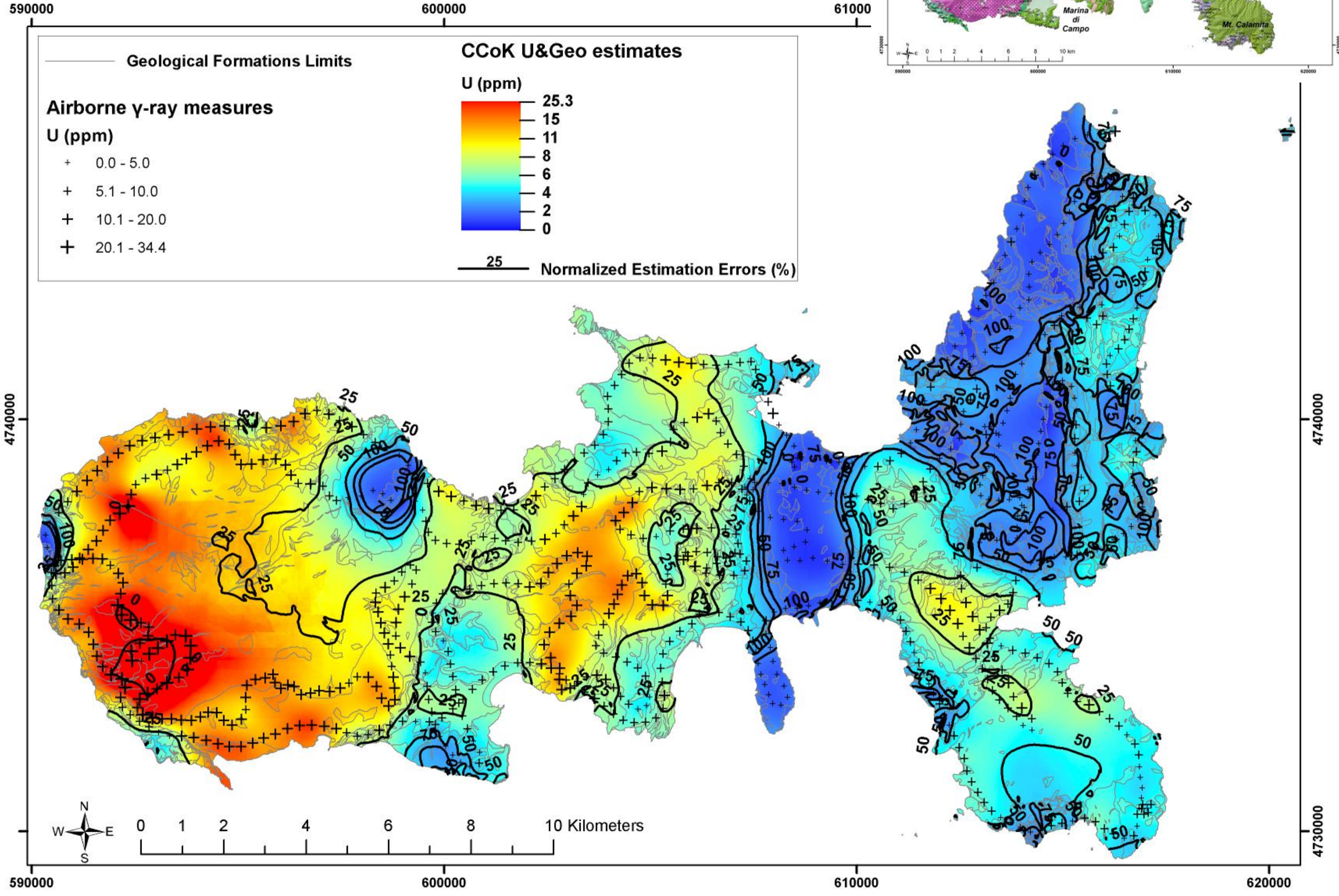


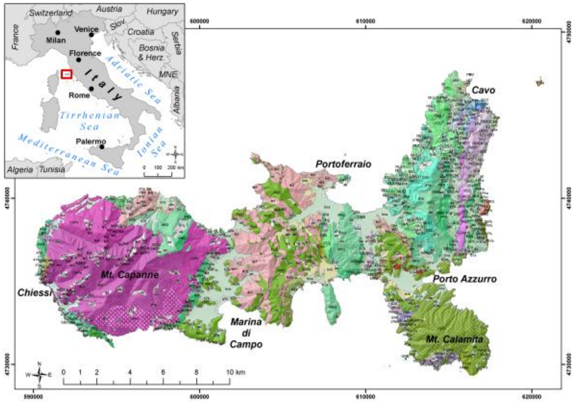
Potassium (%) map



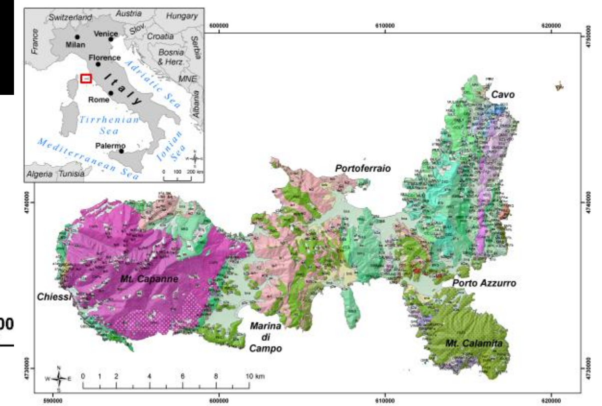


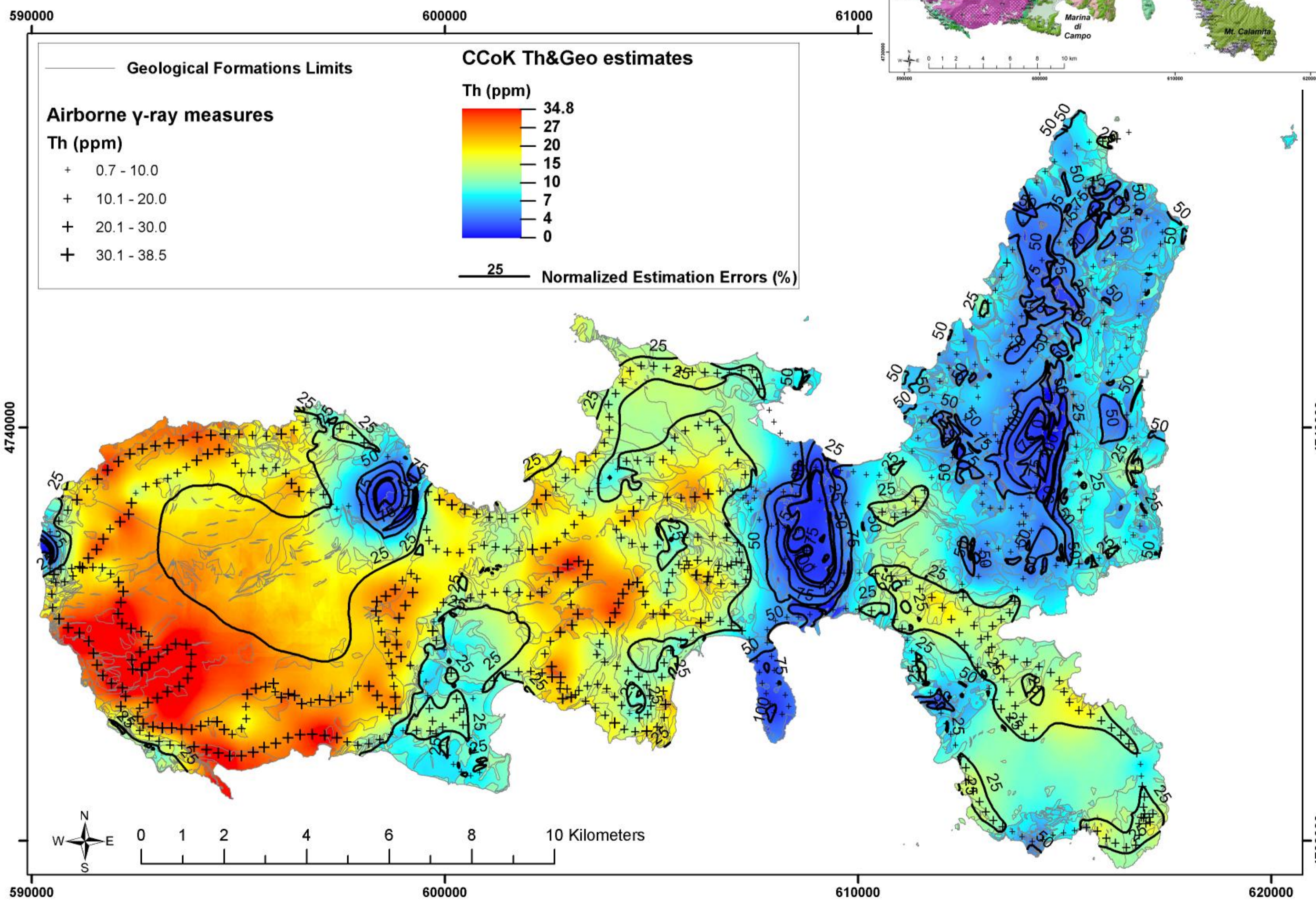
Uranium (ppm) map





Thorium (ppm) map





Conclusions and prospective

- Realization of a fully automated γ-ray spectrometer (MCA_Rad) and its efficiency characterization with an overall uncertainty of less than 5%.

- Construction of the map of radioactivity content of Tuscany territory using over 1900 data.

- Realization of a portable scintillation γ-ray spectrometer (ZaNaI) and development of an alternative approach on calibration and spectrum analysis procedure using natural sites and FSA-NNLS method.

- Realization of extensive measurements (80 sites) investigated both in-situ using ZaNaI (FSA-NNLS method) and in laboratory using MCA_Rad showed excellent correlation between them.

- Realization of an airborne γ-ray spectrometer (AGRS) and successfully implementation of the FSA-NNLS method for spectra analysis.

- Realization of the first AGRS survey over Tuscany region territory and realized some preliminary maps for radioelement distribution in Elba island.

Realization of the radioactivity content map of Veneto territory.
Industrialization of AGRS γ-ray spectrometer.
Investigation of radioactivity content in building materials

Peer-reviewed scientific papers

1. Guastaldi E. et al. (2012). A new geostatistical approach for interpolating airborne γ-ray survey based on geological constrains. Geoderma. (Submitted)

2. Xhixha G. et al. (2012). Fully automated gamma-ray spectrometer for NORM characterization. Journal of Environmental Radioactivity. (submitted)

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

Conference proceedings and papers not peer-reviewed

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. Puccini A. et al. (2011). Measurements of natural radioactivity with a portable gamma-ray spectrometer in Sardinian granite dimension stones. 6th International Conference of Applied Geophysics for Environmental and Territorial System Engineering.

5. Puccini A. et al. (2010). Employment of portable gamma-ray spectrometer in survey and mapping of intrusive complexes: a case study from the Buddusò pluton (Sardinia). Atti 85° Congr. Soc. Geol. It., vol. 11, 297-298.

6. 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.

7. Puccini A. et al. (2010). Natural radioactivity in Sardinian granite dimension stones. Atti 85° Congr. Soc. Geol. It., vol. 11, 552-553.

8. Cfarku F. et al. (2009). Përcaktimi i radioaktivitetit alfa - beta total në ujë me metodën e npg (numërues propocional gazor). Buletini i shkencave natyrore Nr. 7, 83 – 88.