Environmental (& *"other"*) Applications *(of Nuclear Techniques)*

Lecture II

In-situ measurements

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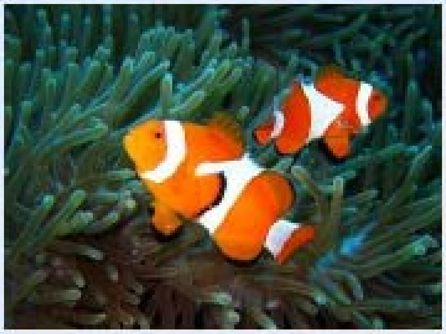
NAPC-PH/NSIL

uk_npss Bristol 2013

IAEA
International Atomic Energy Agency

Environment

Responding to the impacts of environmental changes

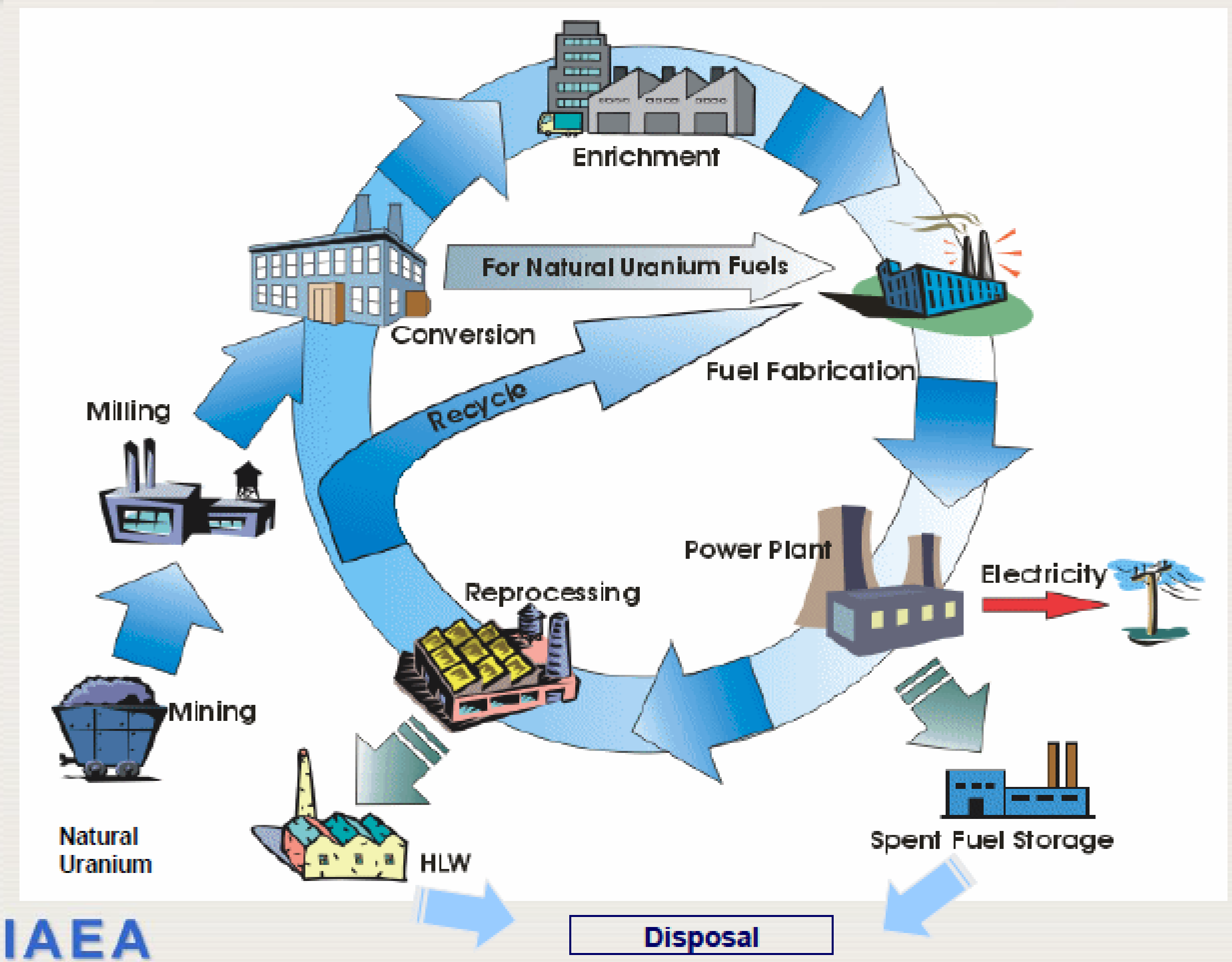


- Establishing approaches, protocols and standards for environmental assessments

- Assessing the impacts of climate change, ocean acidification and other contaminants on seafood, biodiversity and the marine environment

- Providing reliable environmental
- radioactivity concentration data

Nuclear Fuel Cycle



What is NORM?

Courtesy of Paddy Regan Univ Surrey

- **N**aturally **O**ccurring **R**adioactive **M**aterials
- Two main sub-groups:
  - Cosmogenic (from cosmic ray interactions)
    - ^{14}C (from $^{14}N(n,p)^{14}C$), ^{7}Be, ^{26}Al
  - Primordial (i.e. very old)
    - Single nuclei (e.g., ^{40}K)
    - Decay chains (^{232}Th, ^{235}U, $^{238}U/^{226}Ra$)

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Other nuclides in the 'background'

- Man-made ('anthropogenic') radionuclides in the environment.
  - Nuclear weapons tests / Chernobyl / Fukushima
    - Fission fragment daughters such as ^{137}Cs, ^{90}Sr, ^{131}I
    - ^{241}Am
    - ^{239}Pu
    - Neutron capture products (e.g., ^{134}Cs)

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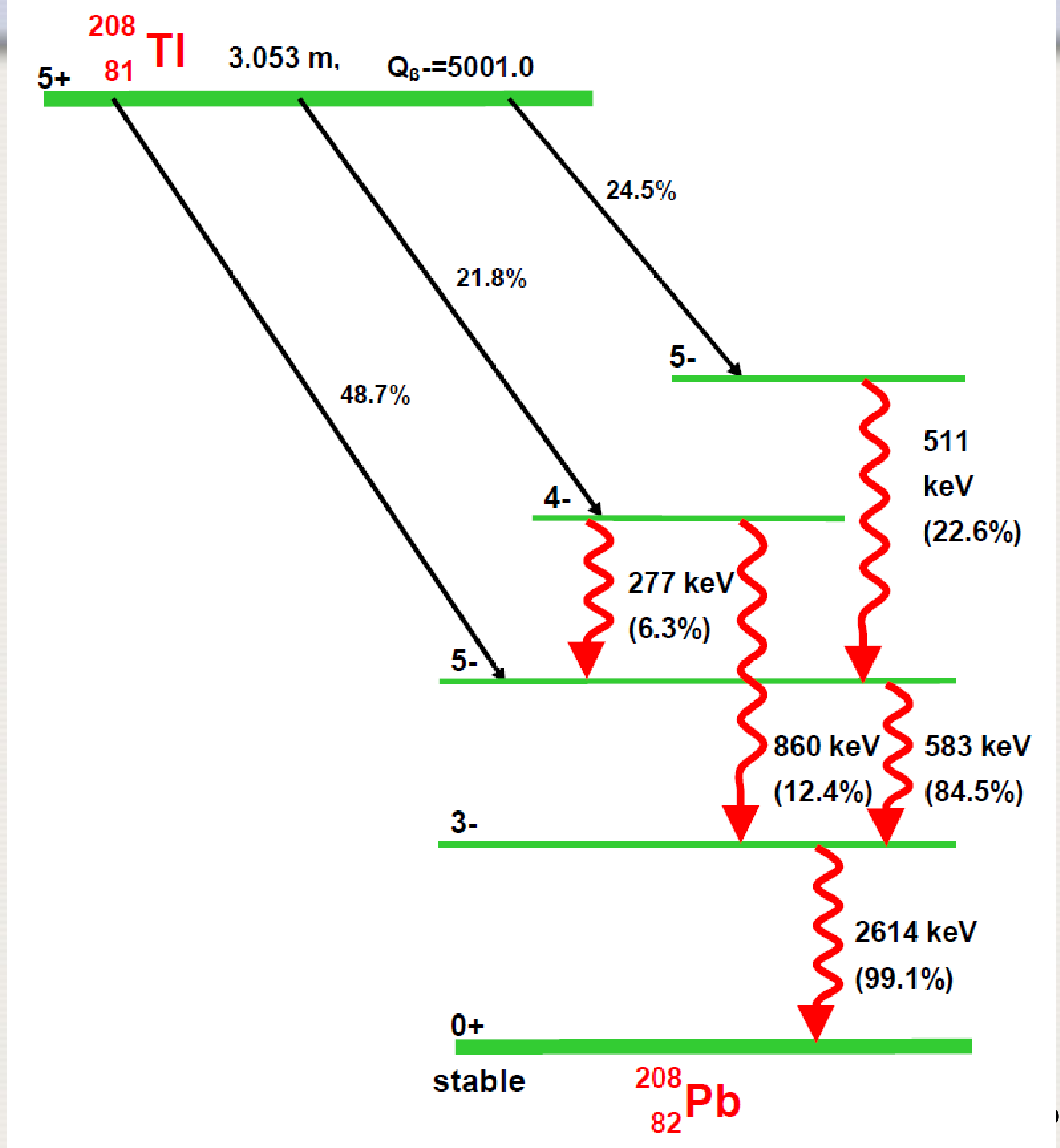
How do you measure the NORM?

- You could use alpha spectrometry

Parent Nucleus → Daughter Nucleus	Decay Half-Life	Energy of main α decays and % of decays with this energy.
$^{210}Po \rightarrow {}^{206}Pb + \alpha$	138.4 days	5.304 MeV (100%)
$^{234}U \rightarrow {}^{230}Th + \alpha$	2.455 x 10^5 years	4.774 MeV (71%) & 4.722 MeV (28%)
$^{235}U \rightarrow {}^{231}Th + \alpha$	7.038x10^8 years	4.399 MeV (55%) & 4.366 MeV (17%)
$^{238}U \rightarrow {}^{234}Th + \alpha$	4.468x10^9 years	4.198 MeV (79%) & 4.151 MeV (21%)

….or gamma-ray spectrometry.



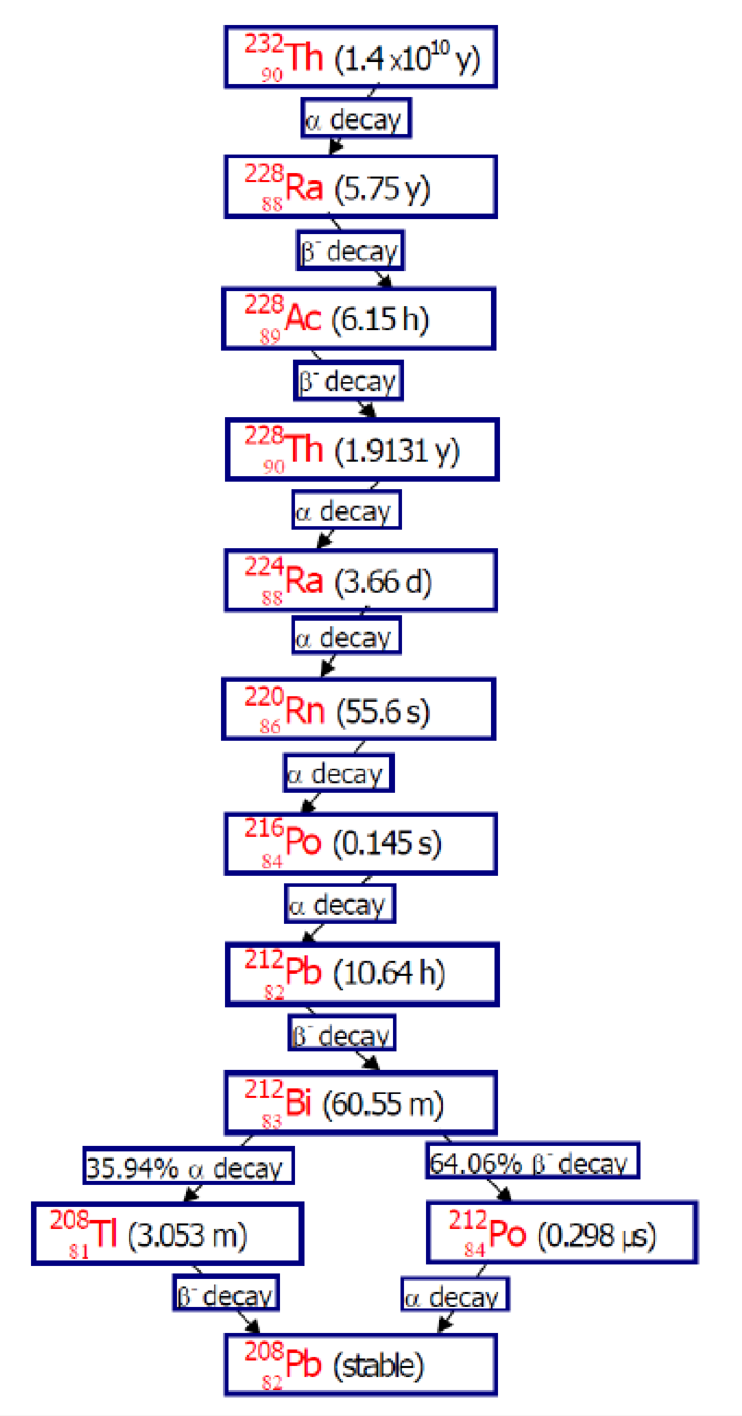


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Table 2.3 Decay details of the Uranium (^{238}U) decay chain [WAH07]

Parent nuclide	Half-life $t_{1/2}$	Decay Mode (% branch)	Decay Energy (MeV)	% Intensity	Daughter nuclide	γ-emission Energy (keV)	% γ-emission Intensity
^{238}U	4.5×10^{9} y	α (100)	4.198	79.0	^{234}Th	49.55	0.063
			4.151	20.9		113.50	0.0102
^{234}Th	24.10 d	β (100)	0.199	70.3	^{234m}Pa	63.28	4.1
			0.104	19.2		92.37	2.4
			0.103	7.6		92.79	2.39
^{234m}Pa	1.17 m	β (99.84)	2.269	98.2	^{234}U	1001.03	0.837
			1.224	1.007		766.38	0.294
		IT (0.16)	*		^{234}Pa	73.92	*
^{234}Pa	6.70 h	β (100)	0.642	19.4	^{234}U	131.30	0.029
			0.472	33.0		946.00	0.021
^{234}U	2.5×10^{5} y	α (100)	4.7746	71.38	^{230}Th	53.20	0.123
			4.7224	28.42		120.90	0.0342
^{230}Th	7.5×10^{4} y	α (100)	4.6870	76.3	^{226}Ra	67.672	0.373
			4.6205	23.4		143.872	0.0483
^{226}Ra	1600 y	α (100)	4.7843	94.45	^{222}Rn	186.21	3.59
			4.601	5.55		262.27	0.0050
^{222}Rn	3.8235 d	α (100)	5.4894	99.92	^{218}Po	511.00	0.076
^{218}Po	3.10 m	α (99.98)	6.0024	100.0	^{214}Pb	**	
		β (0.02)	*		^{218}At		
^{218}At	1.60 s	α (100)	6.0024	100.0	^{214}Bi	*	
^{214}Pb	26.8 m	β (100)	0.671	48.9	^{214}Bi	351.93	35.1
			0.728	42.2		295.22	18.2
			1.023	6.3		241.99	7.12
^{214}Bi	19.9 m	β (99.98)	3.272	18.2	^{214}Po	609.31	44.6
			1.542	17.8		1764.50	15.1
			1.507	17.02		1120.29	14.7
		α (0.02)	5.452	53.9	^{210}Tl	1238.11	5.78
			5.516	39.2		2204.21	4.98
^{214}Po	164.30 μs	α (100)	7.6868	99.99	^{210}Pb	799.7	0.0104
^{210}Tl	1.30 m	β^- (100)	4.209	30.0	^{210}Pb	*	
			1.863	24.0			

* No data ** No γ-rays observed

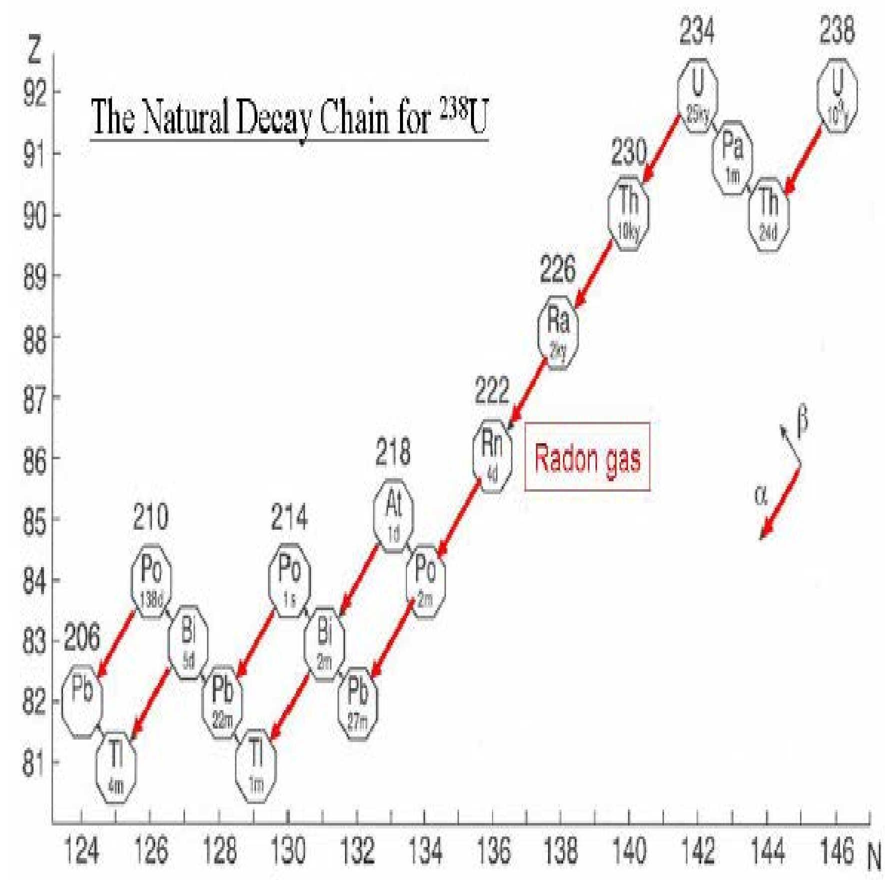

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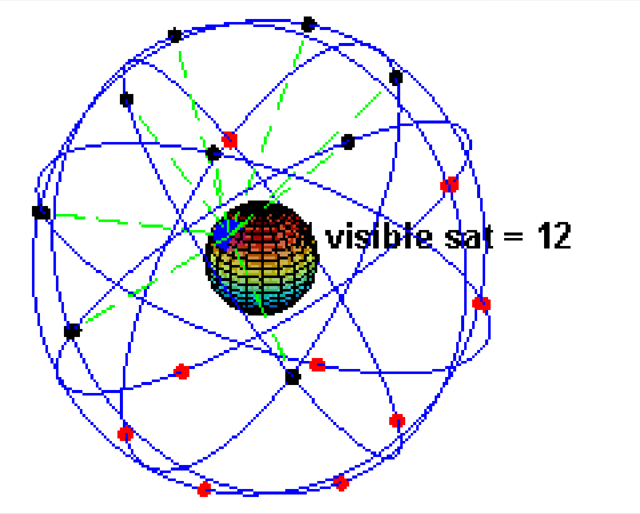
Table 2.3 Decay details of the Uranium (^{238}U) decay chain (continued) [WAH07]

Parent nuclide	Half-life $t_{1/2}$	Decay Mode (% branch)	Decay Energy (MeV)	% Intensity	Daughter nuclide	γ-emission Energy (keV)	% γ-emission Intensity
^{210}Pb	22.3 y	β (100)	0.0166	84.0	^{210}Bi	46.54	4.25
			0.0631	16.0			
^{210}Bi	5.013 d	β (100)	1.1615	100.0	^{210}Po	**	
^{210}Po	138.376 d	α (100)	5.3043	99.99	^{206}Pb	803.10	0.00122
^{206}Pb	Stable end product						

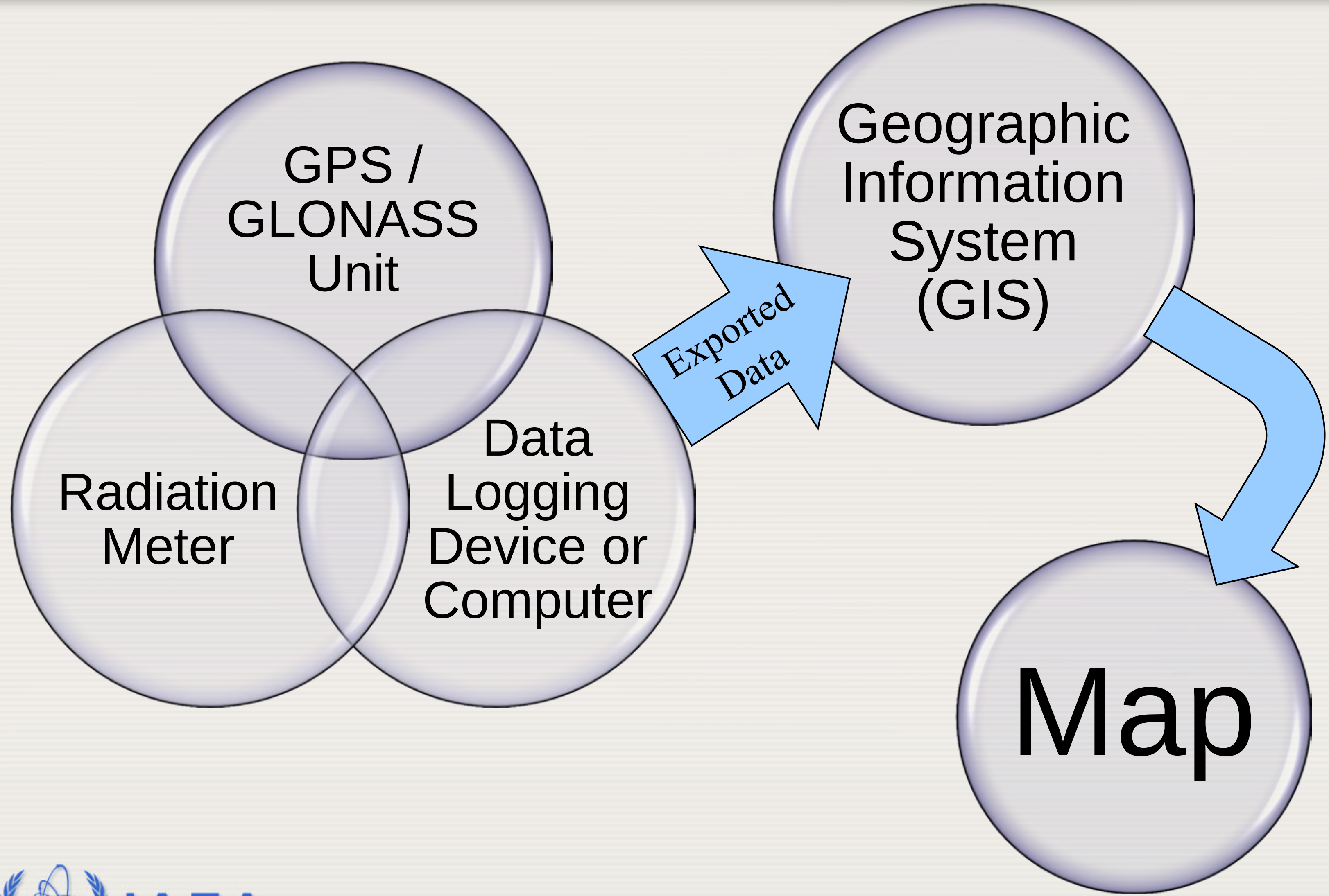
* No data ** No γ-rays observed

Global Navigation Satellite Systems

- Global Navigation Satellite Systems:
  - Global Positioning System (GPS)
  - Globalnaya Navigatsionnaya Sputnikovaya Sistema (GLONASS)
- Constellation of satellites transmit messages that include:
  - Time the message was transmitted
  - Satellite position at time of message transmission
- GPS receivers use the signals to determine their current location, the time, and their velocity



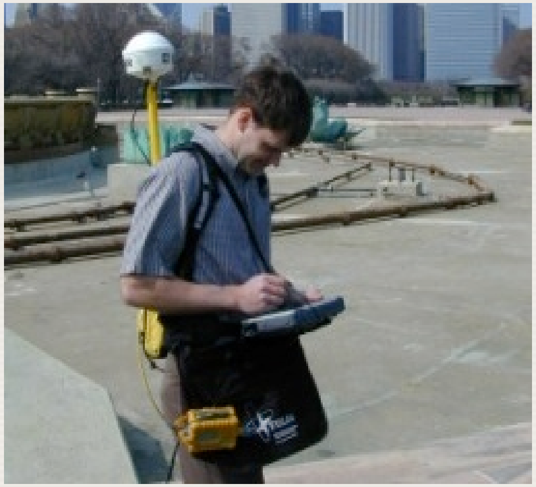
Geospatial Data Collection Systems



Geospatial Data Collection Systems

Systems may be developed from separate components:

- GNSS receiver / antenna
- Radiation meter with digital output
- Mobile computer
- Data collection software / firmware



Systems are now available that integrate many or all of these components into one device

An ideal analytic technique

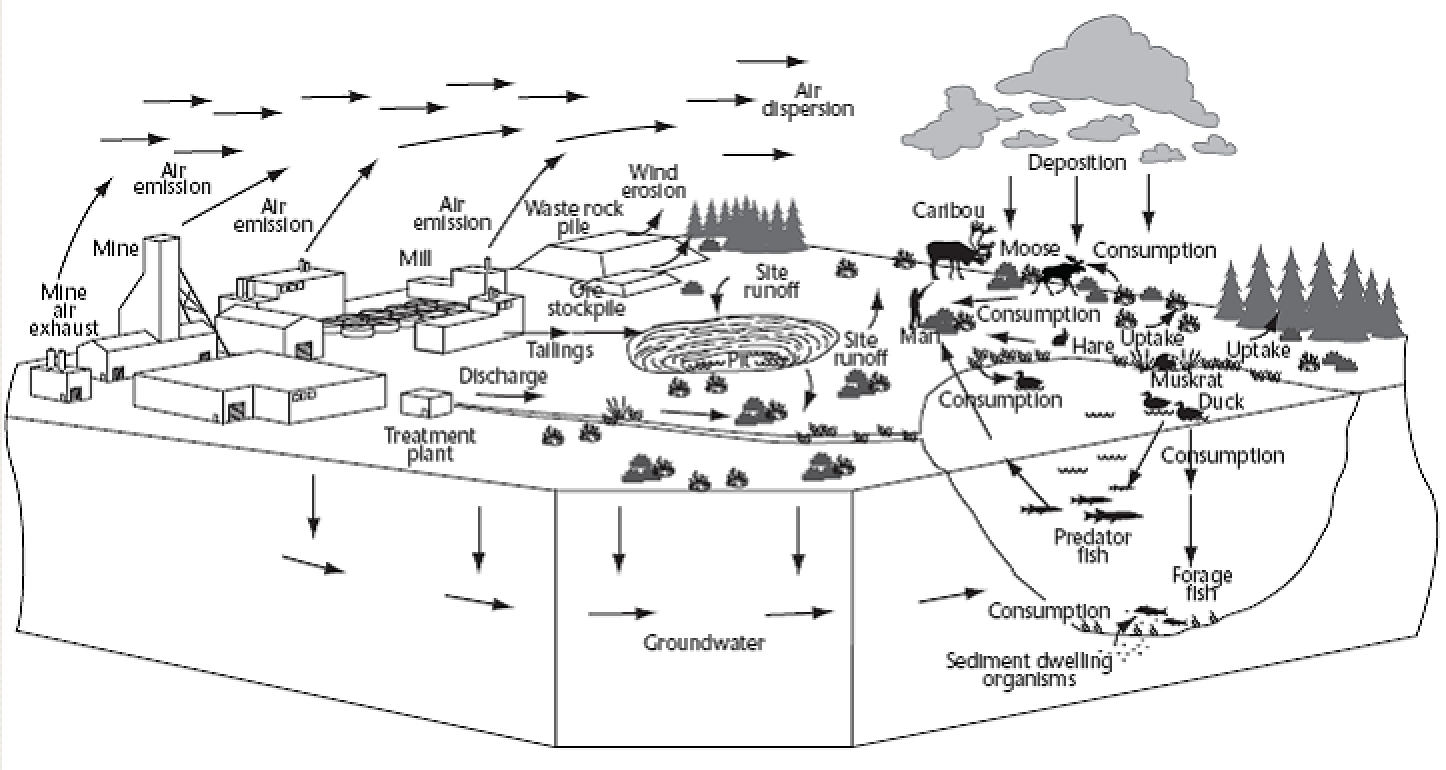
- Uncertainty of measurement result
  - Uncertainty is the parameter associated with the result of a measurement that characterizes the dispersion of the values that could reasonably be attributed to the measurand
  - **Minimal possible uncertainty !**
- Time delay in obtaining the results
  - **Immediate !**
- Cost
  - **Minimal !**

In real life what matters is

- Fitness for purpose
  - o To which extent the analytical method fulfils the expectations in regard to the results of analysis
- Analytical Problem definition
  - o What my sample is?
    - Matrix type
    - Homogeneity of distribution of the property
  - o What do I need to assess in it?
    - Analytes (COC)
  - o What is the expected level of presence of the analyte?
    - Mass fraction, activity concentration
  - o How accurate and uncertain can be the results?
    - Depends on the purpose of the characterization

Cycle monitoring

Problem: Monitoring of Radionuclides and/or other Hazardous Substances in the Environment



Example of conceptual model for a mining / milling site

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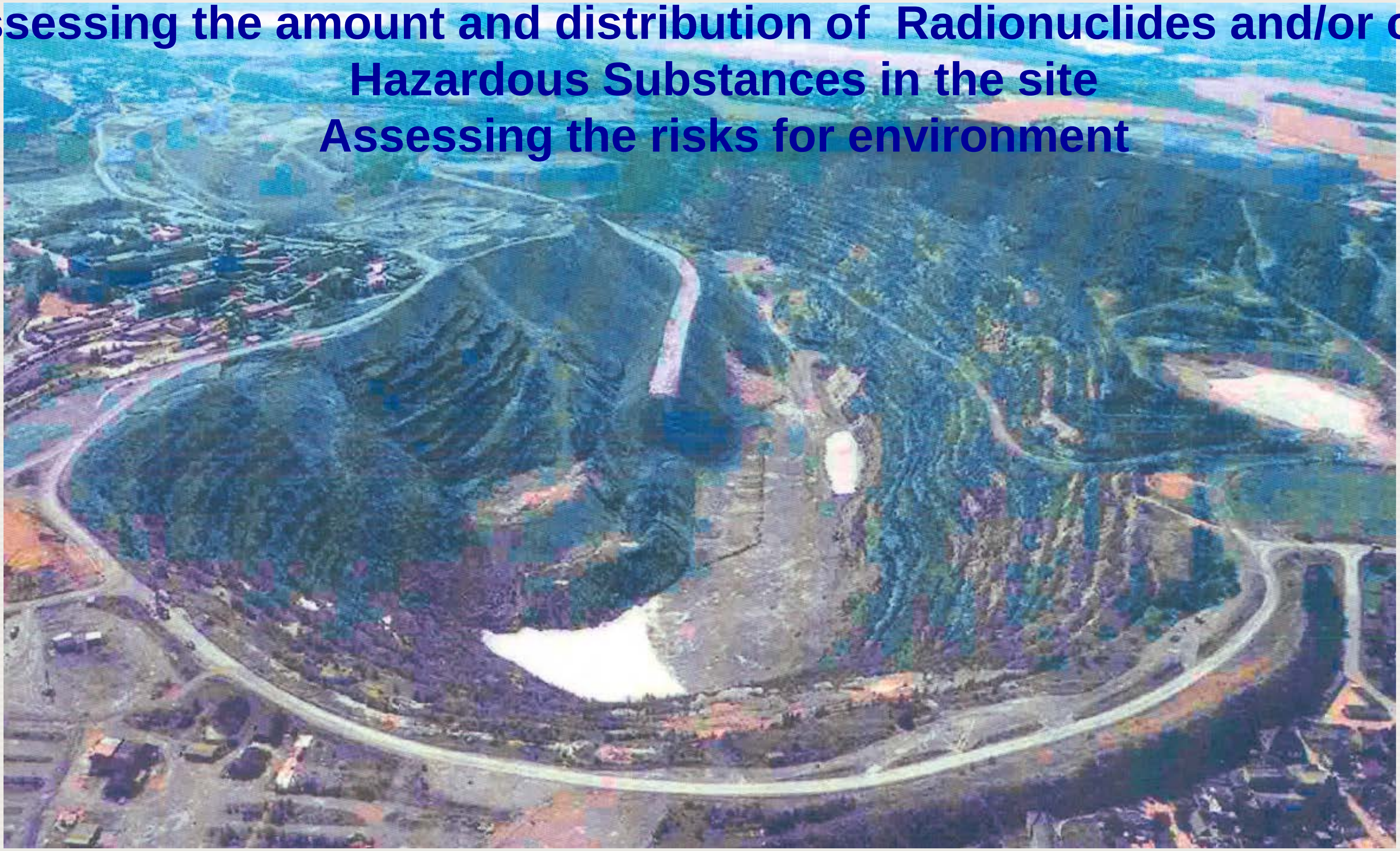
Characterization of sites for remediation

Problems:

Assessing the amount and distribution of Radionuclides and/or other Hazardous Substances in the site

Assessing the risks for environment



Typical cases of radiologically affected sites

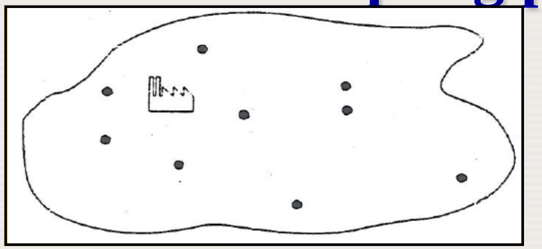
- Uranium mining / milling sites
- Sites with increased amounts of NORM
  - mining of phosphate rocks, REE, bismuth, zirconium, titanium
- Sites affected by discharges (accidental or planned) of radionuclides
- Nuclear weapons test areas
- Military sites
- Nuclear industry or other radiological facilities accidents

Site characterization challenges

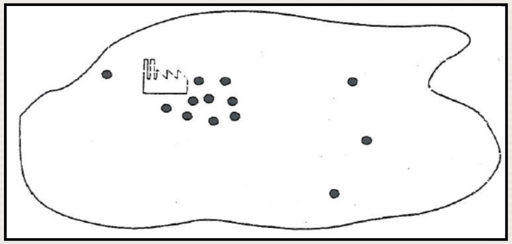
- Samples may differ by composition and aggregation
- The concentration of contaminants of concern (abundance) is unknown
- Heterogeneous spatial distribution of the COC
- Need to analyze different compartments (soil, water, biota)
- Large amount of samples required to evaluate the status and extent of the contamination

Characterization based on laboratory analysis

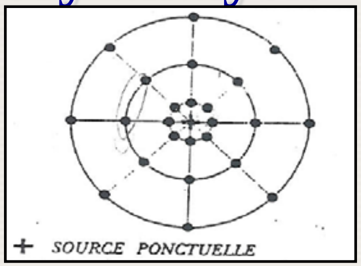
Different sampling plans for further laboratory analysis



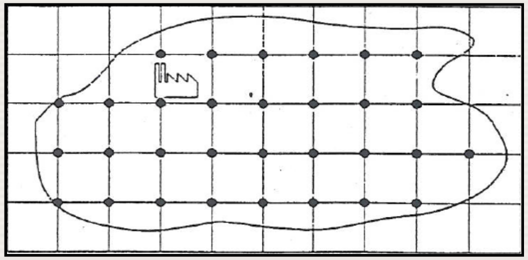
Random sampling



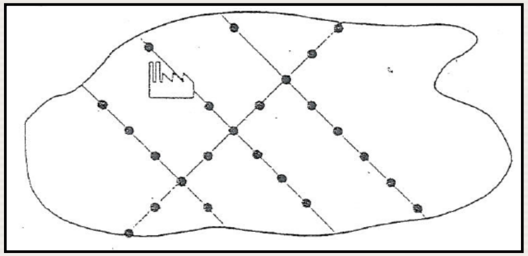
Appraisal sampling



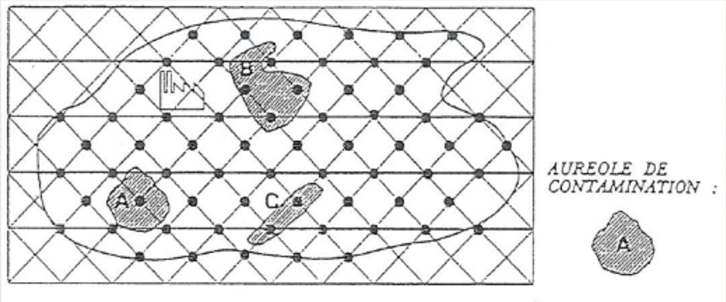
Circular grid sampling



Regular sampling



Profile sampling

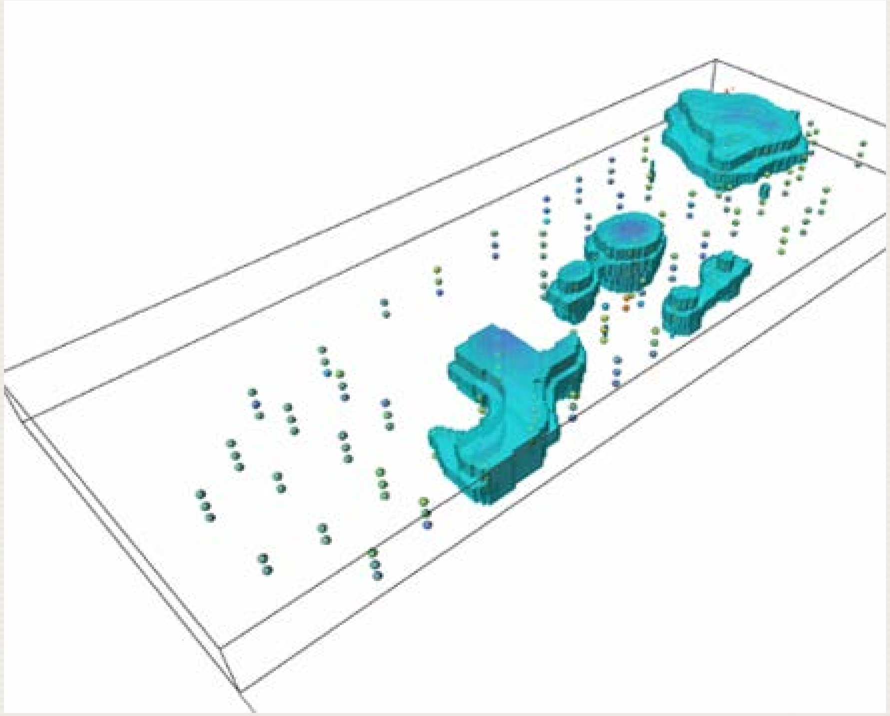


Specific search sampling

Sampling in the site

Requires of previous knowledge (history)
- Not always known
- Possibly biased or wrong

→ Large amounts of samples sent for analysis might not necessarily produce useful information



Balanced order of priorities in site characterization

- Is there contamination (Y/N)
  - Trigging level corresponding to established (or to be established) regulation
    - Presence or not
- How to assess the extent (spatial distribution) of the contamination?
    - Knowledge of previous activities + logic sampling campaign
    - In-situ fast screening
- Accuracy required for the results…
  - High? Poor? Acceptable?
- Additional needs:
  - To avoid misclassifications (false positives/ negatives)
  - To minimize hazardous exposure
  - To reduce costs
  - To shorten time delay for decision taking

Analytic techniques for radiological assessment

Sample	Laboratory	In-situ
Aerosol / gas	-	Radon (alpha sp.)
APM	Large volume collectors through filter media + alpha, beta and gamma Sp.	Continuous measurement by alpha
Soil sediments tailings	Gamma (NaI or HPGe) Alpha , Beta Radon (etch track + lab)	Dose rate meters Gamma (NaI or HPGe) Radon (alpha or beta)
Water bodies	LSC (alpha – beta separation)	Gamma
Biota, biomonitors	Total alpha-beta Gamma (NaI or HPGe)	-

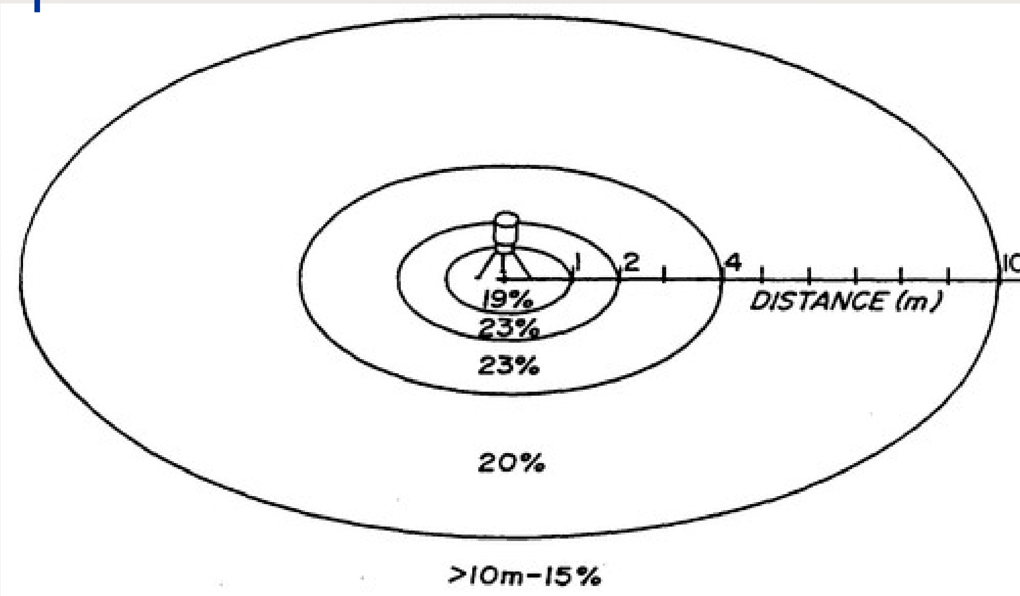
Laboratory vs. in-situ

Gamma spectrometry:

	Laboratory	In-situ
Relative uncertainty of the result	5 - 10 %	20 %
Measurement time (min)	~ 180	30
Cost (USD)	~ 200	~ 50 - 100
Samples per 100 m^2	min 30	~ 400
Turn-around time	weeks	days

Laboratory vs. in-situ

Gamma spectrometry: Sample size



Marinelli beaker: ~ 1 dm^3 Area: 10 m x 0.3 m ~ 95 m^3 (85 %)

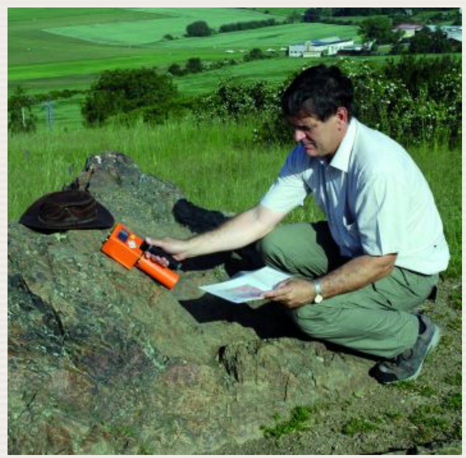
In-situ techniques for radiological assessment

Surface measurements:

Shift from Gas-filled (GM / PIC / PC) to

Low resolution gamma spectrometry (Scintillation detectors)

NaI(Tl), BGO, LaBr CdZnTe



Measurement time:

For dose rate ~ 1 s

For radionuclide activity concentration ~ 1 – 5 min.

In-situ techniques for radiological assessment

Surface measurements
High resolution gamma spectrometry
HPGe semiconductor detectors

Measurement time:
For radionuclide activity concentration ~ 30 minutes

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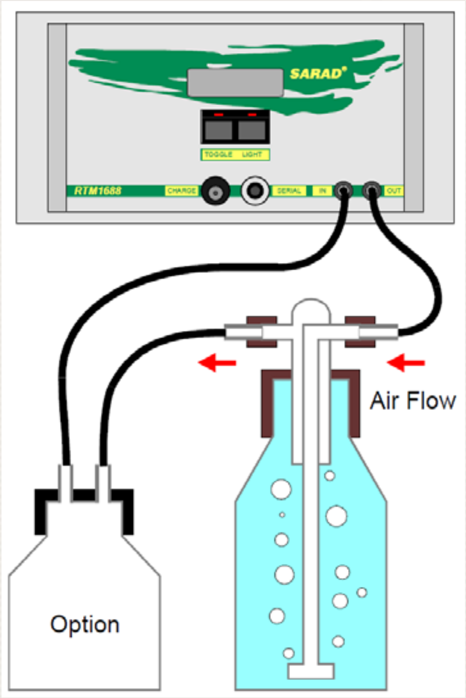
In-situ techniques for radiological assessment

Radon measurements



Measured radiation hazards:

- Soil gas radon / thoron activity concentration
- Radon activity concentration in aerosols
- Dose rate (gamma scintilliation detector)
- Gamma emitting radionuclides activity concentration in soil

Measurement techniques / instruments:

- Soil gas radon / thoron activity concentration

- air pressure, temperature and humidity sensors,
- built-in flow regulated pump
- spectrometer to process the measured signal.
- 4 measurement chambers, each one containing a 2 cm^2 ion implanted silicon detector.
- Measurement time 15 minutes
- Radon: ^{218}Po ($T_{1/2}$ = 3.1 m, E = 6.115 MeV)
- Thoron: ^{216}Po ($T_{1/2}$ = 0.145 s, E = 6.906 MeV)

Unit SARAD RT-1688

Measurement techniques / instruments:

- Soil gas radon / thoron sampling probes (large volume)

Hand drilling set with gravel head,
packer probe
alternative machine drill and generator

Measurement techniques / instruments:

- Sampling procedure (large volume probe)

1) A hole is made with the hand-operated drilling systems and,

2) the packer probe is introduced into the hole.

Measurement techniques / instruments:

- Sampling procedure (large volume probe)

3) The packer probe is kept inside the hole and the packer sealing is inflated with a small hand pump.

Measurement techniques / instruments:

- Sampling procedure (large volume probe)

4) The packer probe's tube is connected to the instrument's inlet and the measurement is started.

Measurement techniques / instruments:

- Soil gas radon / thoron sampling probes (small volume probe)

Measurement techniques / instruments:

- Sampling procedure (small volume probe)

1) The hollow probe (with attached sharpened disposable tip)

is punched into the ground.

Measurement techniques / instruments:

- Sampling procedure (small volume probe)

2) The punch wire is inserted into the hollow probe, so the sharp tip can be pushed out and a small sampling volume is formed.

Measurement techniques / instruments:

- Sampling procedure (small volume probe)

3) The punch wire is extracted

Measurement techniques / instruments:

- Sampling procedure (small volume probe)

4) The radon measurement instrument is connected and the measurement cycle (integration time 5 minutes, measurement time min. 15 minutes) is started

Measurement techniques / instruments:

- Radon / thoron activity concentration in aerosols

Multipurpose unit A2M-4000

- air pressure, temperature and humidity sensors,
- built-in flow regulated pump
- 3 built-in spectrometers to process
  - soil gas activity measurement made with 4 chambers with 2 cm^2 ion implanted silicon detectors.
  - Alpha and beta measurements of aerosols using an arrangement of a 4 cm^2 filter and 2 ion implanted silicon detectors
  - Input (3^{rd} channel) to connect a NaI detector

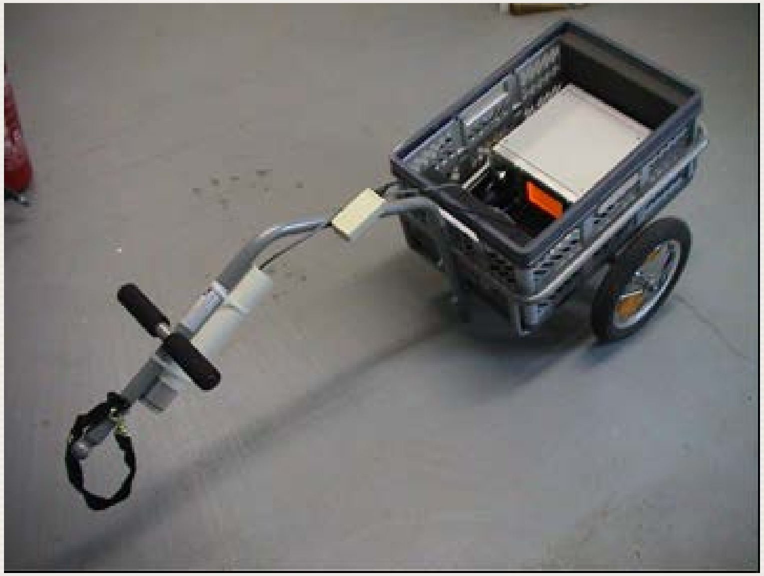
Measurement techniques / instruments:

- (Gamma) Dose rate measurement at 1 m above the soil surface using a back pack system (Tetra Tech Inc.)

- 2” x 2” NaI(Tl) detector (Ludlum 44-10),
- A counting unit recording total counts (Ludlum 2350-1)
- GPS sensor (GlobalSat BU-353) with enabled WAAS and EGNOS capabilities, thus providing improved accuracy in positioning (± 2 m).
- A portable computer with dedicated software allowing collecting the measurement result and the GPS coordinates every second.
- Measurement time 1 s
- Walk pace ~ 1 m/s

Measurement techniques / instruments:

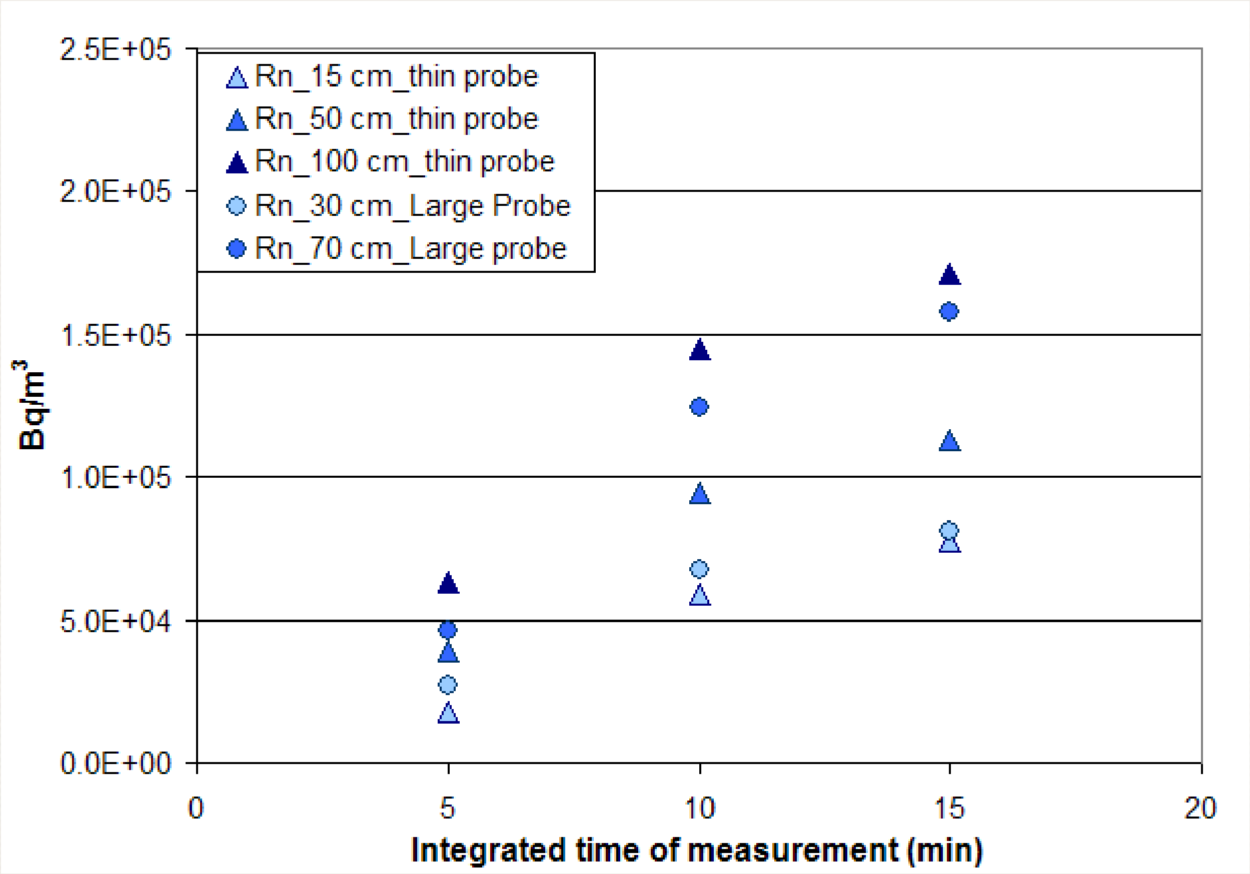
- Gamma spectrometry measurements using a transportable system (SARAD)



- 2” x 2” NaI(Tl) detector,
- A2M4000 multipurpose unit
- Measurement grid 10 x 10 m pattern
- GPS sensor.
- A portable computer with dedicated software allowing collecting the measurement result and the GPS coordinates.
- Measurement time 60 s

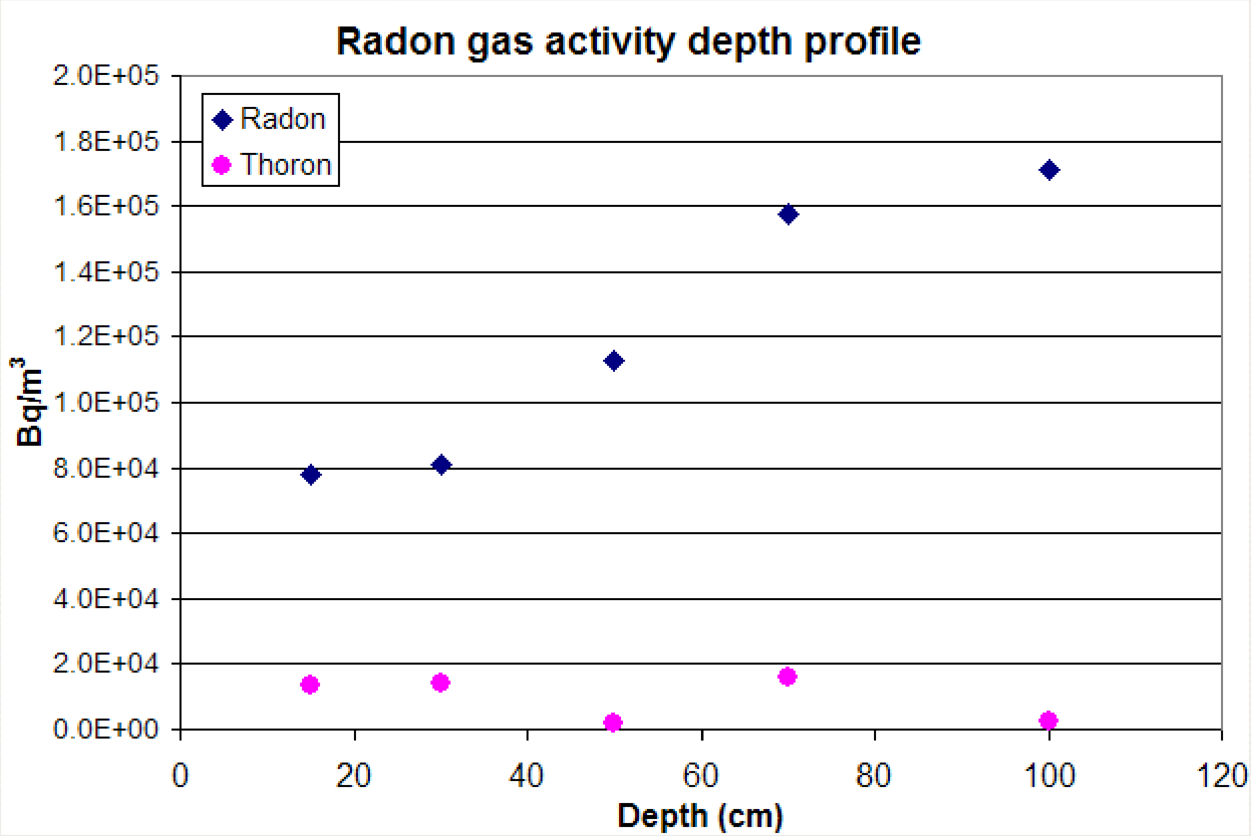
Results: Radon activity in soil gas

- Measurements using two different sampling probes



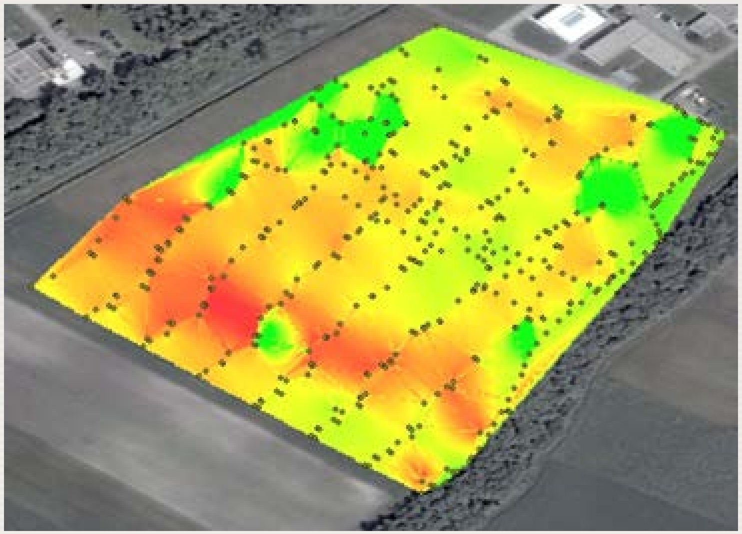
Results: Radon activity in soil gas

- Change in activity vs. depth

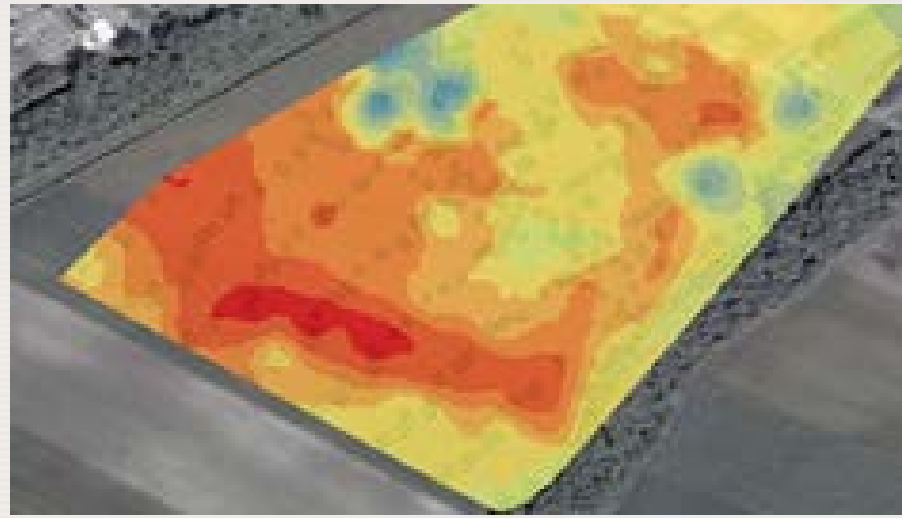


GIS representation of results

Dose rate or gamma results:



Natural neighbor interpolation:
Finds the closest subset of input samples to a query point and applies weights based on proportionate areas

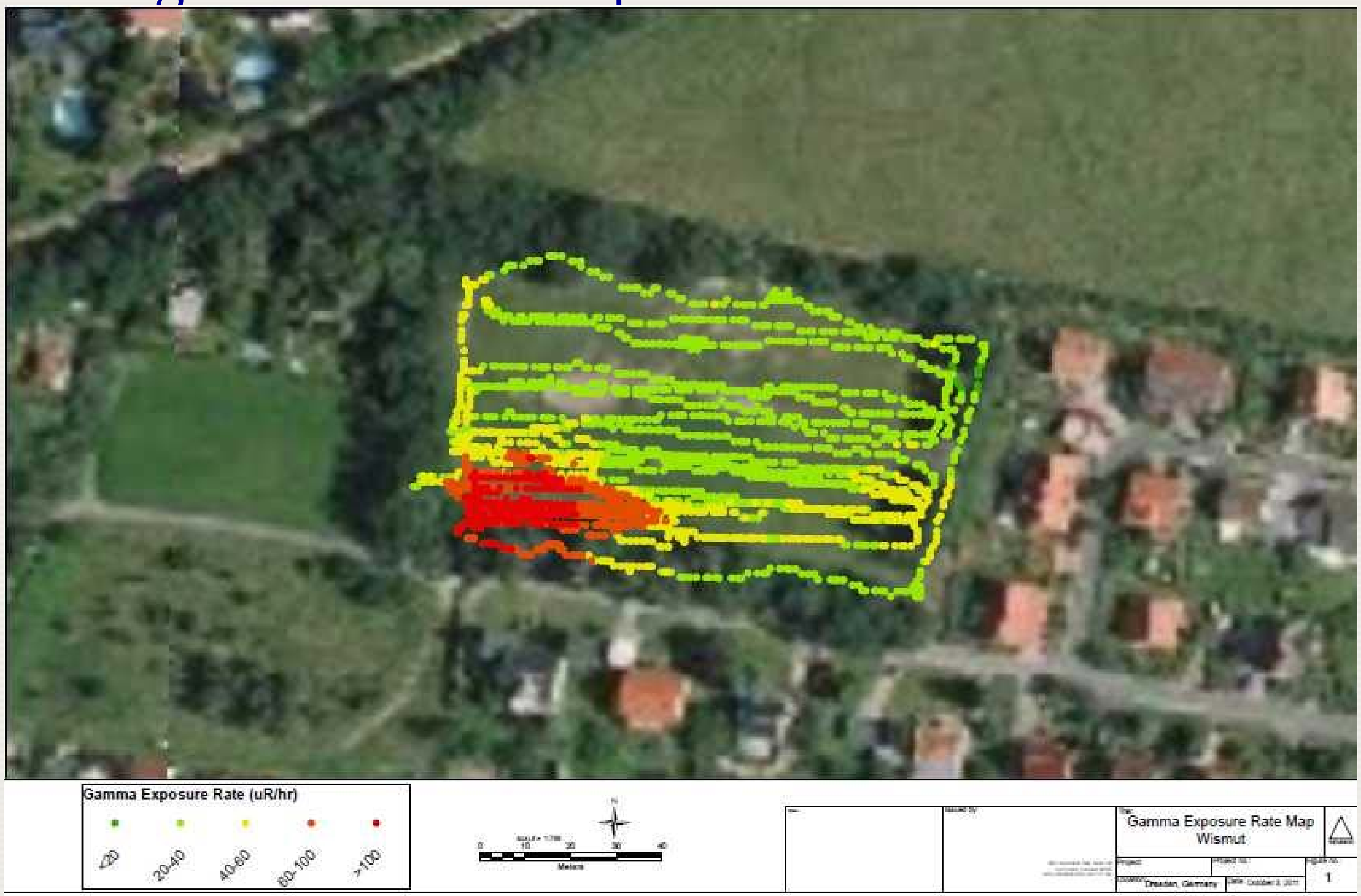


Kriging: based on the regionalized variable theory, the spatial variation represented by the values is statistically homogeneous throughout the surface

ArcGIS

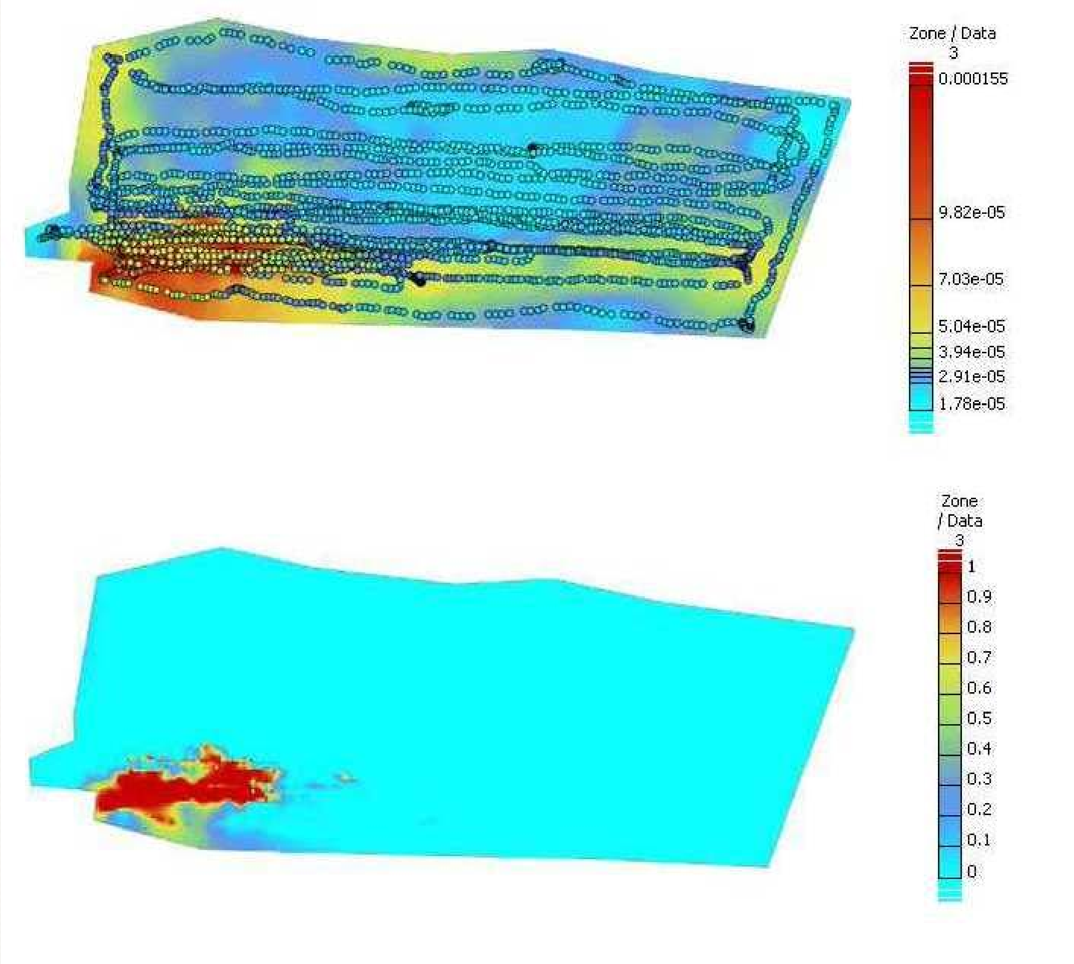
Results: Dose rate measurements using backpack system

- Representation as colour-coded plot of the dose rate (uR/h) data in a geo-referenced map



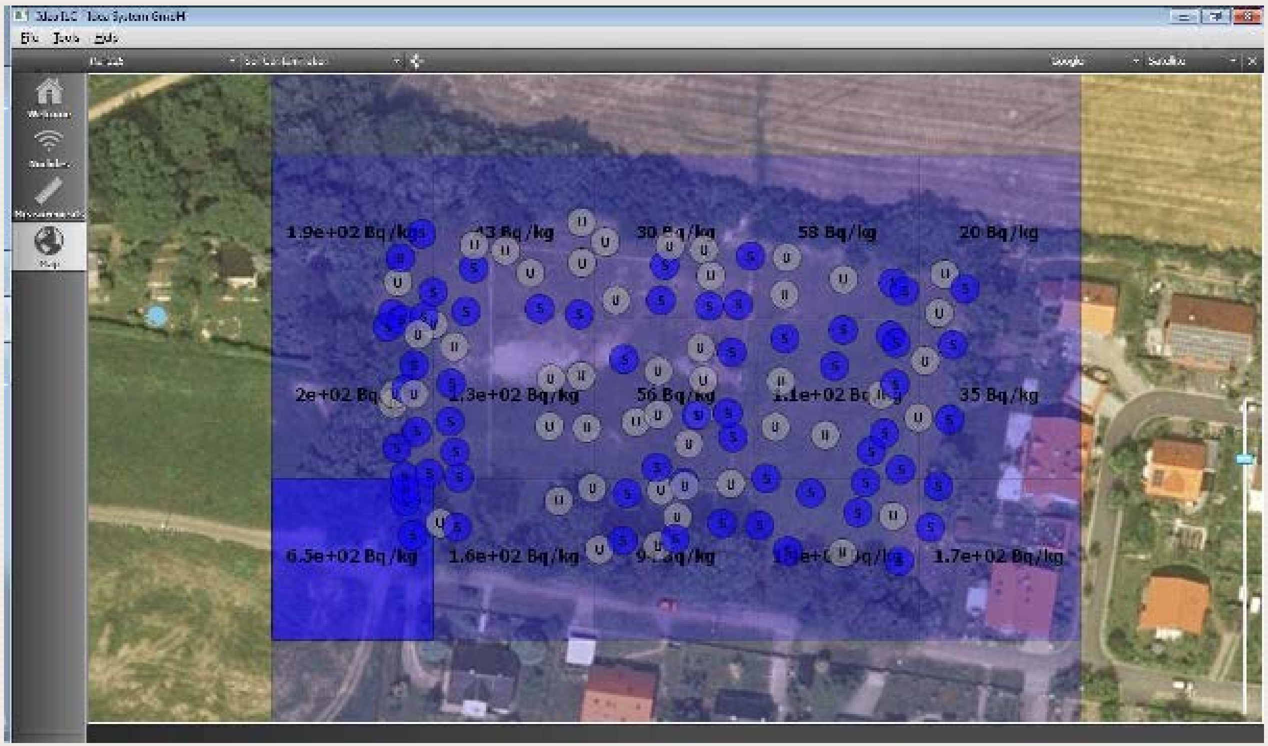
Results: Dose rate measurements using backpack system

- Interpretation of the dose rate (R/h) measurements with Kartotrak (Geovariances, CEA)



Results: Gamma activity concentration measurements

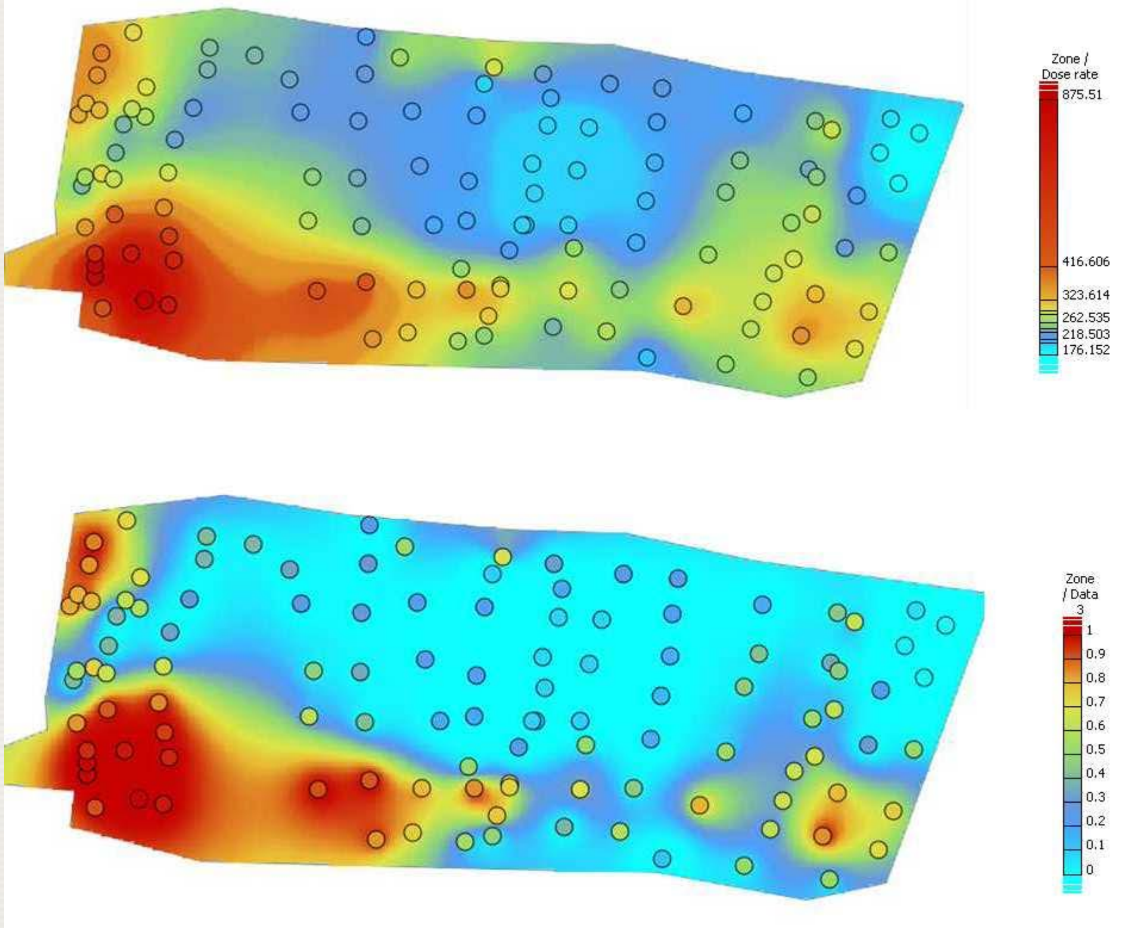
- Interpretation with IDEA ILC software (SARAD)



using a retrospective algorithm the average ^{226}Ra mass activity (Bq/Kg) is calculated for sections of the measured site area

Results: Gamma activity concentration measurements

- Interpretation of the activity concentrations (Bq/kg) results with Kartotrak (Geovariances, CEA)



Analytic techniques for inorganic analysis

Sample	Laboratory	In-situ
APM	Large volume collectors through filter media + XRF, PIXE, INAA AAS, ICP-OES, ICP-MS	-
Soil sediments tailings	Direct: XRF, PIXE, INAA Digestion + AAS, ICP-OES, ICP-MS	XRF
Water bodies	TXRF, ICP-MS, AAS	TXRF
Biota	Direct: XRF, PIXE, INAA Digestion + AAS, ICP-OES, ICP-MS	-

Laboratory vs. in-situ

X-ray spectrometry:

	Laboratory	In-situ
Relative uncertainty of the result	5 - 10 %	20 %
Measurement time (min)	~ 15 - 30	5
Costs (USD)	~ 40 (XRF) ~ 150 (ICP)	~ 5

Laboratory vs. in-situ

X-ray spectrometry: Sample size

Pellets, loose powder: ~ 4 g

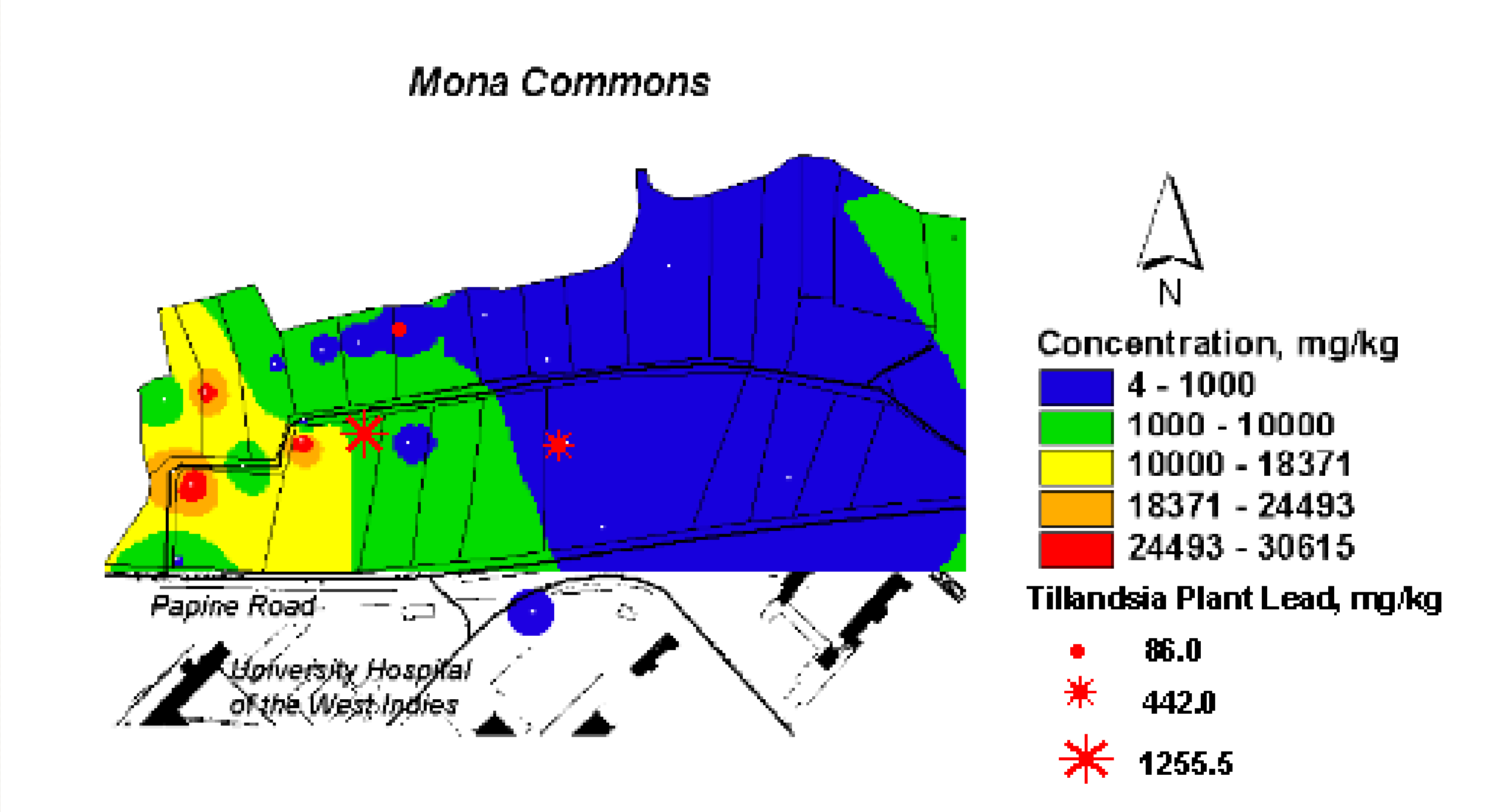
Composite sample

Probed layer: ~ 2 – 5 mm

Measurement at different depths

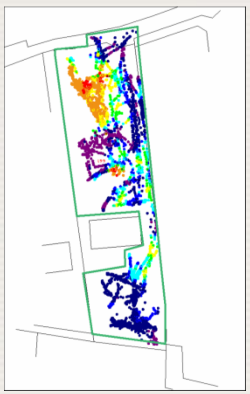
GIS representation of results

X-ray spectrometry results:
Lead contamination due to backyard melting in Kingston, JAM

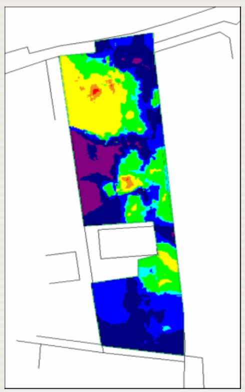


Combined approach

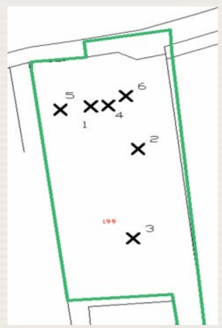
- Optimization of tools for sampling design and interpretation of results



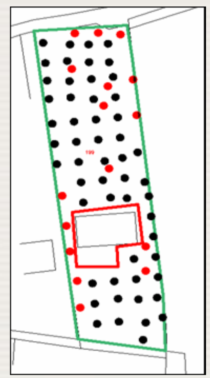
In-situ dose rate survey



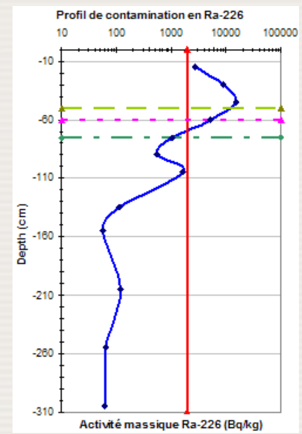
Geo-statistics interpolation Krigging



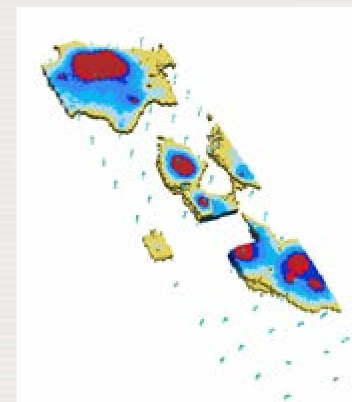
Lab analysis



Design for bore-hole sampling



Analysis and depth profile distribution In-situ or laboratory



3-D reconstruction

Observations

- Radiological characterization must comprise different types of measurements
- Sampling design as well as the interpretation of the measurement results depend on local specific safety regulations, on the intended use of the site after remediation, and on the foreseen method of containment or removal of the contaminants
- Gamma measurements carried out to reveal a surface distribution pattern mainly reflect the near surface contamination, and are influenced by the density of the soil, the energy of the measured radiation and cannot be used as a direct indication on the amount and depth distribution of the contamination.

Observations

- Short interval dose rate measurements while moving along the terrain are useful to achieve a fast screening of the near surface contamination and to locate areas with increased radiation levels. Such information is of extreme value to decide which type of measurements and sampling to be performed for radionuclide identification and concentration activity estimation

- Discrete Gamma spectrometry measurements at points of a given sampling plan are useful to identify gamma emitting radionuclides in a cost-effective way.

- Additional depth profile measurements are required to gather data allowing modelling the distribution in depth, which is in turn valuable to optimize the volume of material to be removed or to optimize containment design and elements.

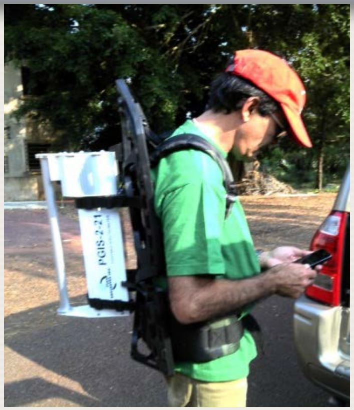
Observations

- Radon and thoron measurements are required to determine the hazards for inhalation in the case of pollution with NORM.
- There is a need to compare the performance of other type of gamma detectors and sampling heads. For example, probes for bore-hole analysis can be useful for soil depth profile studies, whereas special sampler designs could be useful for measuring activity concentration in water bodies at different depths.
- The Kartotrak areal representation of the dose rate results gathered by using two different measurement methodologies are comparable. The continuous dose rate measurements provide a fast screening, whereas the grid gamma spectroscopic measurements could be used to create areal distribution of radionuclides.

3 different "Backpack" Systems

PGIS-2-21 (PicoEnvirotec)

- NaI(Tl) 3 x 3"; Android controlled, GPS incorporated, real-time DR, U(Ra), Th, K, Cs

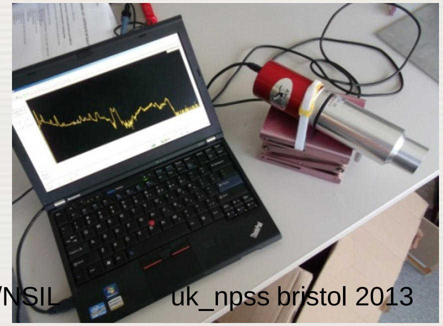


Gamma Surveyor II (GF Instruments)

- BGO 1x1.5"; GPS: serial and bluetooth
- DR & search modes, Spectrum mode

Canberra

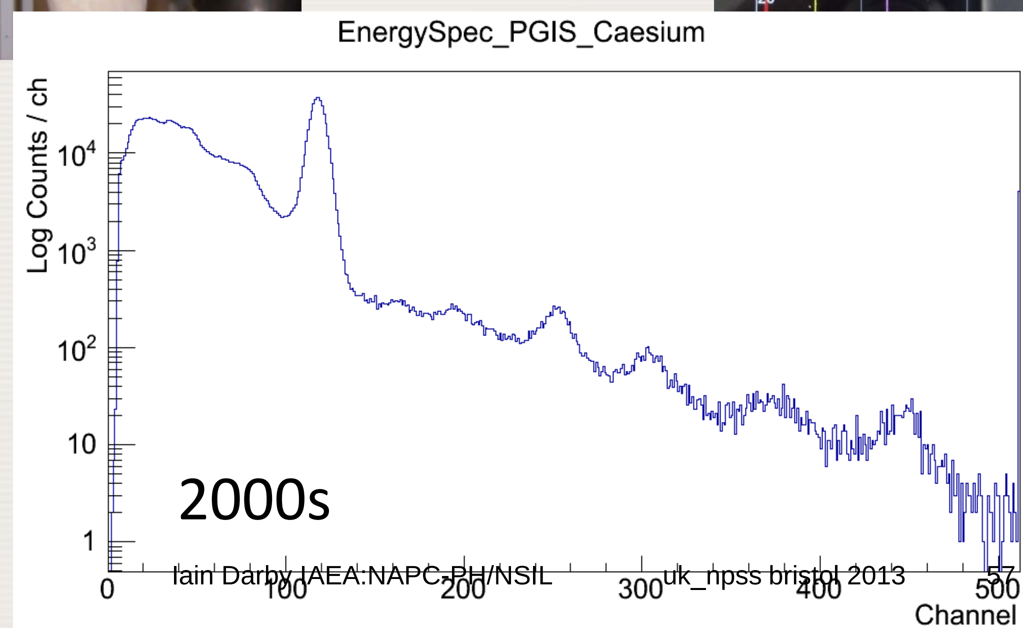
- LaBr (1.5 x1.5") + OSPREY
- Improved E-resolution



Equipment : PGIS-2-21

- PicoEnvirotech
- NaI(Tl)
  - 0.347l (21” cu)

- GPS
- Anroid operated
- Bluetooth control
- Real-time DR mapping
- auto-calibration
  - nat K, Th

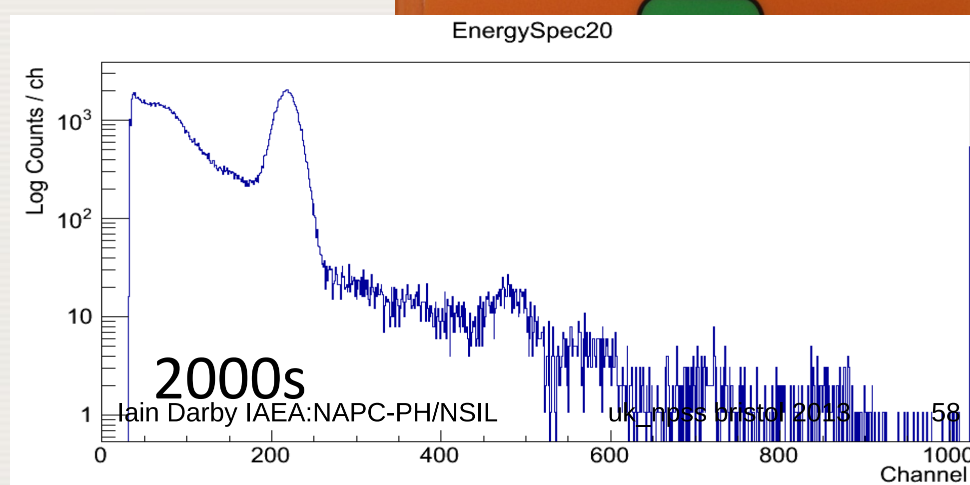


IAEA

Equipment : Gamma Surveyor II

SG/TC Trainee

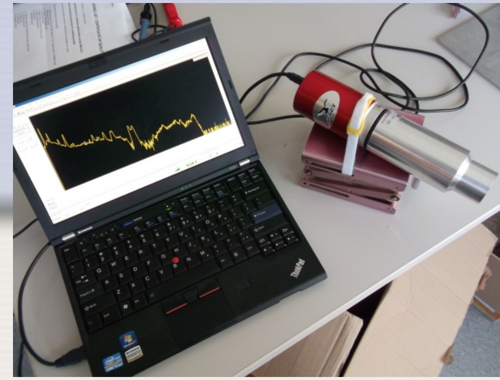
- GF Instruments CZ
- BGO 1 x 1.5”
  - 1024 channel
  - K, U, Th assays
  - DR & search modes
  - Spectrum mode
- GPS
  - serial
  - Bluetooth



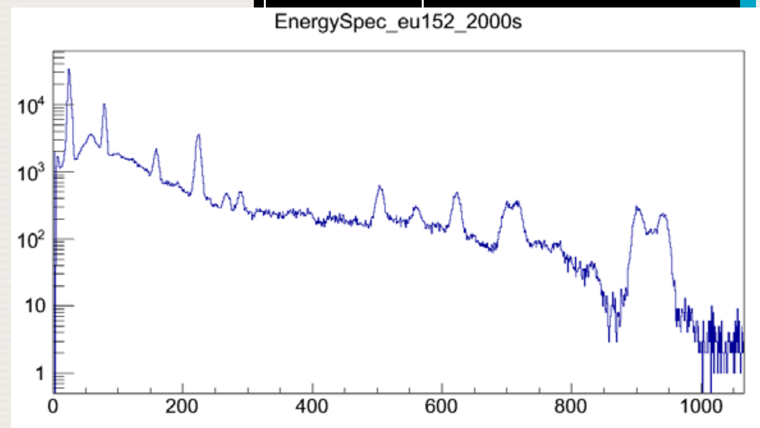
IAEA

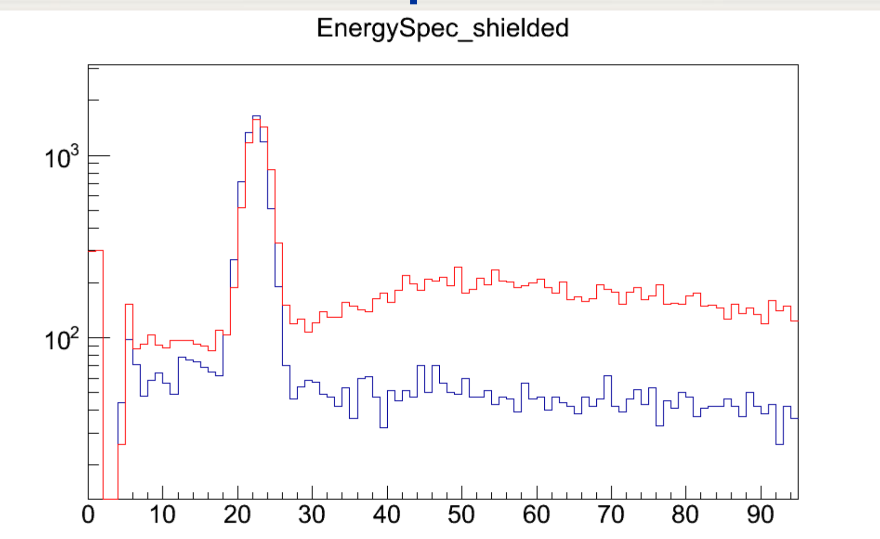
Equipment : LaBr + OSPREY™

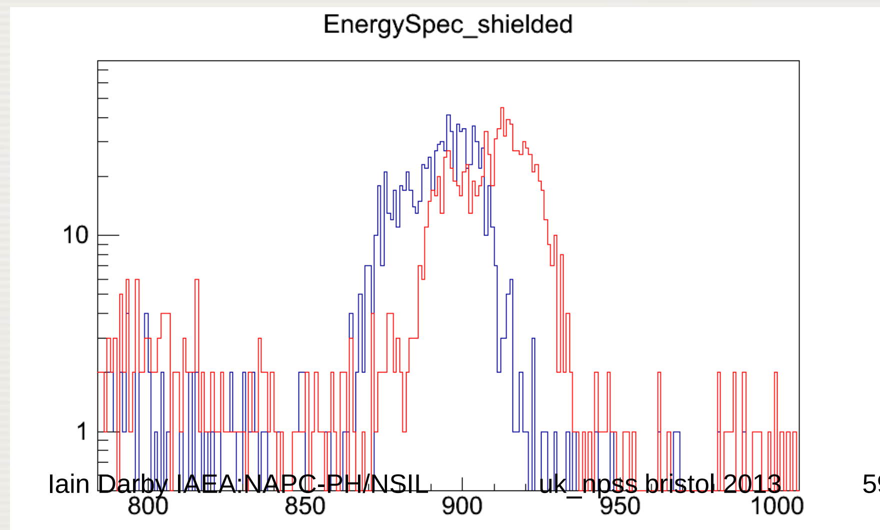
- Canberra
- $LaBr_3$(:Ce) 1.5 x 1.5
- Digital MCA
- Genie2k controlled
- 2048ch
- 506V usb powered



138Ce ≥0.9E+14 Y 0.251% 2ε: 100.00%	139Ce 137.641 D ε: 100.00%	140Ce STABLE 88.450%
137La 6E+4 Y ε: 100.00%	138La 1.02E+11 Y 0.08881% ε: 65.60% β-: 34.40%	139La STABLE 99.9119%
136Ba STABLE 7.854%	137Ba STABLE 11.232%	138Ba STABLE 71.698%

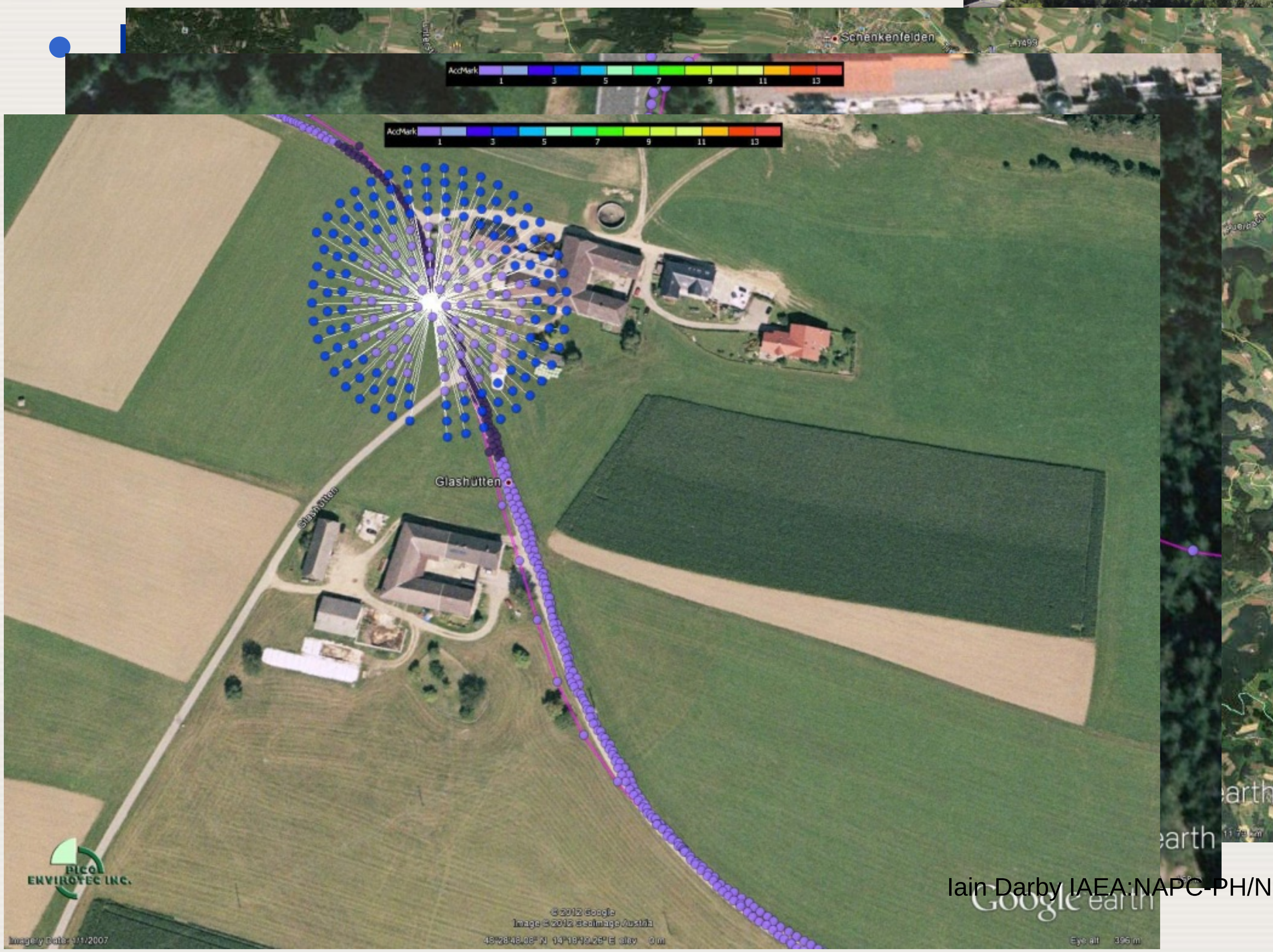






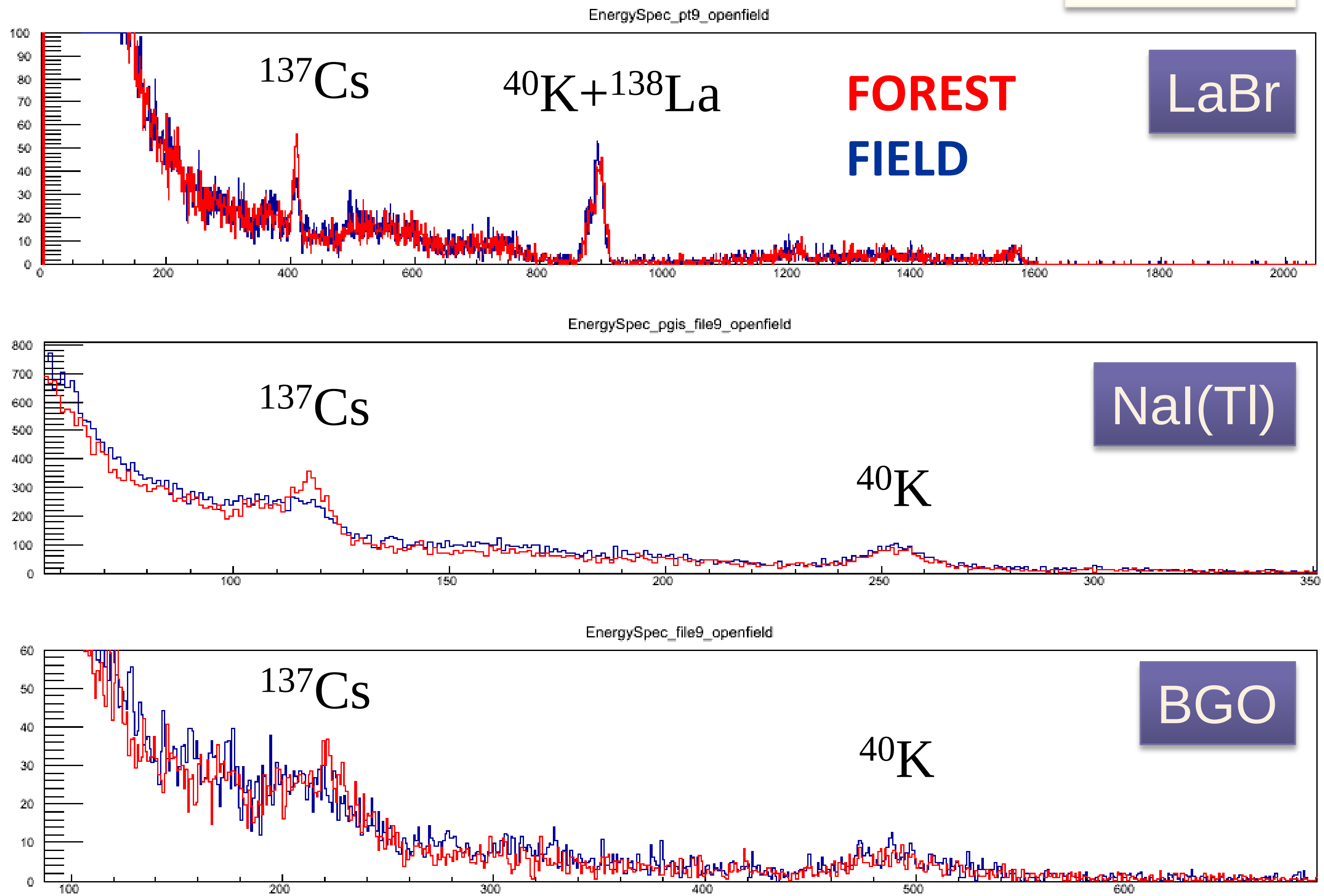
Austrian Radon Project

- Austrian Radon Project



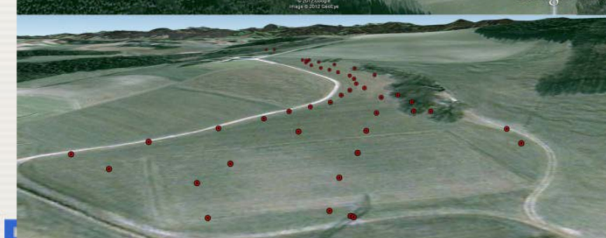
Evaluation of detectors performance: Austrian Radon Project

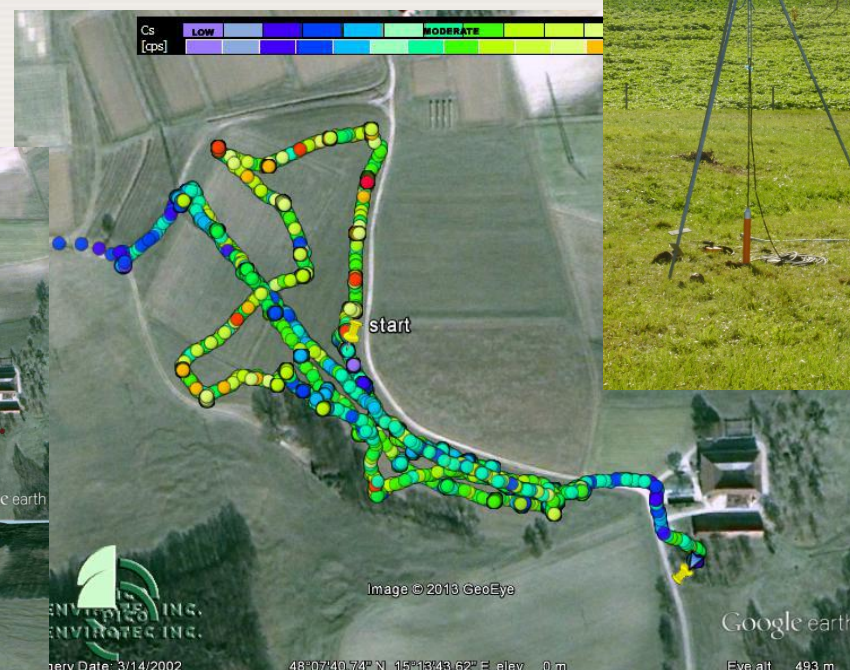
t = 300s



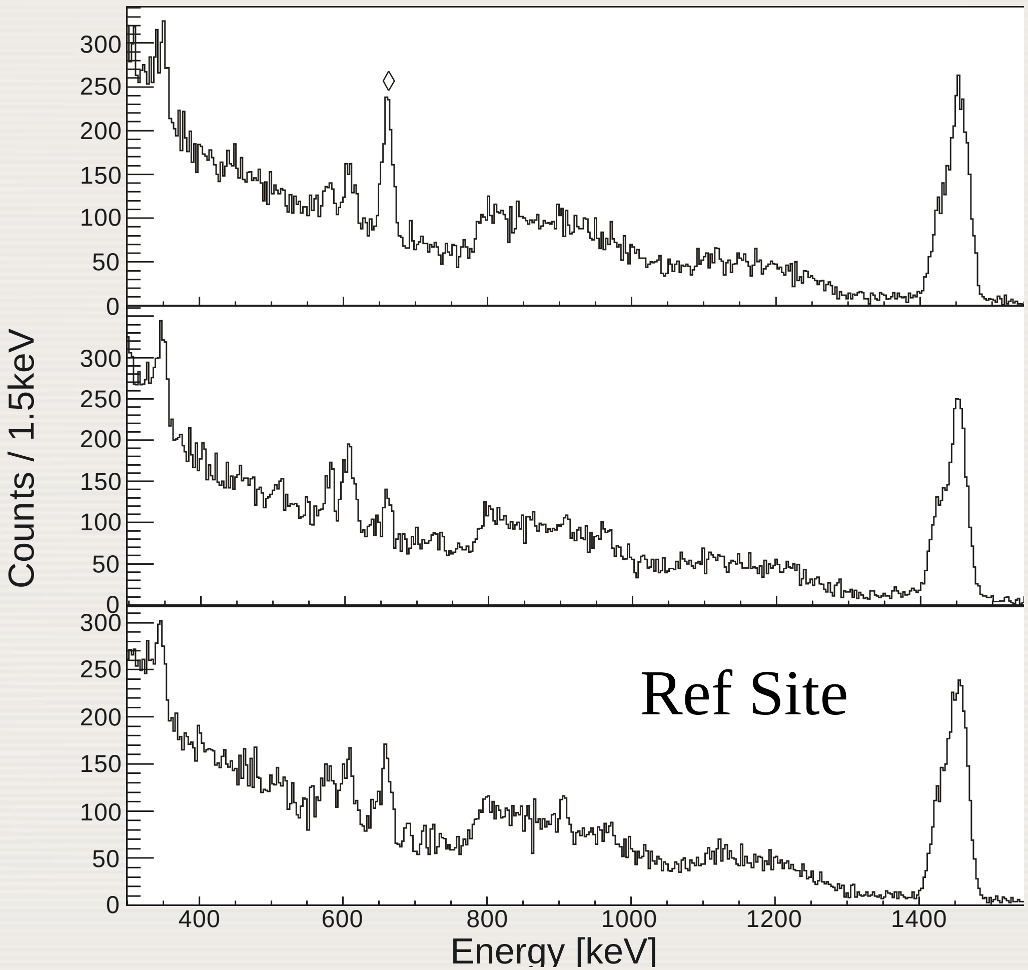
2012 results: Joint project with Soil Science Unit (NAFA) – Feasibility for ^{137}Cs soil erosion study

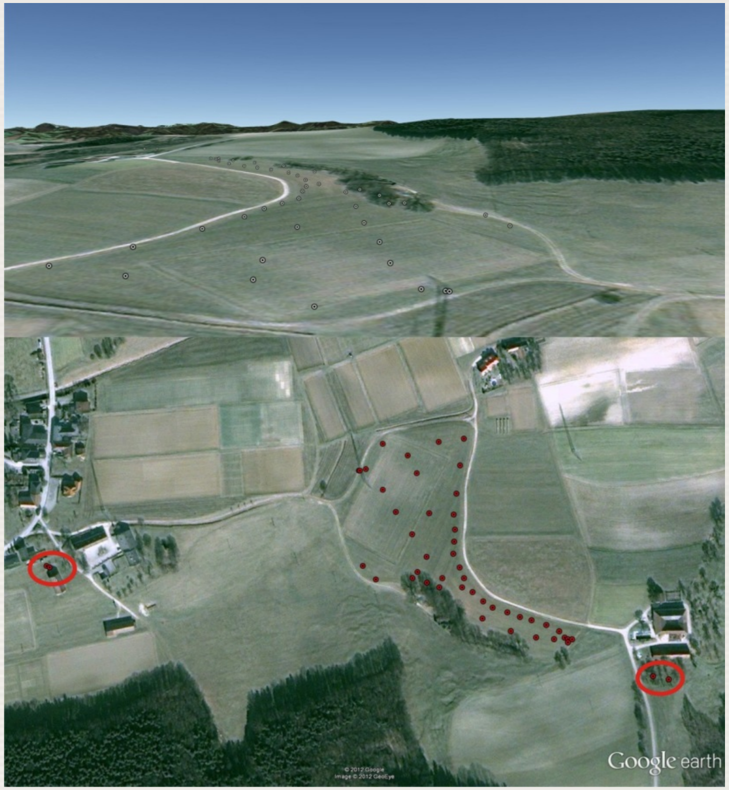
- Feasibility of the instruments assessed for surface and bore-hole measurements
- Activity levels and changes detectable with LaBr detector





Data from Grabenegg





Uranium exploration and mining

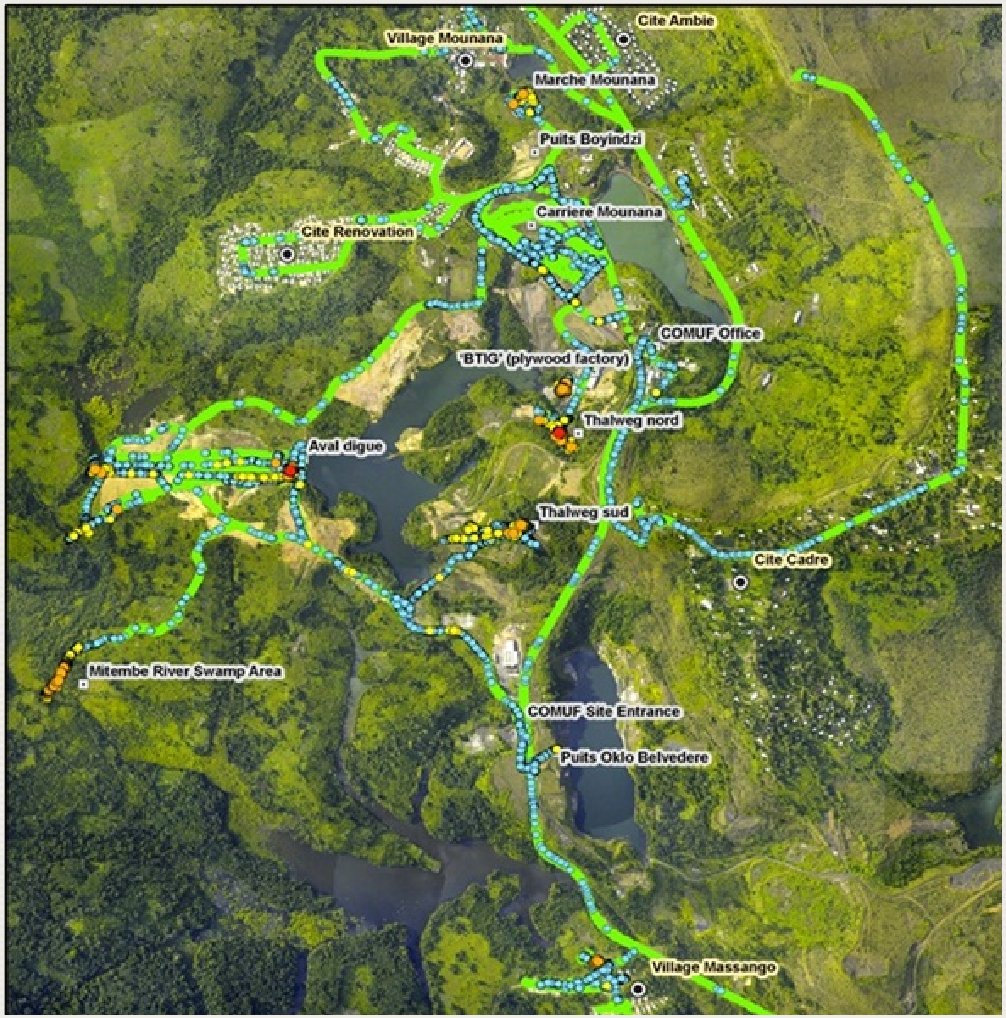
MEGATEM, Dash 7

NSIL/NEFW activities: Demonstration of capabilities for surface characterization of NORM affected sites

Gabon (July, 2012)

- 114 165 measurements (5 instruments)
- 2 ½ days of measurements
- Detailed surveyed areas

Dose rate (nSv/h)	Number	(%)
10 - 114	61 267	56.7
114 - 250	24 771	22.9
250 - 500	11 660	10.8
500 - 1140	5 283	4.9
1140 - 10400	5 036	4.7



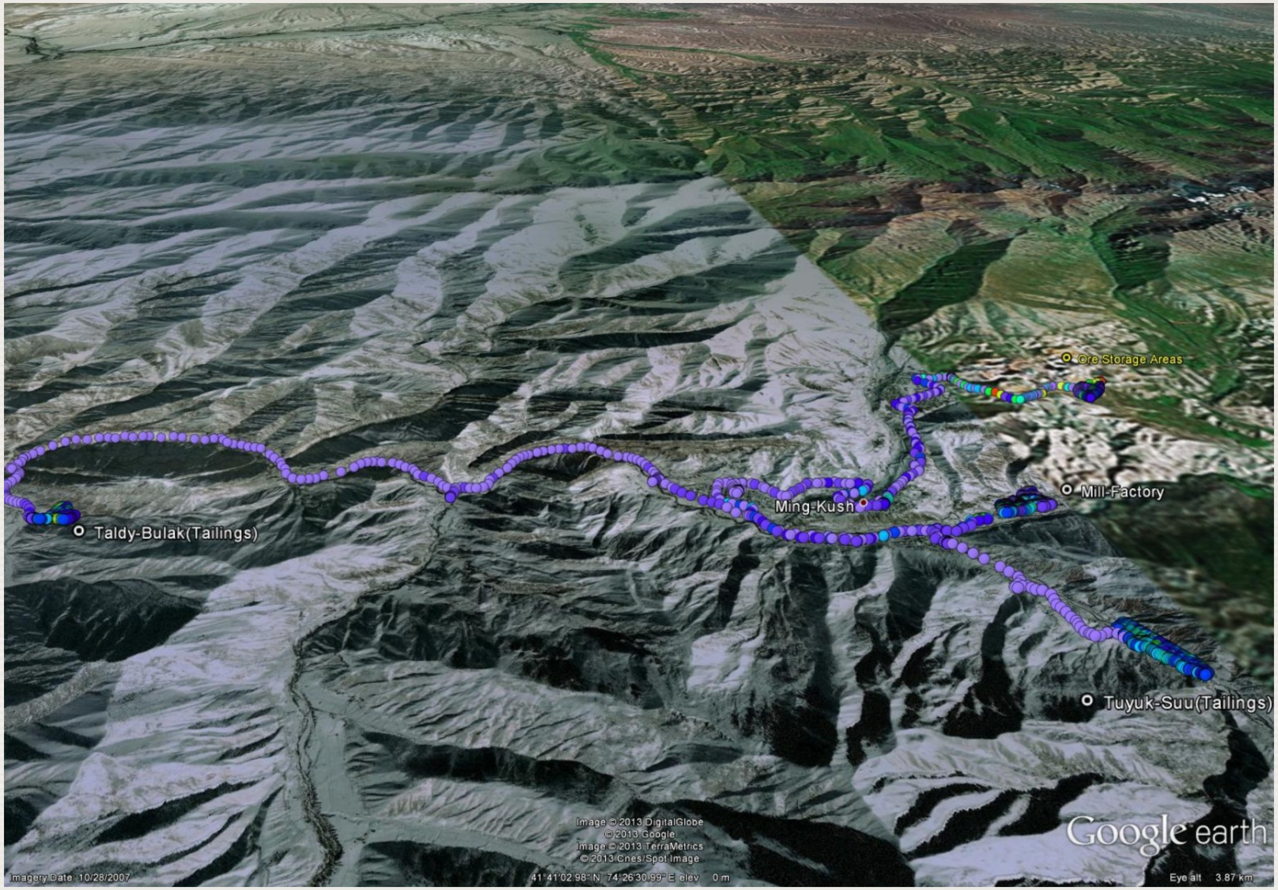
IAEA

Radiological characterization of sites
Expert mission to Azerbaijan (TC project AZB9005)

- Maps of surface DR for 3 areas (2 remediated sites, one disposal facility)
- 4 different instruments
- Total area surveyed: ~ 30 ha (16.04, 6.32 and 7.66)
- Total length of each pathway: ~ 15 km (5.4, 2.9, 6.5)
- 2 ½ days of measurements

NSIL / NSRW: Training on surface characterization of NORM affected sites

Kyrgyzstan (May, 2013), 21 ha



ENVIRONMENTAL MONITORING

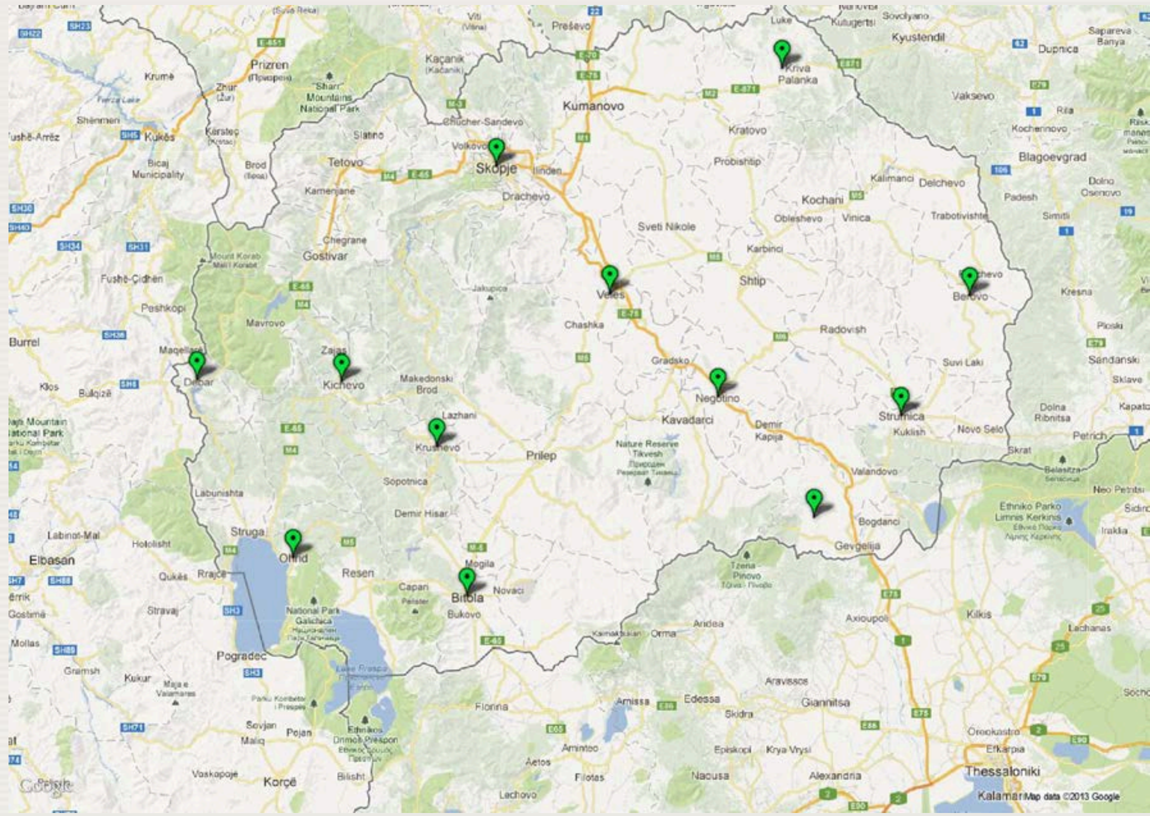
- Gamma dose rate
- Air
- Fallout
- Soil,
- Water
- Food (local and/or imported)

Courtesy of
Lidija Nikolovska
Public Health Institute
Skopje, Republic of Macedonia

ENVIRONMENTAL MONITORING

Macedonia

- Gamma dose rate
- 12 stations
- Air sampling
- 3 locations
- Fall-out
- 3 locations
- Rivers,lakes
- 6 locations
- Soil
- 3 locations



ENVIRONMENTAL MONITORING

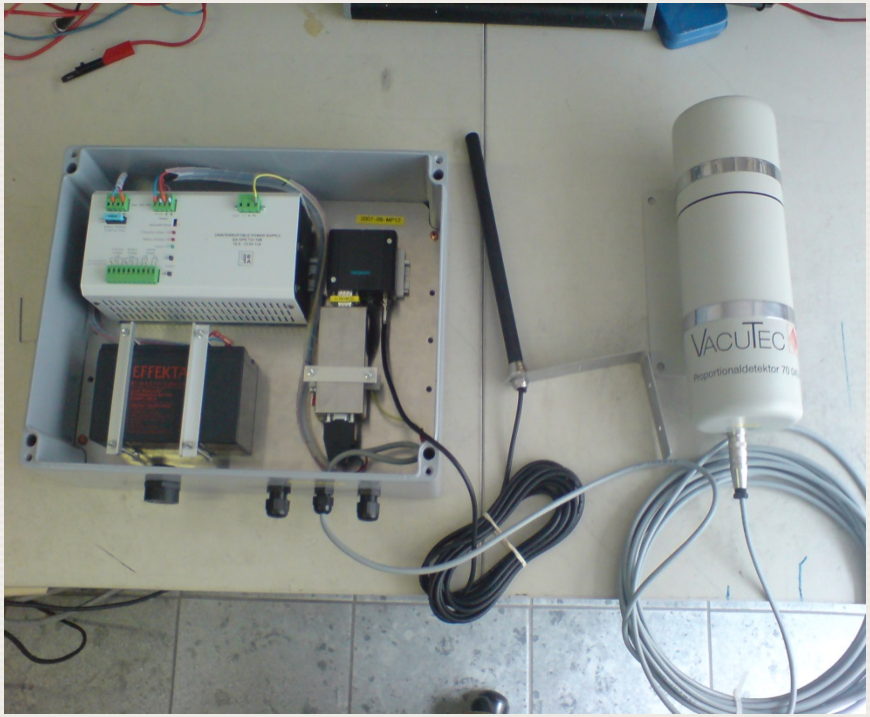
Gamma dose rate network

- 1. Measuring Units (12x):
  - Installed 2m above ground
  - Radiation Detector Vacutec 70045A
  - 12 V lead battery,
  - UPS,
  - GSM Modem MC35i+SIM Card
  - GSM antenna,
  - IP67 housing
- 2. Data center:
  - PC with MEVIS software
  - GSM modem connected to the PC via a COM/Serial Port

ENVIRONMENTAL MONITORING

Gamma dose rate network – Dose rate probes

- Radiation Detector Vacutec 70045A
- Proportional counter
- tube (430 cm3)
- Measuring value:
- ambient dose rate
- equivalent H*(10)
- 10 nSv/h-10 mSv/h
- Accuracy: ±5%
- Energy range:
- 30 keV ... 3 MeV



ENVIRONMENTAL MONITORING

Air sampling

- Weekly sample
-     Total alpha/beta activity

- Monthly sample
-     Gamma spectrometry

- six-months sample
-     (Sr-90)

V=600-900m^3/h

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ENVIRONMENTAL MONITORING

Analytical methods

- Total alpha and beta counting
- Gamma spectrometry
- Chemical separation (Sr-90)
- Radon counting (active and passive)

ENVIRONMENTAL MONITORING

Analytical methods

- Alpha/Beta counting

- Low background measuring system,
- EURYSIS C-P, four gas flow 2π detectors,
- Ar/Me gas mixture (90:10)

- Standard calibration sources
- Sr90, Am-241

ENVIRONMENTAL MONITORING

Analytical methods

- Gamma spectrometry

- Two Canberra systems
- - HPGe, Rel.Eff:25%
- - HPGe, Rel.Eff:21%
- - software: GENIE 2000

L.A.S.A. Lab, INFN and Physics Dept. of University of Milano

Courtesy of Alexandra Ioannidou
Aristotle University of Thessaloniki, Greece

S. Manenti, L. Gini and Flavia Groppi
L.A.S.A Lab., INFN and Universita degli Studi di Milano, Italy

Publications on Fukushima accident

- The 1st publication in *Journal of Environmental Radioactivity* about Fukushima accident was from our Nuclear Physics laboratory.
- A publication as collaboration of all Greek laboratories with GAEC published at *Radiation Meausrements* Journal.
- A combined publication between Milano and Thessaloniki lab there is in *JRNC*.
- A combined publication between Milano and Thessaloniki lab with air mass trajectories analysis published at Atmospheric Environm. journal.

Environmental Science & Technology
ARTICLE
pubs.acs.org/est

Tracking of Airborne Radionuclides from the Damaged Fukushima Dai-Ichi Nuclear Reactors by European Networks

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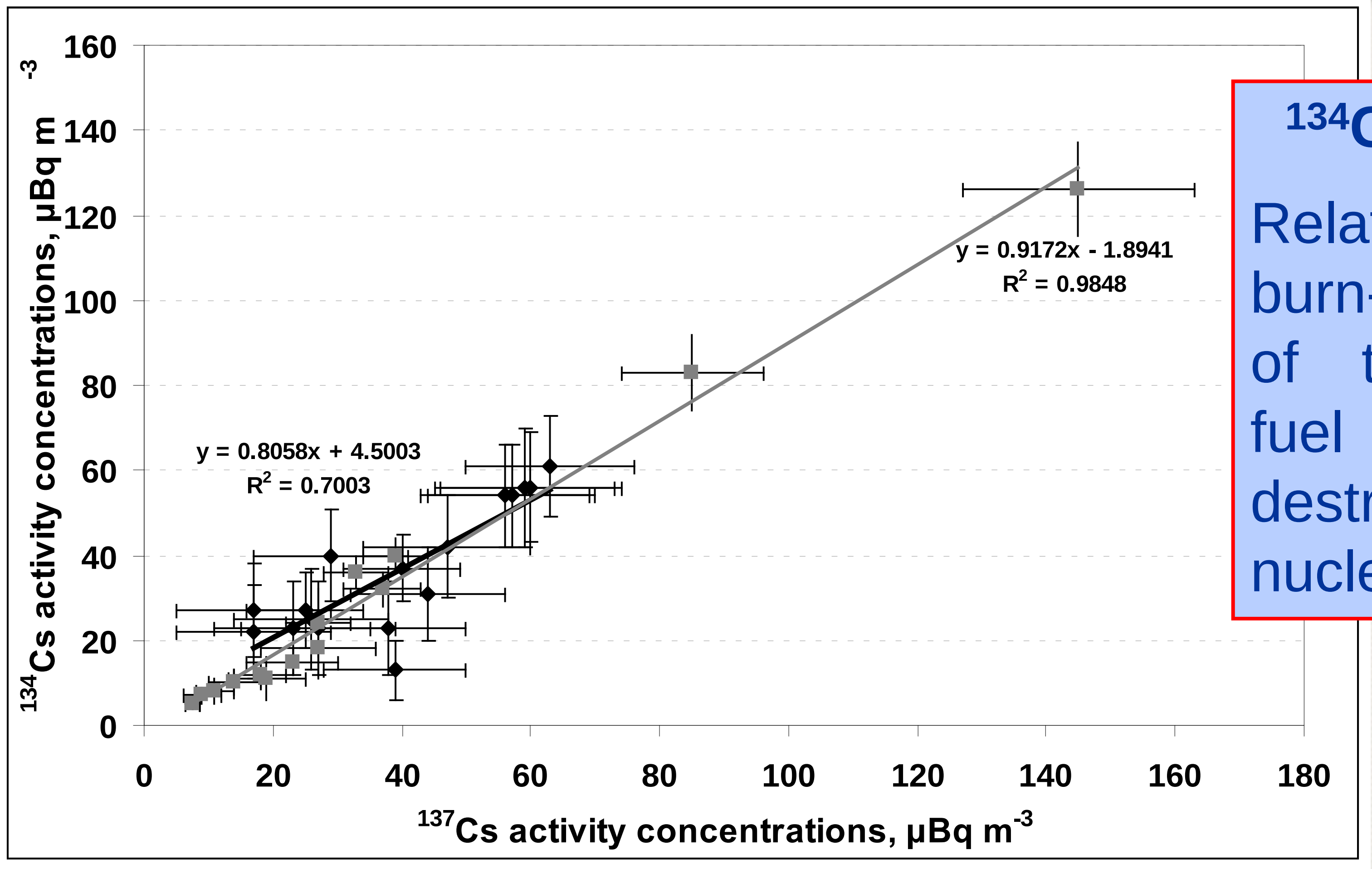
$^{134}Cs/^{137}Cs$ activity ratio = 1

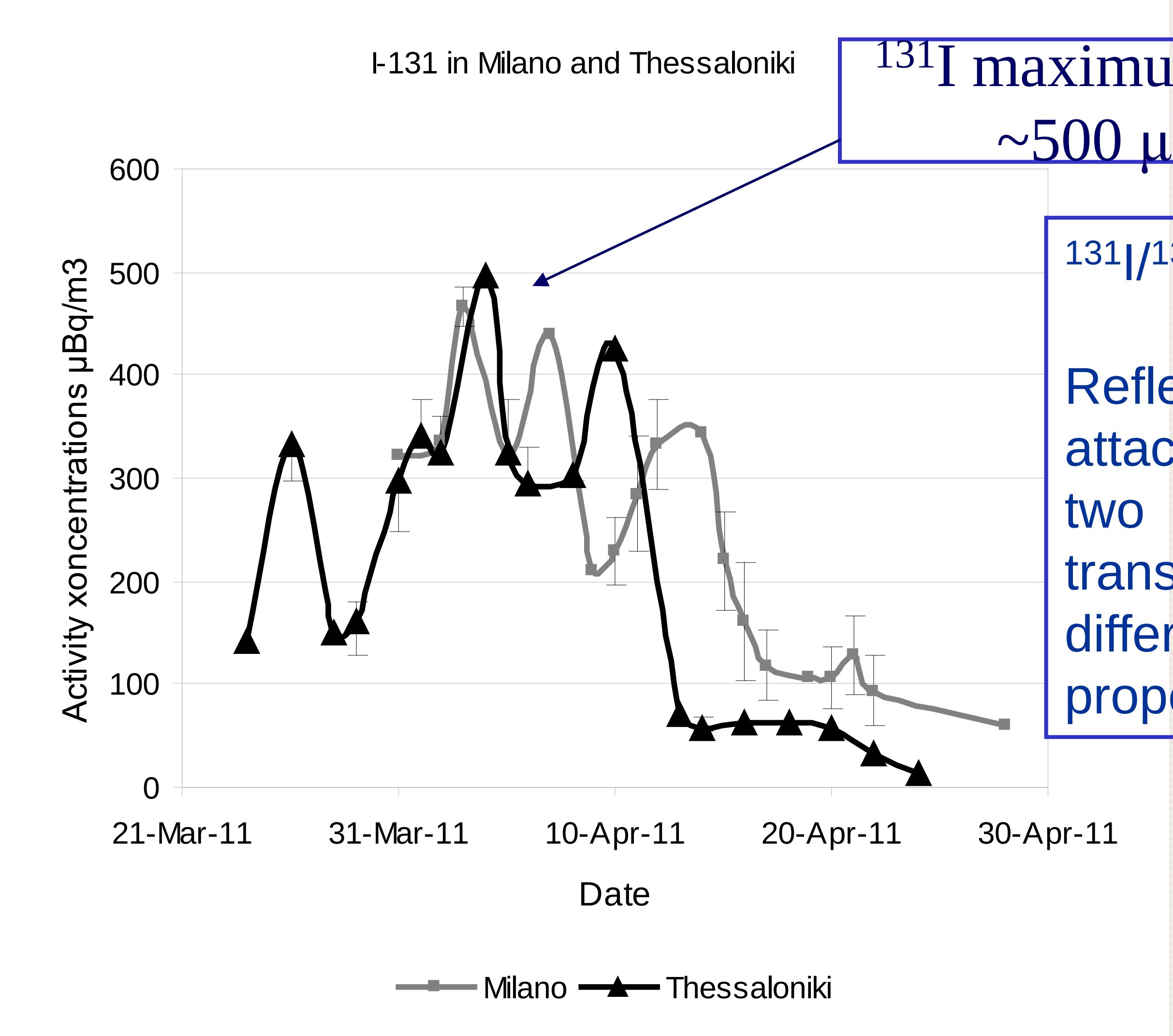


$^{134}Cs/^{137}Cs$ =1

Related to the burn-up history of the nuclear fuel of the destroyed nuclear reactor

^{134}Cs concentrations versus ^{137}Cs concentrations in Milan (45°), Italy (black dots) and Thessaloniki (40°), Greece (gray dots)

^{131}I atmospheric concentrations

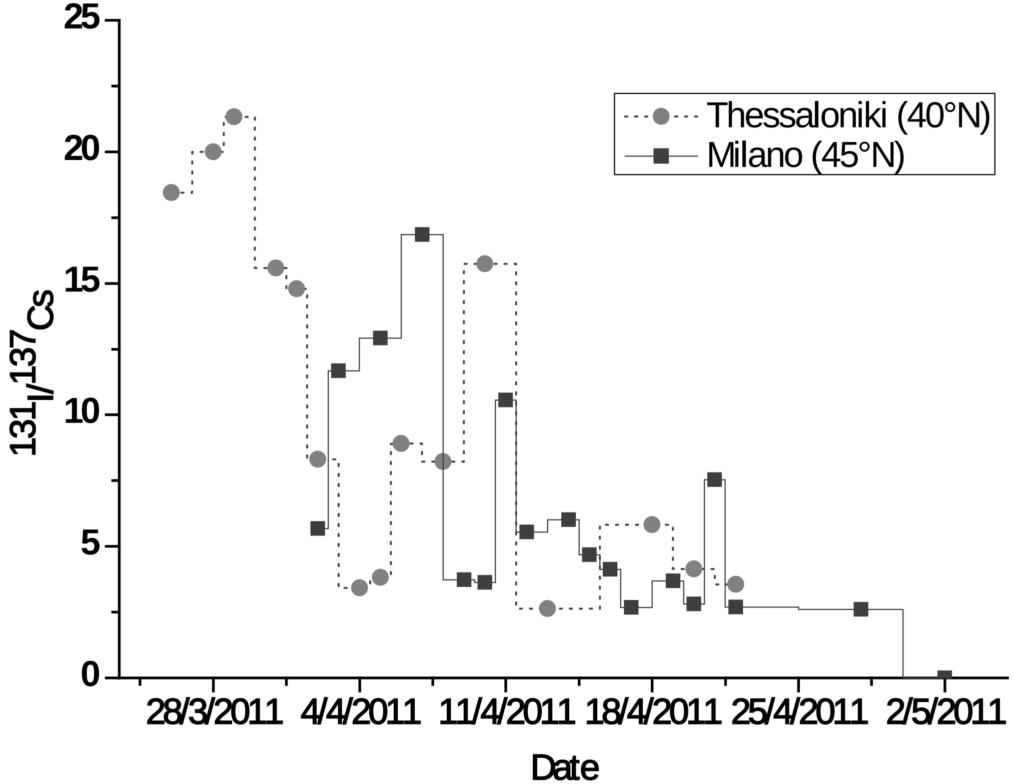


^{131}I maximum 3-4 April ~500 µBq m^{-3}

^{131}I/^{137}Cs decrease with time

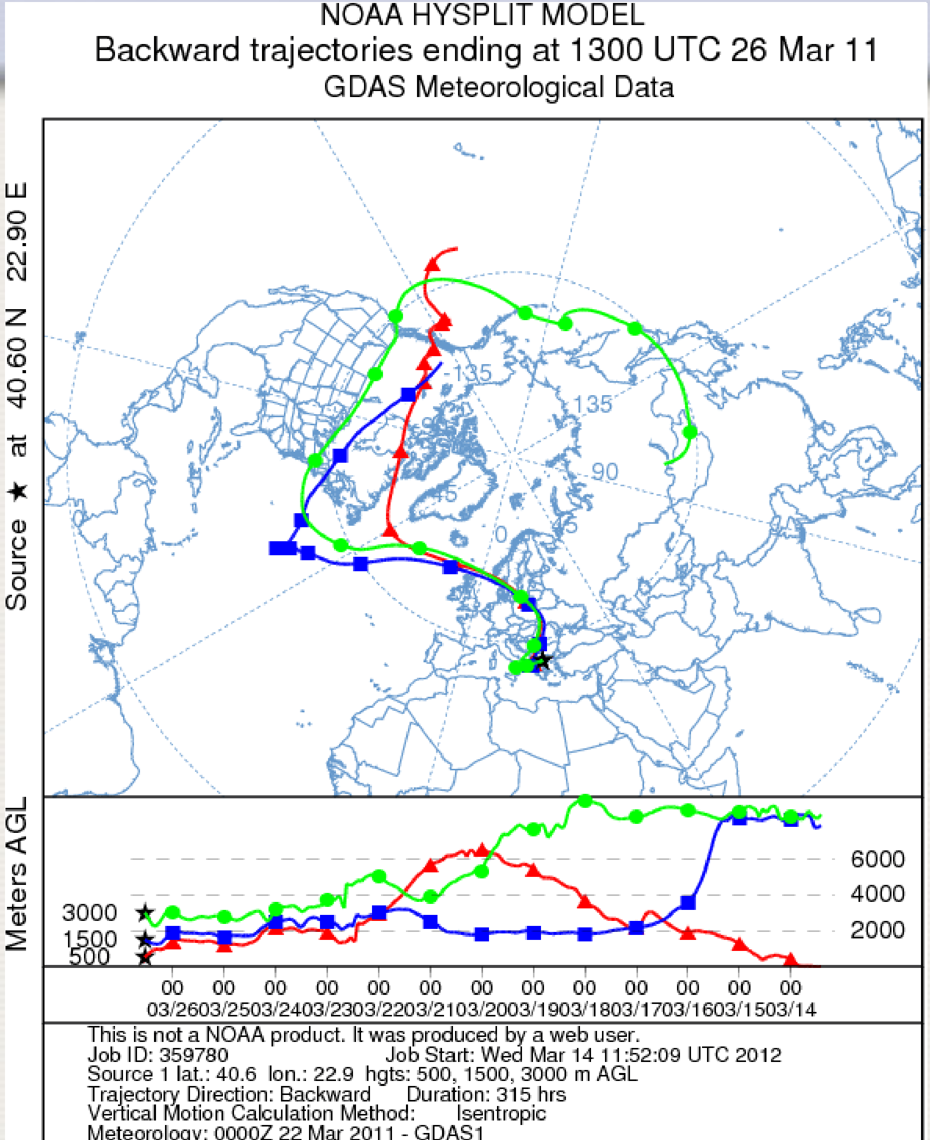
Reflects the different volatility, attachment and removal of the two isotopes during transportation due to their different physico-chemical properties.

$^{131}I/^{137}Cs$ atmospheric concentrations



$^{131}I/^{137}Cs$ activity ratio in Milan (45°) and Thessaloniki (40°)

Back trajectories analysis

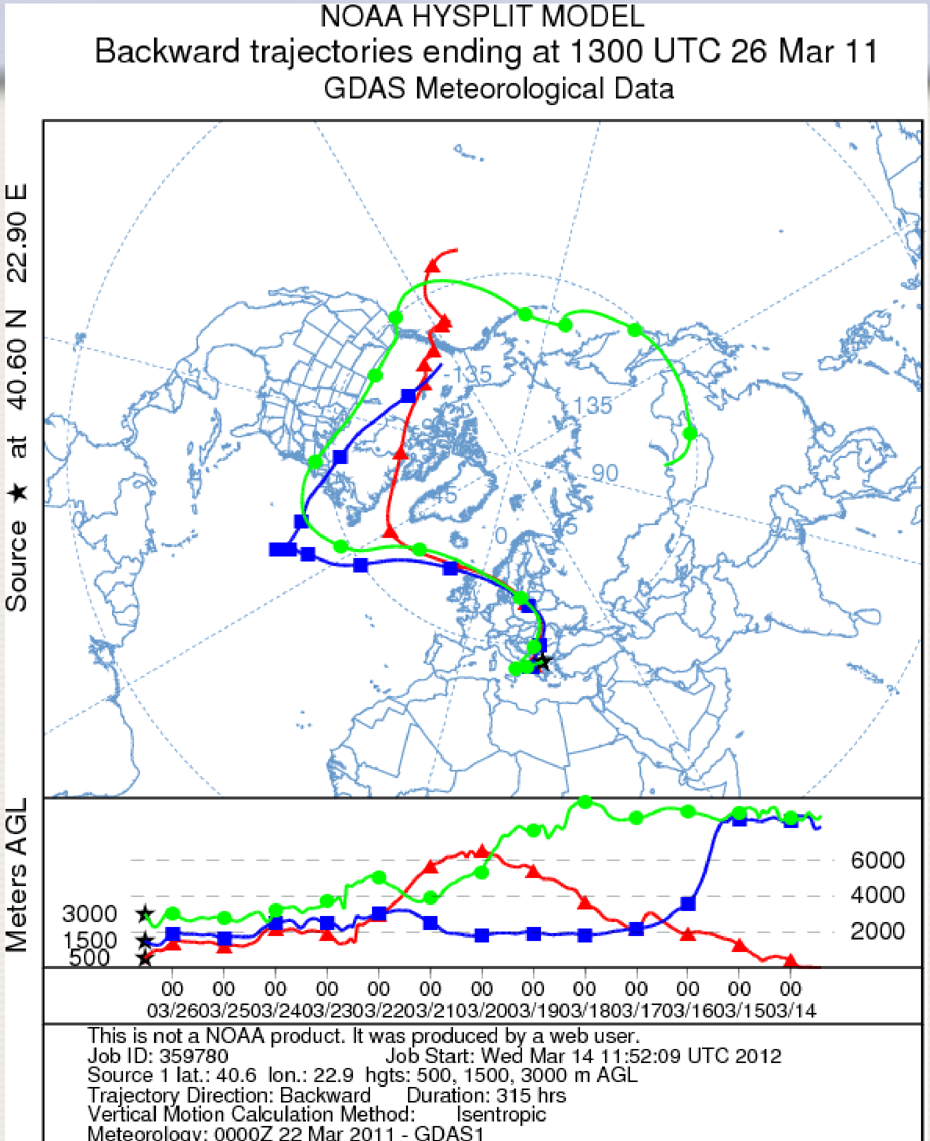


The NOAA HYSPLIT model was used to assess the transport pattern and to explain the deviation in radionuclide activity concentrations found.

Thirteen days (312) back trajectories were calculated for different arrival height and for 12 UTC time.

The trajectories are labeled every 24h by a filled symbol.

Back trajectories analysis



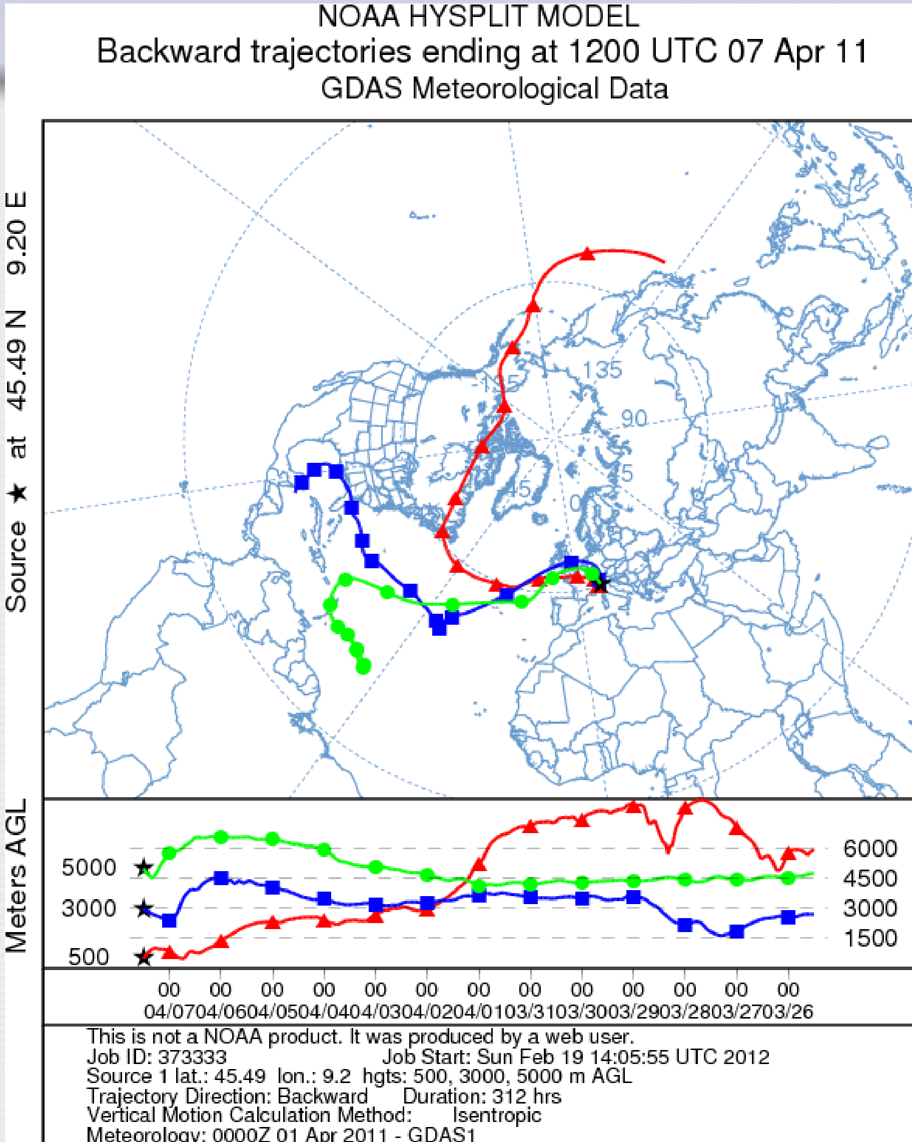
The first maximum in concentration of ^{131}I (332 µBq m^{-3}) was observed at Thessaloniki on **26th of March** 2011.

Air masses were lifted rapidly and transported over the North America to Europe at height of 500 m.

Air masses were also traveled at higher atmosphere levels from Japan. It is also possible that radioactive particles were transported at higher altitudes and may have been removed in the the lower layer of the atmosphere due to various reasons, e.g. rainfall characteristics, fog formation or growth of aerosol particles and their deposition.

IAEA

Back trajectories analysis

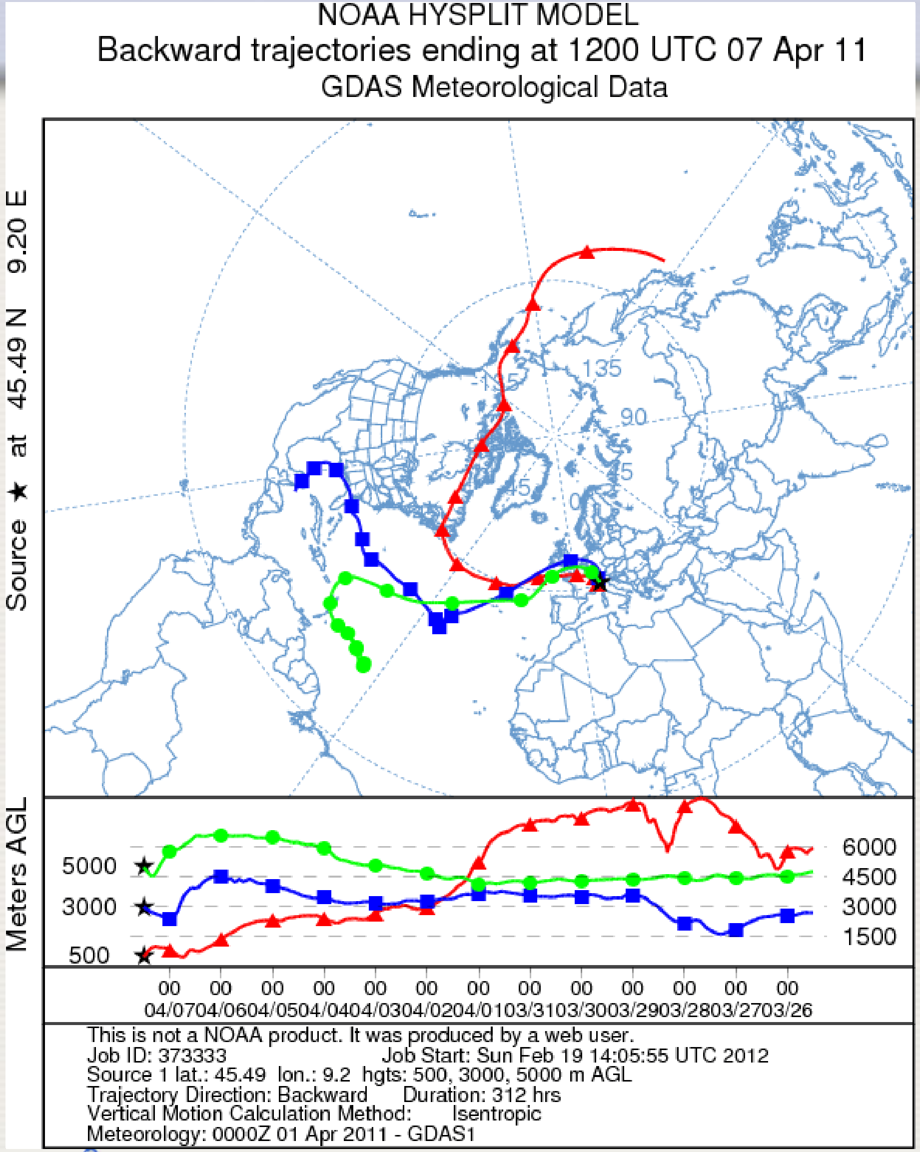


An example of transported air mass at **07 of April 2011** at Milano is presented.

The results showed a direct transfer from Fukushima across the Pacific Ocean, a transport through the North Pole and a pathway through the Greenland and Iceland at height of 500 m to Milano.

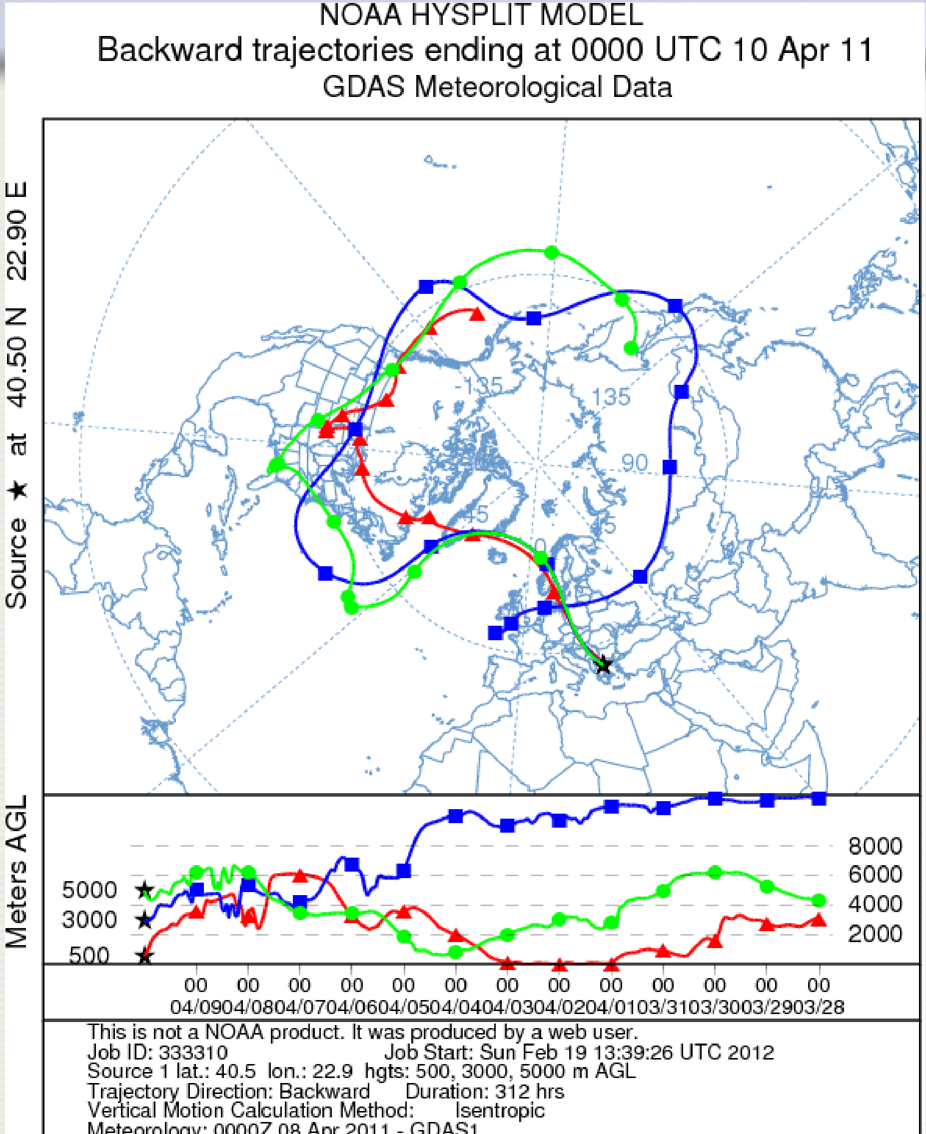
The air masses at higher altitudes were rapidly transported, while the air masses at 500 m exhibited rather slow transport.

Back trajectories analysis



Although the second maximum of concentration of ^{131}I that observed at Milan on 07th of April can be attributed to the advection of air masses from Japan at altitude of 500 m, however, the back-trajectory analysis for the same day at Thessaloniki indicates no transport of air masses from Japan, at least for height of 500 and 1500 m.

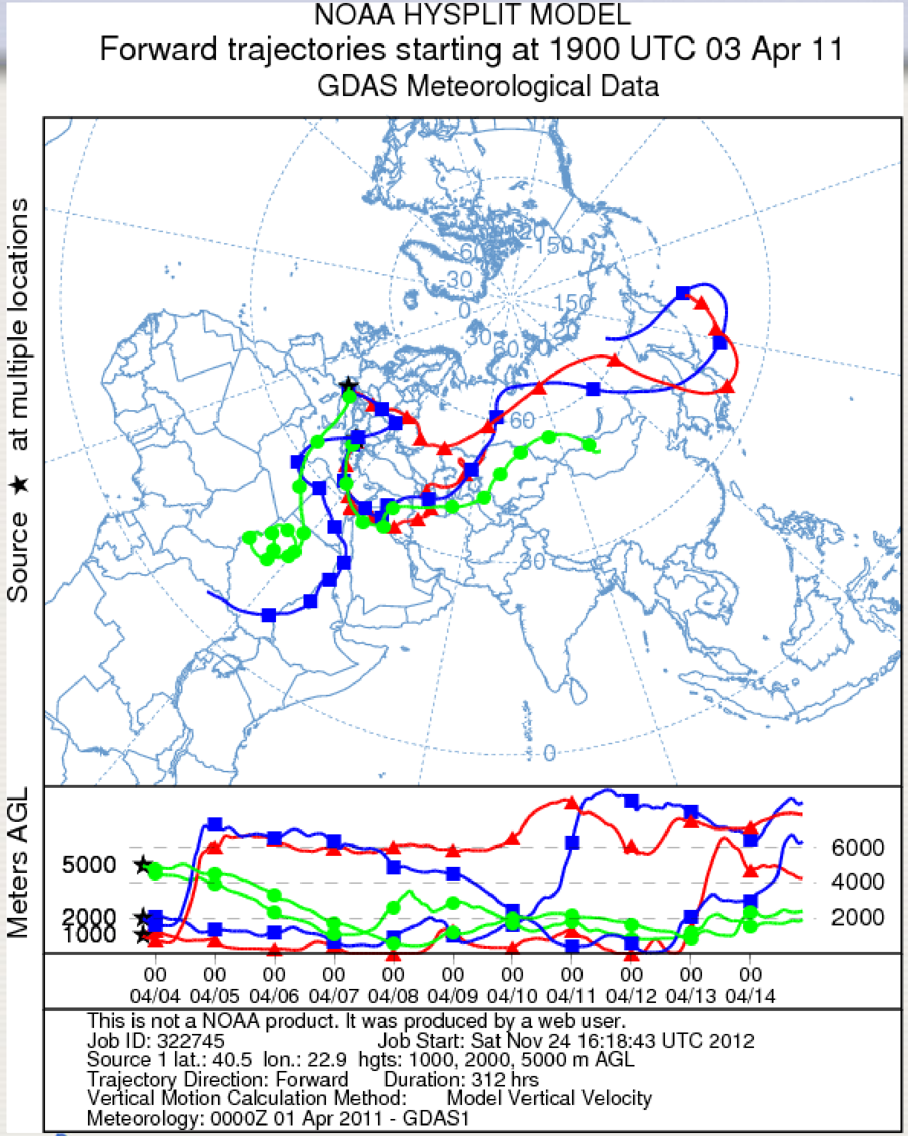
Back trajectories analysis



On **10th of April** back-trajectory analysis showed a direct transfer from Fukushima across the Pacific Ocean, a transport through the North Pole and a pathway through the Greenland and Iceland to Thessaloniki.

The air masses on that day reach Thessaloniki from Northwest direction and this is possible the reason why no maximum concentration was observed at Milan.

Back trajectories analysis

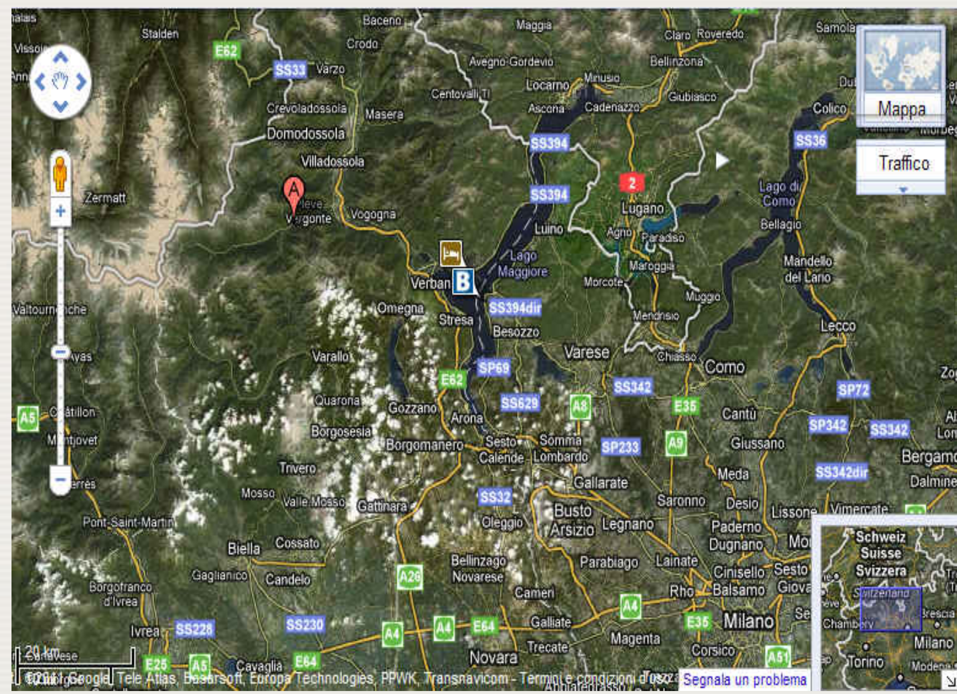


On April 3, 2011 at 19:00 UTC the back-trajectory analysis indicates the transport of air masses from Japan both in Italy and in Greece but at different arrival heights. As Fig. 8 shows the air masses started above Japan at around 2 km. In Italy the air mass moved down, travelled near the ground and arrived above Italy at height of 1 km. On the other hand, the air mass after being near the ground for almost 2 days rose to arrive at height of 2 km above Thessaloniki. Both transport pathways can explain the maximum concentrations that were observed at the regions of study.

^{131}I, ^{137}Cs and ^{134}Cs in milk

Samples of sheeps and cows milk

collected in Val Anzasca (VB), Italy at 400 m s.l.m.



^{131}I, ^{137}Cs and ^{134}Cs in milk

Fallout isotopes in milk samples

Date of Sampling	Goat Milk				Cow Milk			
	^{131}I mBq L^{-1}	^{137}Cs mBq L^{-1}	^{134}Cs mBq L^{-1}	Ratio ^{134}Cs/^{137}Cs	^{131}I mBq L^{-1}	^{137}Cs mBq L^{-1}	^{134}Cs mBq L^{-1}	Ratio ^{134}Cs/^{137}Cs
9/04/11	246±107	481±52	< 33[a]	-	208± 97	333±44	< 31[a]	
1/05/11	101±68	506±48	< 26[a]		[b]68±67	421±44	< 31[a]	
8/05/11	87±72	448±47	< 26[a]		< 40[a]	263±39	< 26[a]	
16/05/11	< 24[a]	526 ±50	< 30[a]		< 38[a]	302±47	67±35	0.22
21/05/11	[b]77±73	527±63	[b]59±44	0.11	< 53[a]	684±54	< 28[a]	
29/05/11	60 ±46	474±47	69 ±26	0.15	110±58	473±44	< 27[a]	
05/06/11	< 25[a]	398±44	< 33[a]		< 34	354±41	< 27[a]	
11/06/11	< 68[a]	378±55	< 34[a]		77±68	279±37	41±24	0.15
20/06/11	< 32[a]	298±37	< 22[a]		< 28[a]	197 ±35	< 22[a]	
26/06/11	< 29[a]	460±45	< 25[a]		81±60	283±64	< 34[a]	
03/07/11	< 28	796±67	[b]48±30		< 32 [A)]	296±36	< 0.23	

a. MDA
b. Critical level

I-131 1200±350 mBq L^{-1}

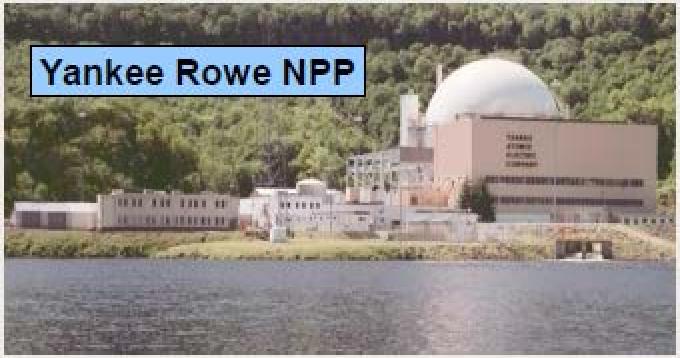
Cs-137 150±30 mBq L^{-1}

CONCLUSIONS

- The Fukushima plume was detected all over Europe
- The presence of more than one peaks of ^{131}I and $^{137,134}Cs$ is an index that air masses continuously transferred from Fukushima, Japan till the end of April, 2011.
- HYSPLIT backward trajectories interpreted the measured atmospheric concentrations
- The relative high concentrations of ^{137}Cs in grass, soil and fresh milk samples, correspond to Chernobyl fallout
- ^{131}I and $^{137,134}Cs$ isotopes were found above their detection limits in all environmental samples but very far below levels of concern

Decomissioning

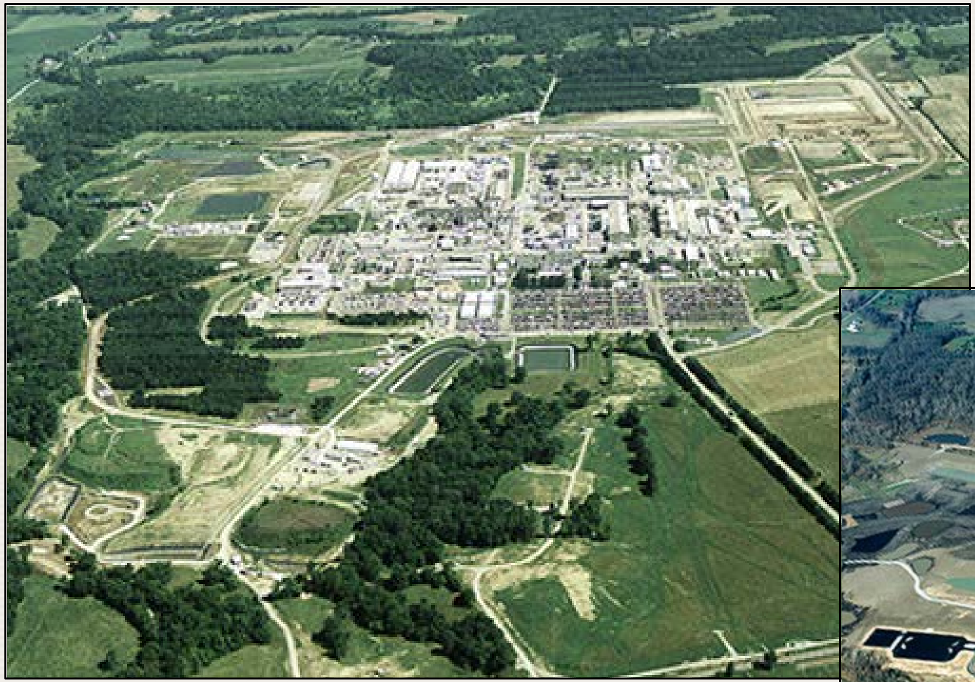
Maine Yankee being dismantled, USA



Disposal of low level waste

Final Covering of the Low Level Waste
Disposal Facility at Centre de la Manche (France)

Fernald Closure Project

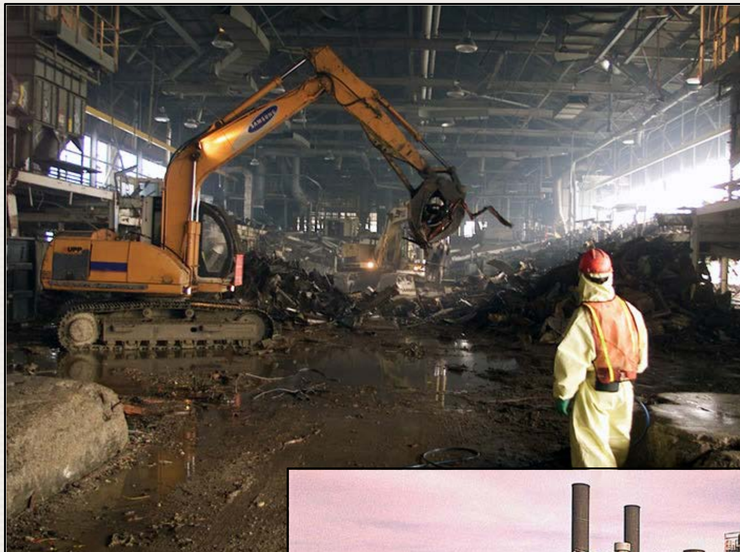


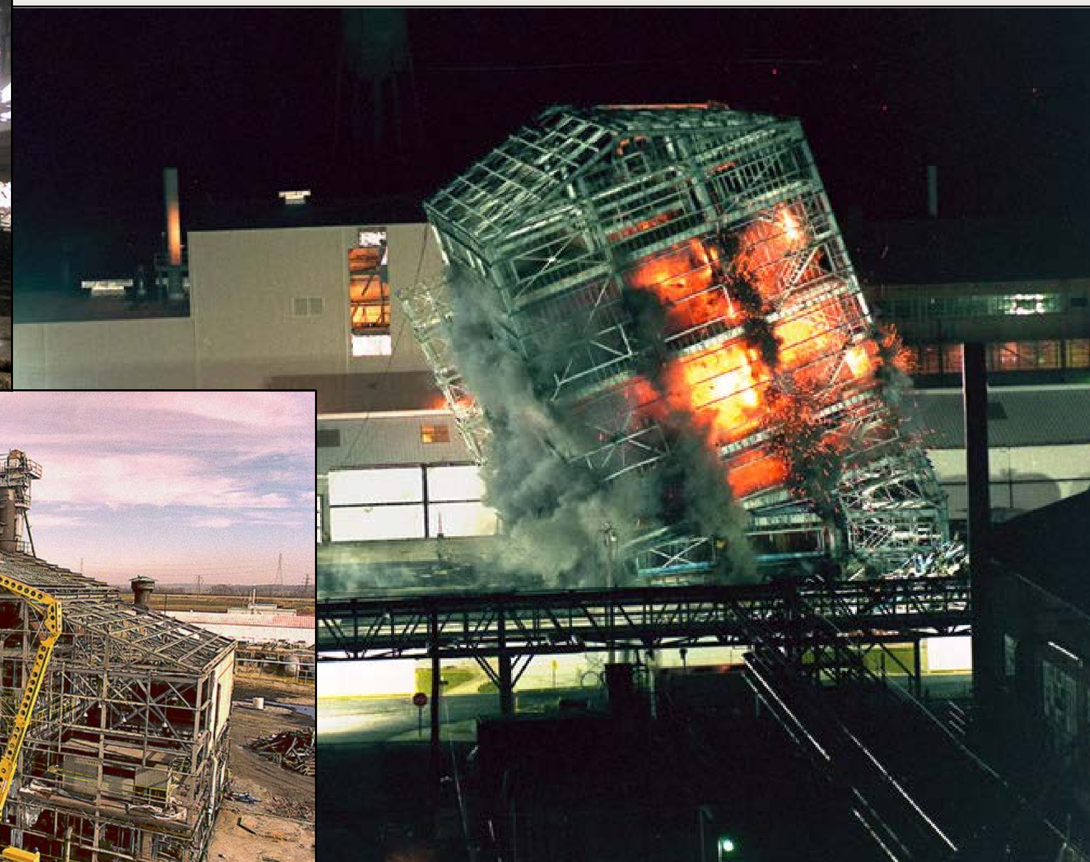
Courtesy of
Gene Jablonowksi
US EPA

Fernald Closure Project

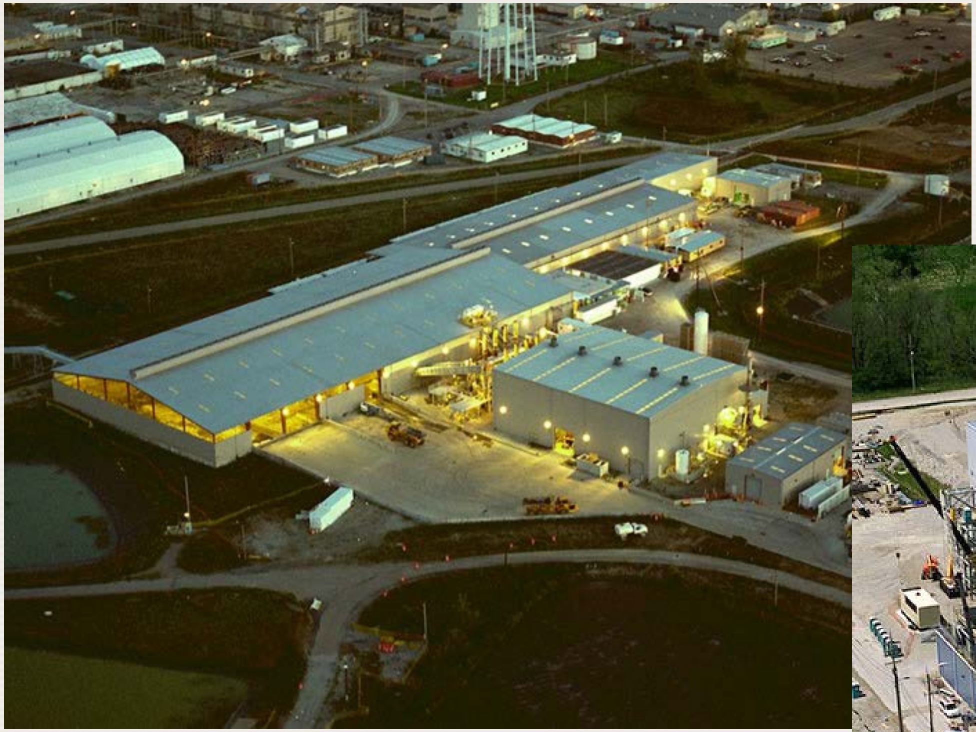
- Former uranium processing and metal products facility
- Operated from 1952 to 1989, then shifted to waste management and remediation
- 4.2 km² (1050 acres) site; 0.5 km² production facility
- Remediation included:
  - Production area D&D of over 200 buildings and structures
  - low level waste in pits and silos
  - soil/debris to be excavated
  - low enrichment uranium in inventory
  - drums of waste in inventory
- Primary contaminants: uranium, radium, thorium
- Site closure completed in October 2006

Fernald Closure Project



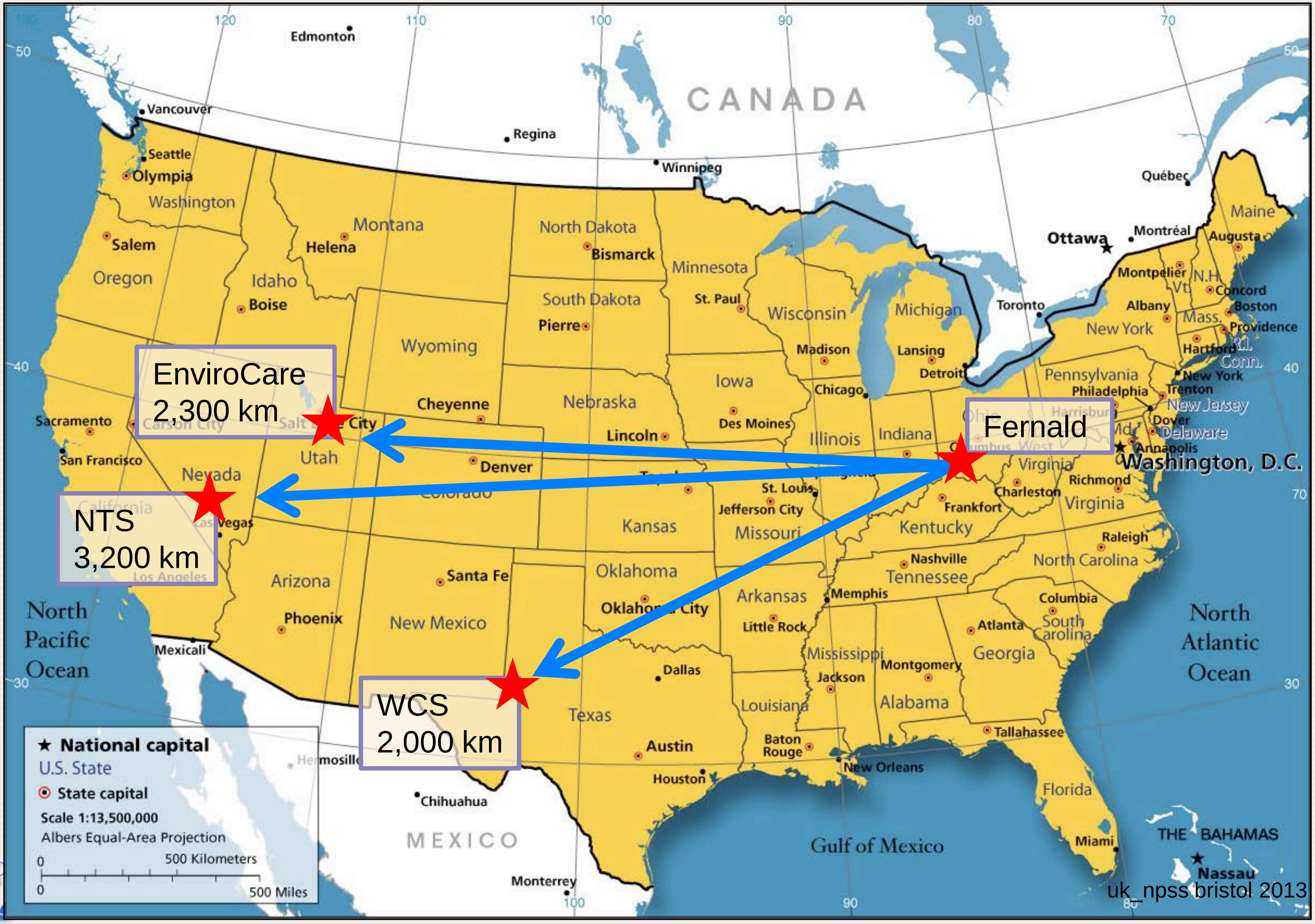


Fernald Closure Project



Fernald Closure Project

Fernald Closure Project



Fernald Real Time Program

- One of the first DOE sites to develop real-time geospatial measurement systems to support remediation
- Soil characterization systems typically comprised of:
  - Global Positioning System (GPS) receiver to determine GPS coordinates of each measurement; includes differential GPS (DGPS) for sub-meter accuracy
  - In-situ gamma spectroscopy equipment for spectrum collection
  - Laptop PC with software that links spectrum information with detector GPS position, and provides a data display for the operator
  - Wireless Ethernet link to computer running Surfer GIS software for real-time map and report production
- “Off-the-shelf” equipment used
- LabView used as system control software

Fernald Real Time Program
Strategy

- Sodium iodide (NaI) detectors on vehicles and carts for rapid 100% coverage of large areas:
  - Scan large flat areas for general contamination patterns
  - Define general excavation boundaries; excavation control
  - Finding hot spots above FRL; areas above OSDF WAC
  - 100% area scans of excavated areas after removal of lifts
  - ~10 hours/hect at 1.6 km/hour with a 0.4 meter overlap/transect
- HPGe for static measurements for accurate quantification:
  - Defining nature and extent of contamination
  - Refine general excavation boundaries
  - Delineation of "hot spot" boundaries
  - Pre-verification of contaminant removal to Final Remediation Levels (FRLs)
  - 65 HPGe shots / 0.6 hect / day using a 5-min count times and 3 units

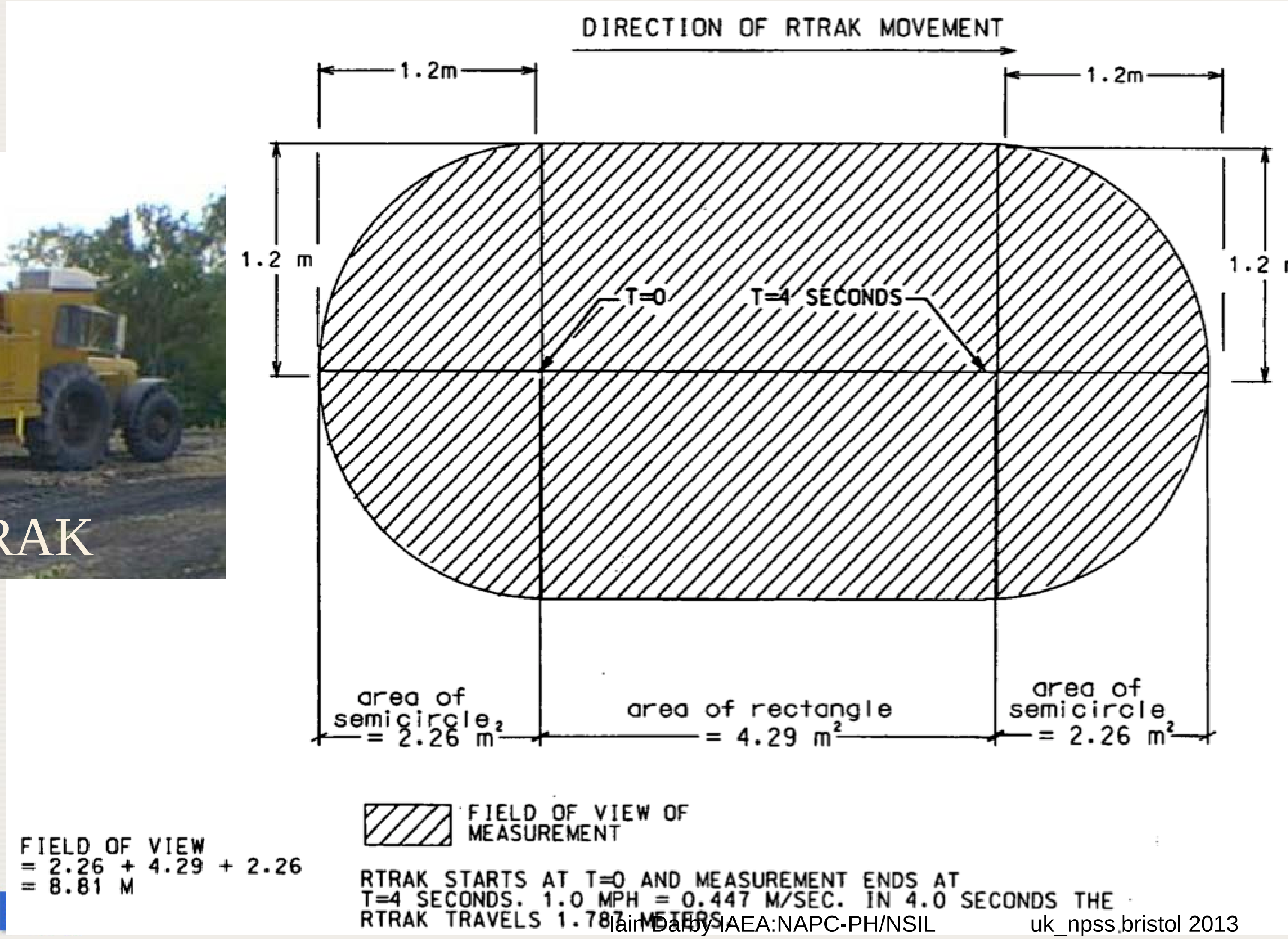
Fernald Real Time Program
NaI Detectors

Sodium iodide (NaI) detectors:

- Uncollimated / unshielded
- 10x10x40-cm (4L) crystals hermetically sealed in aluminum housing
- Crystal/PMT wrapped with non-static polypropylene material for thermal insulation and physical shock protection
- Encased in a polyvinyl chloride (PVC) tube or aluminum enclosure.
- Mounted 31 cm above ground

Fernald Real Time Program

NaI Field of View Example



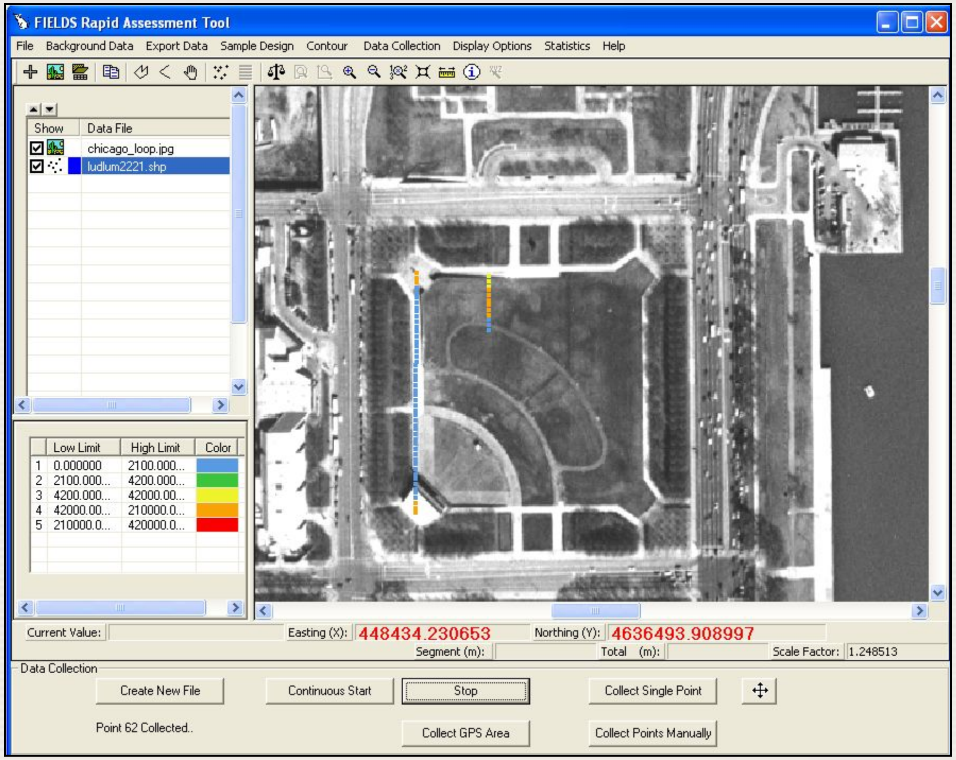
Fernald Real Time Program

More Information

- DOE User Guidelines, Measurement Strategies, and Operational Factors for Deployment of In-Situ Gamma Spectrometry at the Fernald Site, Fernald Area Office, Jan. 2004
  - http://www.lm.doe.gov/cercla/documents/fernald_docs/PROD/468429.pdf
- Measurement Uncertainties and Minimum Detectable Concentrations for the In-Situ NaI Gamma Spectroscopy Systems Used at Fernald, Argonne National Lab, June 2004
  - http://www.ipd.anl.gov/anlpubs/2004/07/50588.pdf
- Real-Time Measurement of Radionuclides in Soil: Technology and Case Studies, Interstate Technology & Regulatory Council Radionuclides Team, February 2006
  - http://www.itrcweb.org/Guidance/GetDocument?documentID=74

Field Analysis and Sampling Tools (FAST)

- Developed in-house
- Data is collected & captured “hands-off”
- Uses multiple devices (Radiation, Air, Soil ….)
  - Manual, Single, and Continuous Point Collection
- Real-time spatial visualization
- Immediate data storage and GIS file creation
- Data export



Discussion and/or presentation of products is for informational purposes only and not an endorsement.

FAST System Components

GPS

Trimble

Leica

Magellan

Garmin

GPS (NMEA) and external devices are linked to a laptop via Serial I/O ports

Heavy Metals

Dose and Count rate data

Chemicals

Air Particulates

- Any GPS that sends a standard NMEA string can be used depending on accuracy required. This gives you real-time locations requiring no post processing.
- Any sensor device both analog and digital can be incorporated into the software. Up to 256 sensors with GPS can be configured at one time.

Discussion and/or presentation of products is for informational purposes only and not an endorsement.

FAST Deployment Options

Mobile

Push Cart System

- Ability to run multiple devices
- Carry batteries for WIFI, GPS, and devices

Personal

Backpack system

- Single device
- Move through tough terrain

Motorized

Kawasaki 4X4 Mule

- Ability to carry multiple devices
- Power plug-in
- Carry multiple people

FAST - Current Devices

Radiation Devices:

Ludlum 2221

Ludlum 2241

Ludlum 2350

Fluke/Victoreen 451P & 451B

Air Monitoring Devices:

MultiRAE

Over 14 sensors available from RAE Systems

(CO, H_2S, VOC's, O_2, LEL, etc.)

Data RAM

Particulates, Temperature, Humidity, etc.

Draeger Multiwarn II

(CO, H2S, VOCs, Toxics)

Soil Monitoring Devices

Niton XLi, XLp, XLt

Innov-X 4000a-s

Over 23 different Metals

Additional devices can be added

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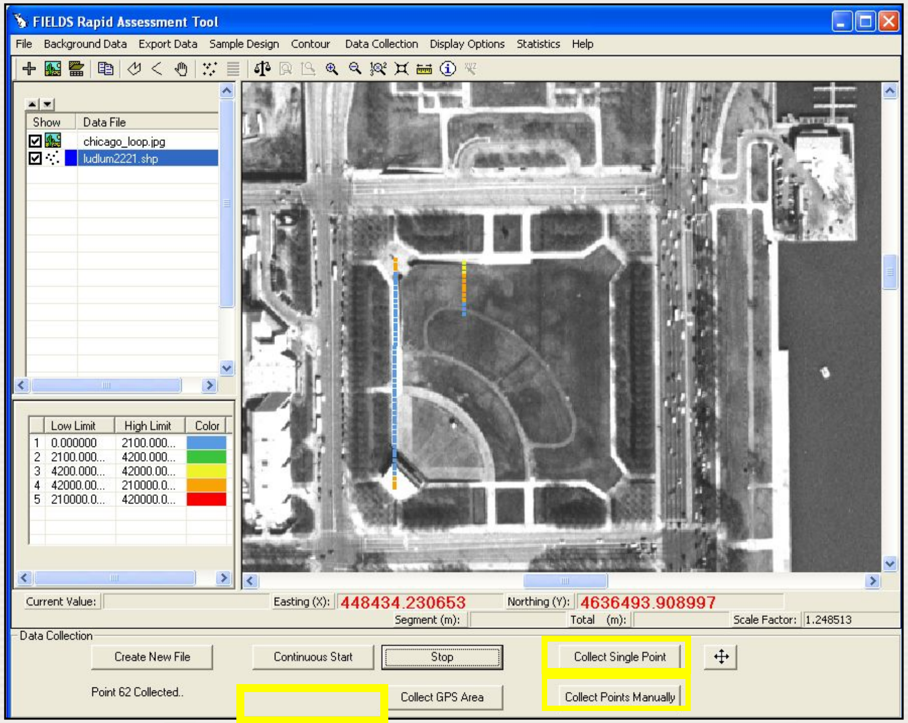
Collecting Data with FAST

Continuous Collection (mapping)



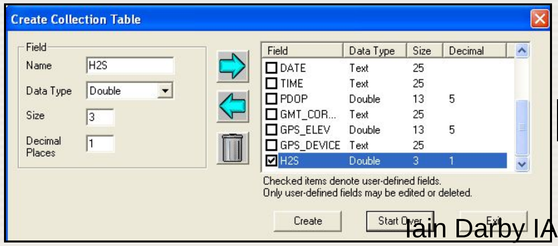

Single Point Collection

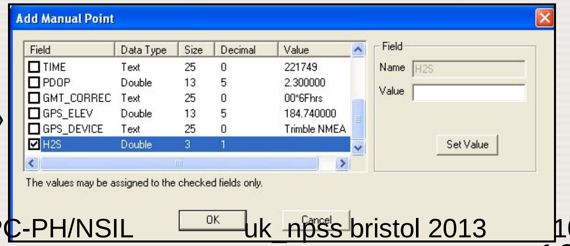




Manual Collection

- No electronic input or not integrated
- An electronic data dictionary can be created



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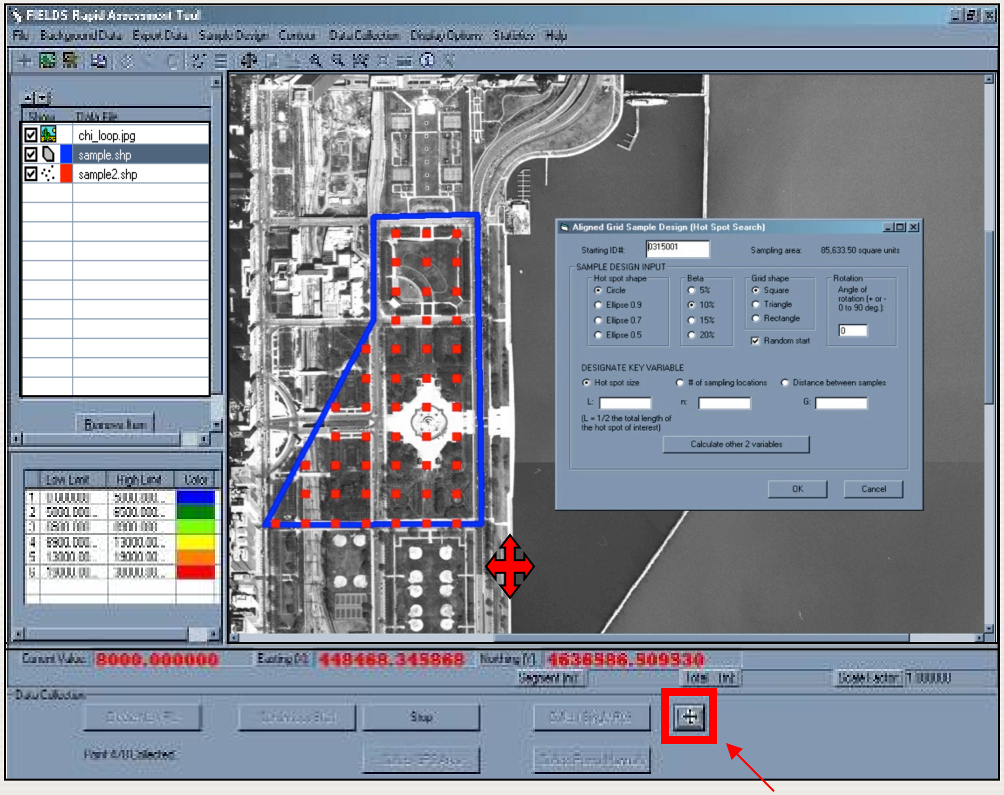
FAST Sample Design

Sample design tools allow instantaneous sample plan development in the field. A navigation tool can then be used to locate sample locations using the GPS.

- How many samples do I take?
- Where should I take samples?

Built in Sample Designs:

- Judgmental
- Random
- Aligned Grid (Hot Spot)
- Unaligned Grid



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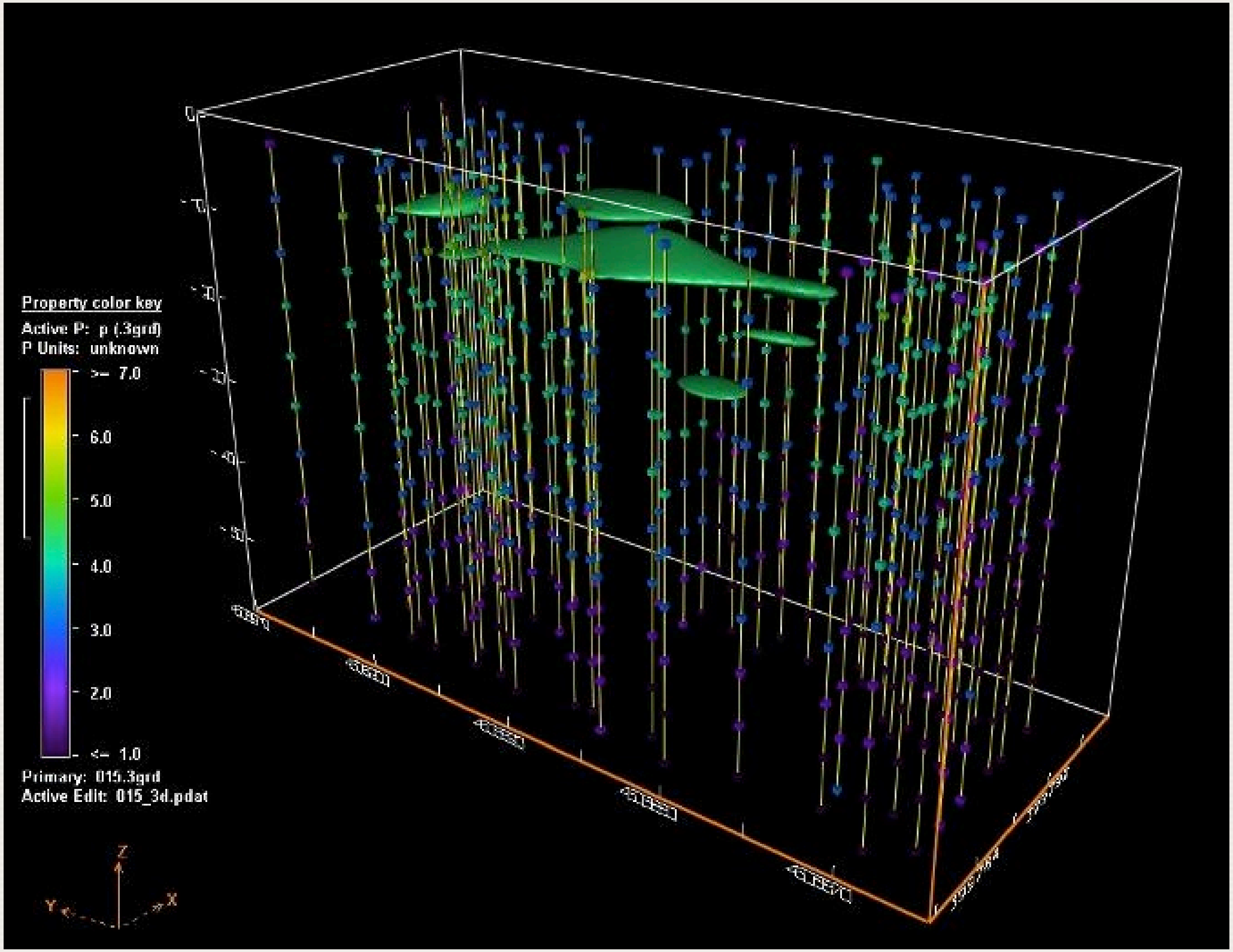
Down-hole Gamma Radiation
3D Model

Data is shown in pico-Curies/gram (pCi/g)

Contours in green represent pCi/g greater than four



GEM ground-based survey with ArcGIS to produce map product. Used to assess variability in urban background.

Total Gamma (cps)

- 0 - 400
- 401 - 600
- 601 - 800
- 801 - 1,000
- 1,001 - 1,200
- 1,201 - 1,400
- > 1,400

Note: the legend colors selected for "Total Gamma" are for display purposes only, and do not represent or imply exceedances of health-based levels.

0 150 300 600 900 1,200 1,500 Feet

Urban background can vary considerably due to historic materials such as granite pavers that may still reside in the shallow subsurface.

Survey Grade GPS with Robotic Laser Total Station

- Survey Grade GPS cm. accuracy x,y,z
- Total Station can be used in heavy canopy, indoors, urban areas
- 360 degree target prism can be moving with continuous data collection

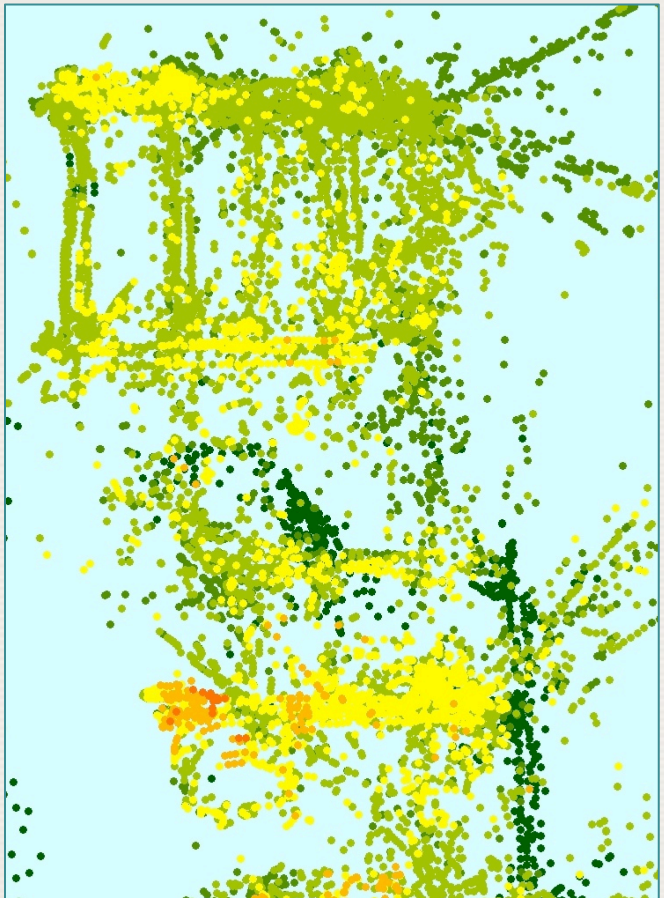
Survey Grade GPS with Robotic Laser Total Station with FAST

- Leica Total Station with integrated GNSS receiver
- Infared laser distance with 360 degree prism
    - 7,000 ft. distance single point
    - 1,000 ft. continuous point
    - Continuous accuracy: 3 mm + 1.5 ppm (typically <0.15 s)
- Set-up on a surveyed position (x,y,z)
- TS fully robotic, auto targeting, and self tracking
    - Single point or continuous data collection
- Integrated with FIELDS FAST software
    - Psuedo NMEA
        - X,Y,Z
        - Serial or bluetooth from data logger to FAST computer
            - Date & time from computer
    - Integrate sampling equipment through FAST
        - 900 Mhz radio modems

Discussion and/or presentation of products is for informational purposes only and not an endorsement.


Panasonic

Common GPS vs. Correction with Robotic Laser Total Station



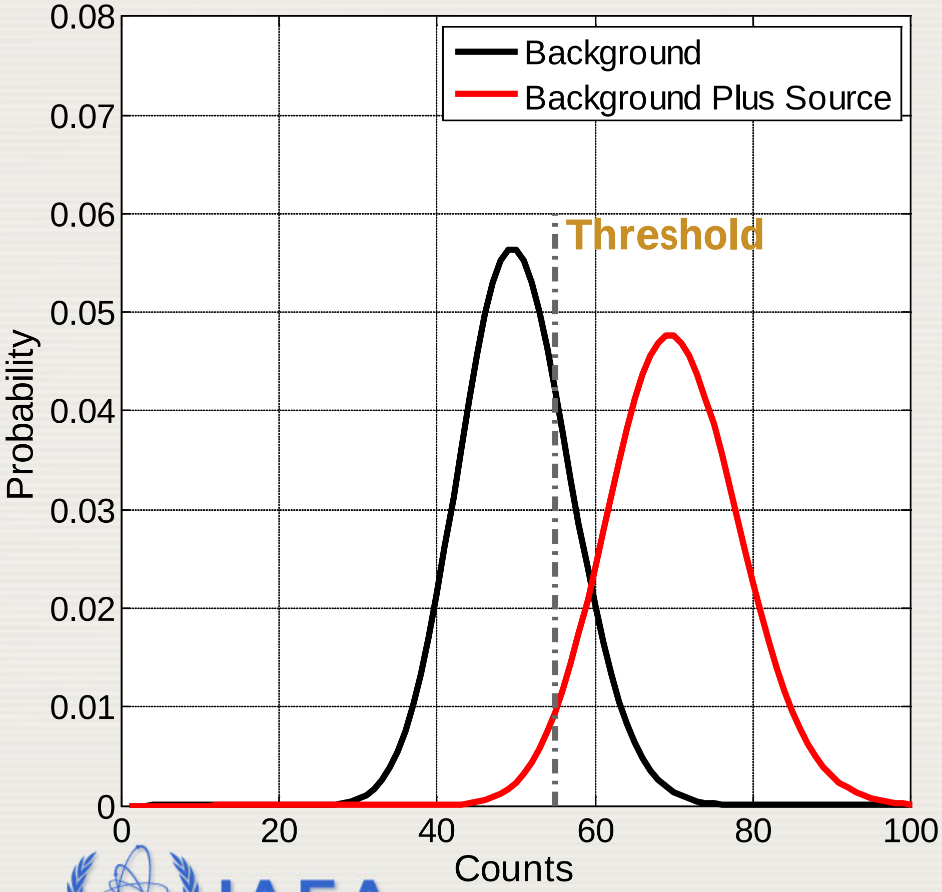
Metal high-rise buildings reduced GPS accuracy

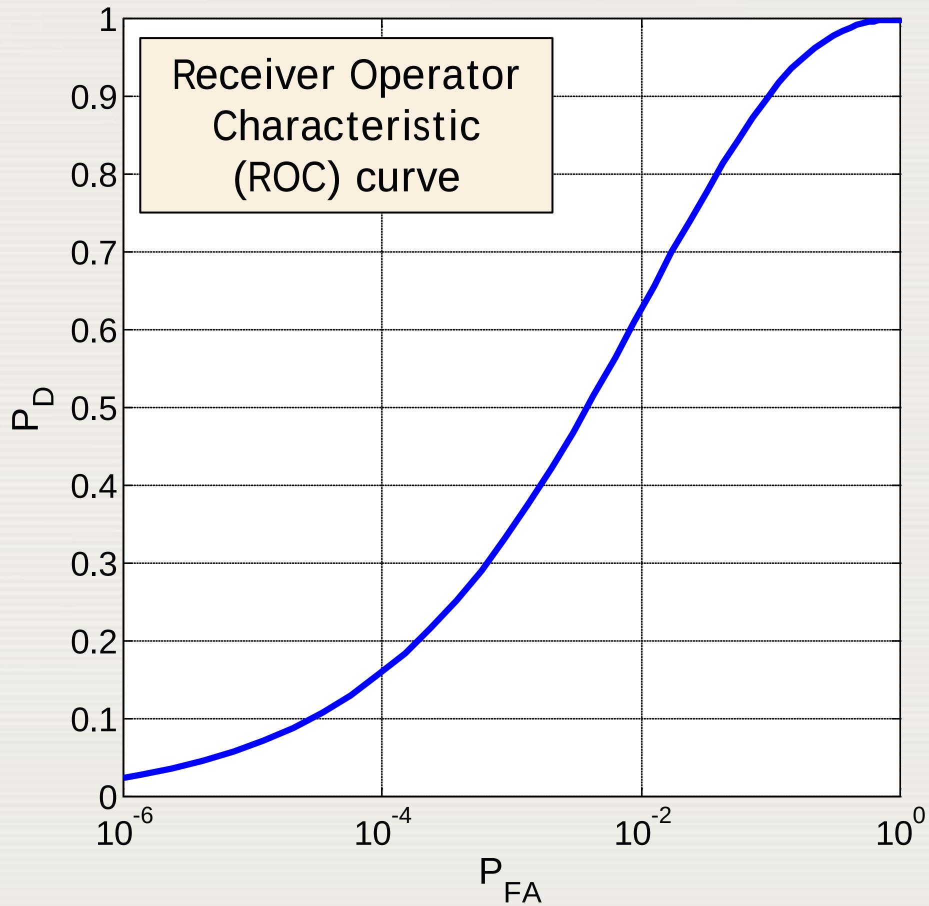
Location corrected with Total Station based on GPS time

Source Detection: A Signal-to-Noise Problem

- Detection of weak sources in the presence of background
- Probability of Detection (P_D) vs. Probability of False Alarm (P_{FA})

Courtesy of Ren Cooper LBNL





Minimum Detectable Activity (MDA)

- Common measure of **detection sensitivity**

- Officially, according to the IAEA (No. RS-G-1.2):

"…the level of activity which is needed to ensure, with some chosen level of confidence…, that the net signal will be detected, according to the criterion that it exceed the MSA."

where

"The minimum significant activity (MSA), often termed the critical level (L_C), corresponds to the smallest signal significantly in excess of the background response for the specific measurement method."

- Or:

The smallest activity you can distinguish from background at some fixed value of P_{FA} and P_D

Minimum Detectable Activity (MDA)

- Consider a system governed by Poisson statistics
- The minimum number of counts required for a source to be distinguished from the background is know as the Critical Level, L_C.
- For a 5% P_{FA};

$$L_C = N_B + 2.326\sigma_B$$

σ_B = standard deviation of background distribution

N_B = Mean number of background counts recorded in a given time period

- If, on average, N_T counts are recorded in the presence of a source, the mean number of source counts is given by;

$$N_S = N_T - N_B$$

- If N_D represents the minimum net value of N_S required to meet a P_{FA} of 5% and a P_D of 95% then

$$N_D = L_C + 1.645\sigma_{N_D}$$

- Where

$$\sigma_{N_D} = \sqrt{2N_B + N_D}$$

Minimum Detectable Activity (MDA)

- Assuming that all variances are from counting statistics

$$\sigma_{N_D} = \sqrt{2}\sigma_{N_B} + 1.645$$

- ND is therefore given by

$$N_D = L_C + 1.645\sigma_{N_D}$$

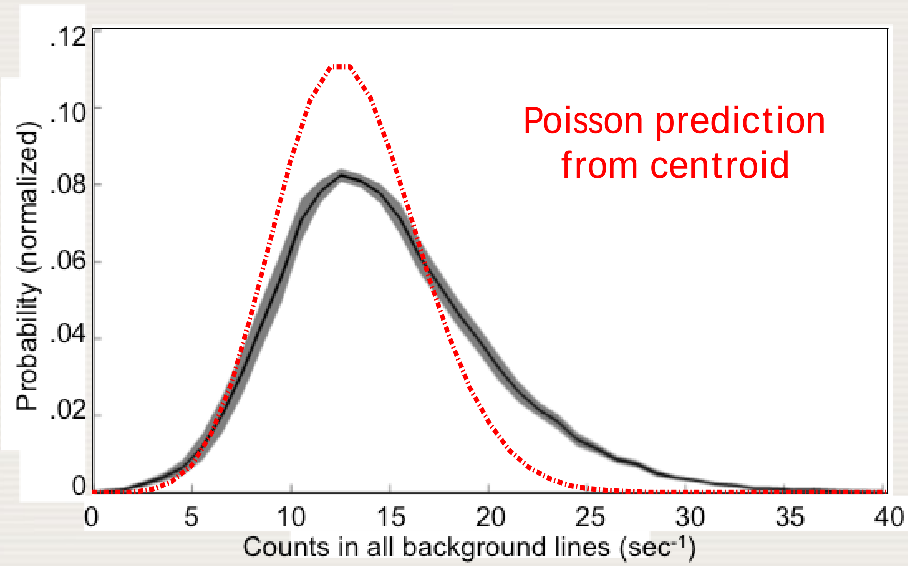
$$N_D = N_B + 2.326\sigma_{N_B} + 1.645\left(\sqrt{2}\sigma_{N_B} + 1.645\right)$$

$$N_D = N_B + 4.653\sigma_{N_B} + 2.706$$

- N_D is the minimum number of source counts required to distinguish the source from background at the chosen values of P_{FA} and P_D.
- It is converted to MDA by accounting for the activity of the source, the counting time, and the absolute detection efficiency

Detection from a Mobile Platform

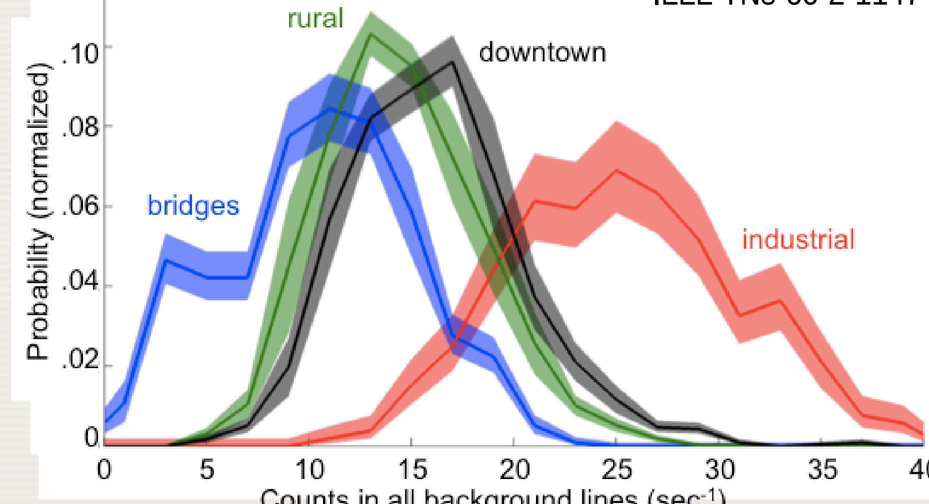
- Detection from ground-based, aerial, and hand-held mobile platforms is significantly more challenging
  - unknown and often unpredictable background reduces detection sensitivity



- Strong spatial and temporal variations (non-Poisson), in highly cluttered (e.g. urban) environments
  - systematic effects dominate

- Drives up P_{FA} or P_D is reduced to maintain P_{FA}

T.J. Aucott *et al.*, IEEE TNS 60 2 1147



- KUT is strongly dependent on environment

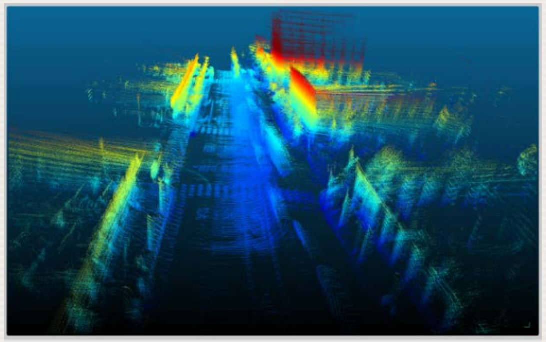
- Knowledge of environment can improve detection sensitivity

RadMAP System at LBNL

Domestic Nuclear Detection Office

Visual Imagery and LiDAR

- LiDAR = Light Detection and Ranging
- Rotating fan beam reflected from surfaces over 360^o
- Location of surface calculated from ToF assuming knowledge of the emission point



- Distribution of returned 3D coordinates known as a point cloud
- Meshing of point cloud reconstructs the world
  - but only what the laser reflects from!
  - resolution can be an issue at large stand-off distances

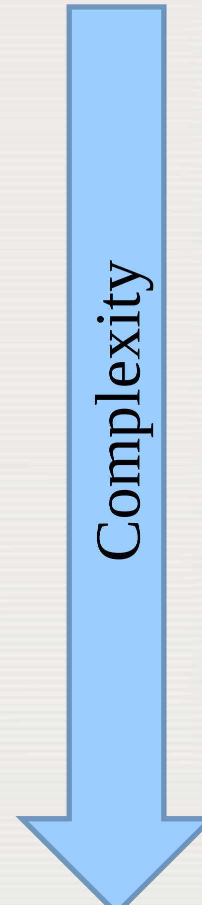
- Surfaces colourised ('painted') with RGB from 4π visual imagery
  - or temperature from an IR camera
  - or material class….
  - or how about with gamma-ray data?

- You now have a world to project and analyse nuclear/radiation data in

- Provides context and can improve detection and imaging sensitivity

Data Fusion

- Auxiliary sensors provide context for radiation data
- Correlations between radiation data and the world



Level 1 – Post-processing

- Registration of individually processed data products
- E.g. gamma-ray image overlaid on visual image
- Spatial awareness for an operator

Level 2 – Real Time Fusion

- Project radiation data into "world"
- E.g. volumetric gamma-ray imaging
- E.g.2 target tracking
- Constrain detection/imaging problem
- Reduce P_{FA}

Level 3 – Real Time Correlation

- Increase dimensionality of data by adding orthogonal info.
- E.g. count rate vs. energy vs. time vs. objects in FoV
- Reduce P_{FA} and increase P_D
- Interface radiation data with multiple intelligence sources*

Volumetric Imaging

- Reconstruction of radiation data into scene defined by LIDAR and/or visual imagery
  - Compton or coded-aperture imaging

- Reconstruction to point cloud constrains the problem and increases speed*
- Improves signal to noise by removing cone background
- Provides context and volumetric image
- Knowledge of depth allows quantitative imaging
- Knowledge of moving objects allows hypothesis testing
- Depends on resolution of LIDAR
- Occlusions can be a problem**

End of Part II
Thanks for your attention

Material (Slides) graciously shared by:

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Alexandra Ioannidou Aristotle University of Thessaloniki, Greece

Gene Jablonowksi USA EPA

Ren Cooper LBNL

(Any errors or omissions belong to Iain Darby)

IAEA
International Atomic Energy Agency