Environmental radiometric techniques applied to Aosta Valley territory

XXVI Giornate di Studio sui Rivelatori

Scuola F. Bonaudi

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Part. 1

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ARPA Valle d'Aosta

Agenzia Regionale Protezione Ambiente - Regional Agency for Environmental Protection

Organization : public body with independent status for administrative, technical-juridical, asset management and accounting purposes.

Mission : preservation and protection of the environment

Activities:

- monitoring and modelling of the various environmental components
- management and surveillance of the environmental and territorial impacts of human activities
- creation and management of the regional environmental information system
- drafting of soil, water and air quality improvement plans
- ...

Environmental radioactivity



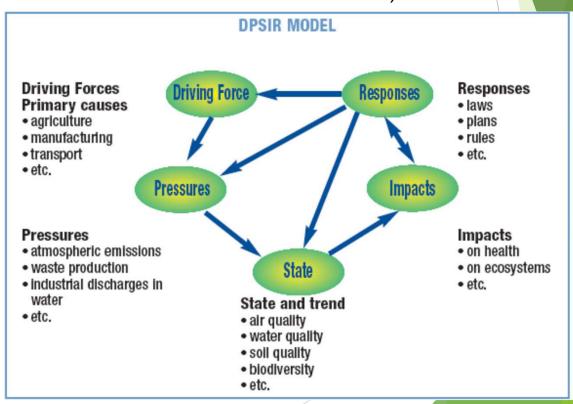
ARPA Valle d'Aosta

Agenzia Regionale Protezione Ambiente - Environmental Protection Agency

Arpa Valle d'Aosta is a member of a **network of 21 Regional Environmental Protection Agencies** managed by Ispra (Istituto Superiore per la Ricerca e la Protezione Ambientale - Italian National Institute for Environmental Protection and Research) in Rome.

Arpa's activities are designed to improve the **knowledge** of the **state of the environment**

The **indicators** are structured according to the environmental information model DPSIR. **DPSIR** includes Driving forces, Pressures, State, Impact and Responses





Surveillance of environmental radioactivity in VdA

Main Activities:

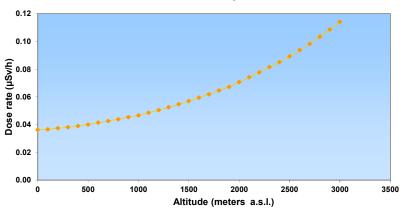
- Monitoring environmental gamma-ray dose rate
- Monitoring artificial radioactivity
 - in foodstuffs
 - in the environment
- Monitoring of natural radioactivity:
 - Radon indoor
 - Radon outdoor (planned)
- Participation to radiological emergency planning (nuclear accident, orphan sources ...)



Environmental gamma-ray effective dose rate

Cosmic contribution :

- caused by cosmic rays interaction with the atmosphere
- Depends on altitude



Terrestrial contribution:

• radionuclides in the earth's crust (K 40 and the natural radioactive series of U 238, U 235 and Th 232)



SILENA MOD 600 CE : high pressure ionization chamber Measurement range : 0.01 µSv/h ÷ 0.01 Sv/h Energy range : 80 keV ÷ 2 MeV Intrinsic uncertainty : 5% Outdoor use Temperature range : -25°C ... 50°C Data transmission: radio,ADSL, GSM



MUNICIPALITY	Place	Altitude a.s.l. (m)
AOSTA	P.za Plouves	581
LA THUILE	Les Granges	1640
COGNE	Gimillian	1788
ETROUBLES	Chevrière	1330
DONNAS	Montey	371
DONNAS	Montey	J/ I







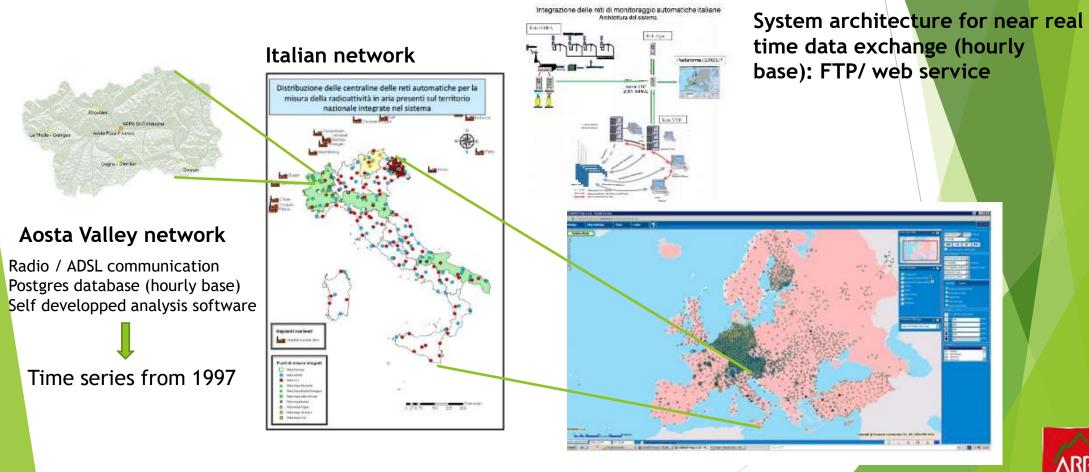






Sensor during a QA procedure

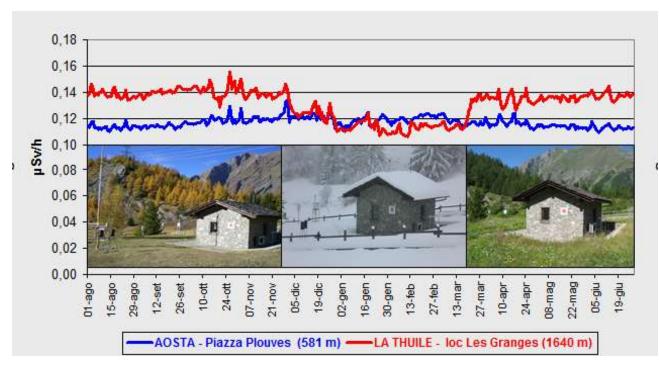




European exchange platform (EURDEP)

In order to set **early warning threshold** we have to study the time series !

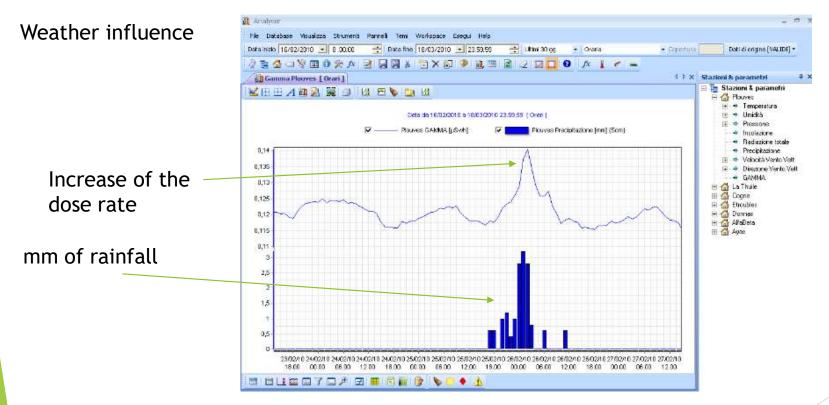
Seasonal variation



The site of La Thuile during winter is covered by a persistent snow layer that shields the gamma rays emitted from the soil



In order to set early warning threshold we have to study the time series !



The rainfall causes the temporal increases of environmental gamma-ray intensity near the ground surface due to the gamma-rays emitted from the radon daughters which are brought from the upper air and accumulated on the ground surface by the scavenging effect of precipitation.



Hints of gamma spectrometry

Detecting gamma-rays is not a direct process since they do not have intrinsic charge

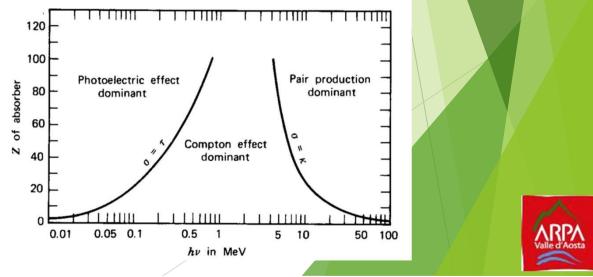
The measurement of these photons is dependent on their interaction with the electrons of the medium.

The incident photons will create fast electrons which will have a maximum energy that is equal to the energy of the incident gamma-ray

There are three major manners in which the photon will interact with the medium:

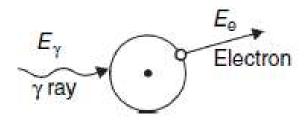
- photoelectric absorption,
- Compton scattering,
- pair production.

These different interactions change their probability of occurring depending on the energy of the gamma-ray and the atomic number of the material.



Hints of gamma spectrometry

Photoelectric absorption



Photoelectric absorption arises by interaction of the gamma-ray photon with one of the bound electrons in an atom.

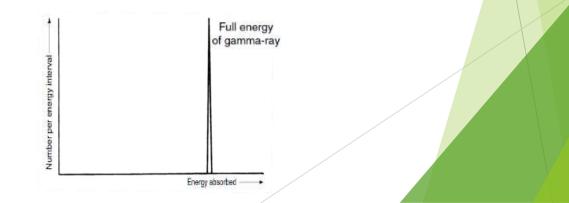
The kinetic energy that this electron carries off is

 $E_e = E_\gamma - E_b$

where E_b is the binding energy of the electron

The atom is left in an excited state with an excess energy of E_b and recovers its equilibrium by electron rearrangement.

It is normally assumed that photoelectric absorption results in the complete absorption of the gamma-ray.





Hints of gamma spectrometry



The Compton scattering interaction is the scattering of a gamma-ray off of a free or unbound electron, thus creating a scattered gamma-ray photon and a recoil electron.

The energy of the *recoil electron* is dependent on the scattering angle

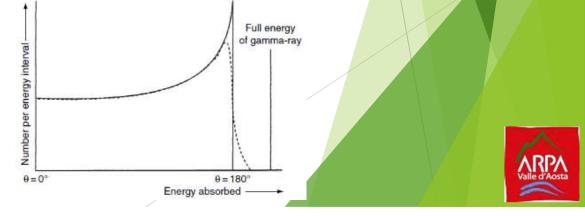
$$E_{\rm e} = E_{\gamma} \left\{ 1 - \frac{1}{[1 + E_{\gamma}(1 - \cos \theta)/m_0 c^2]} \right\}$$

- $\theta = 0$, the scattered photon retains all of its energy and the recoil electron gains no energy.
- $\theta = \pi$, the incident gamma-ray is *backscattered* and the recoil electron moves along the direction of incidence.

Taking electron binding energy into account alters the shape of the Compton response function to some extent, making the sharp point at the maximum recoil energy become more rounded and the edge corresponding to 180 backscatter acquires a slope.

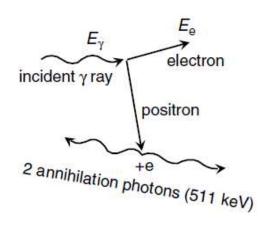
E.,

Scattered y ray



Hints of gamma spectrometry

Pair production



The process takes *place within the Coulomb field of the nucleus*, resulting in the conversion of a gamma-ray into an electron-positron pair

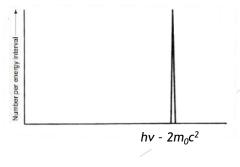
The gamma-ray must carry an energy at least equivalent to the combined rest mass of the two particles (511 keV) each, making **1022 keV** in all. (*In practice*, <u>rather more than 1022 keV</u>)

 $E_{e_{-}} + E_{e_{+}} = E_{\gamma} - 1022 \ (keV)$

The electron and positron created share the excess gamma-ray energy equally, losing it to the detector medium as they are slowed down

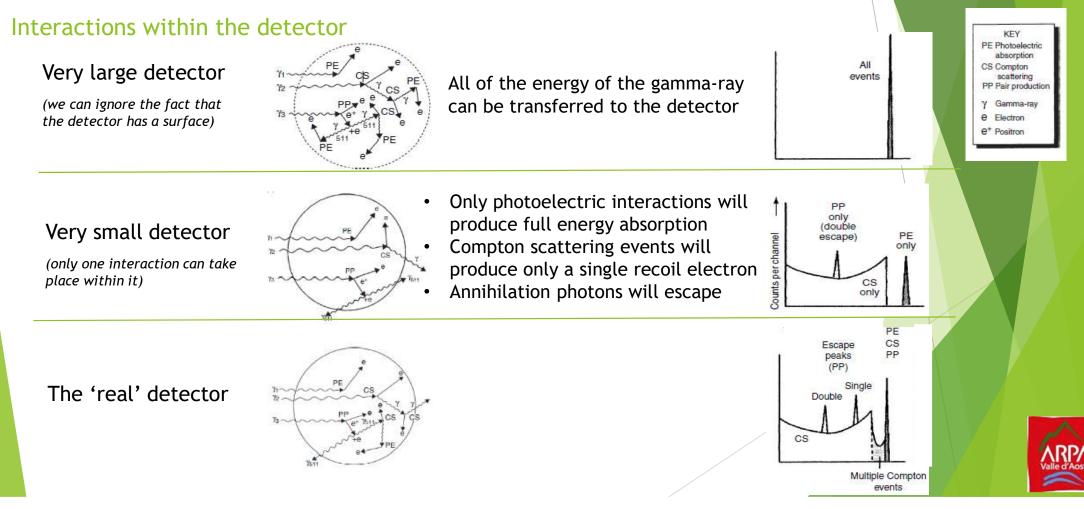
BUT when the energy of the positron is reduced to thermal energies it will annihilate with an electron releasing two 511 keV annihilation photons

If BOTH these two annihilation photons escape from the detector



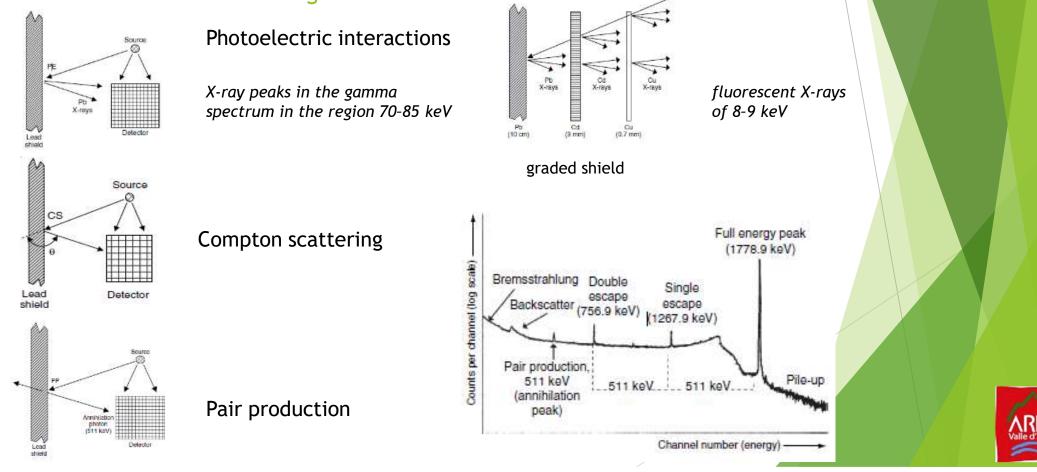


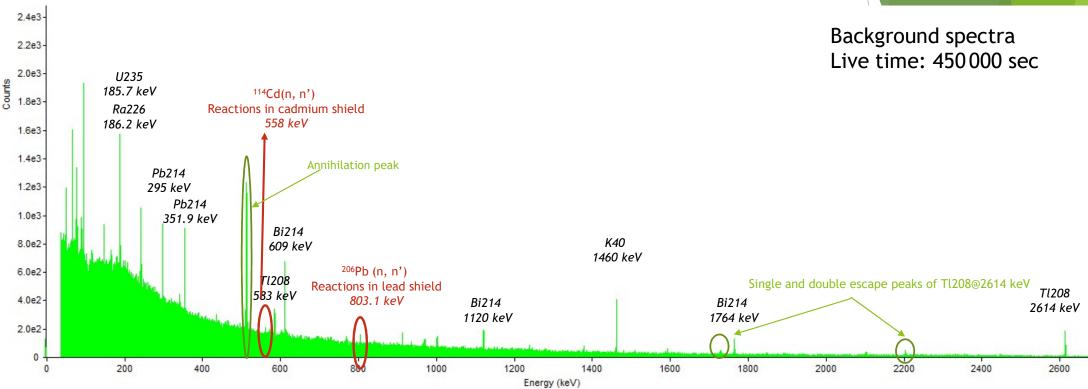
Hints of gamma spectrometry



Hints of gamma spectrometry

Interactions within the shielding





Commonly observed nuclides: K40, Ra226, Pb212, Pb214, Ac228, Tl208, Bi214, Bi212, U235, Pa234, Th234

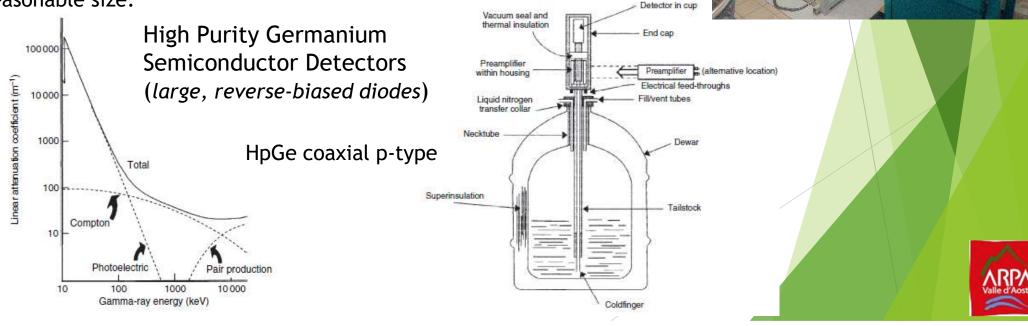
Radon gas can be a problem when counting low activity samples since it is pervasive and its non-gaseous daughter products tend to plate out inside the shield and on the detector cryostat

Cosmic-ray induced reactions: 206Pb (n, n'), 114Cd(n, n') ...



Properties of the ideal detector for gamma spectrometry

- output proportional to gamma-ray energy;
- good efficiency, i.e. high absorption coefficient, high Z;
- easy mechanism for collecting the detector signal;
- good energy resolution;
- good stability over time, temperature and operating parameters;
- reasonable cost;
- reasonable size.



Internal method accredited according to the ISO 17025:

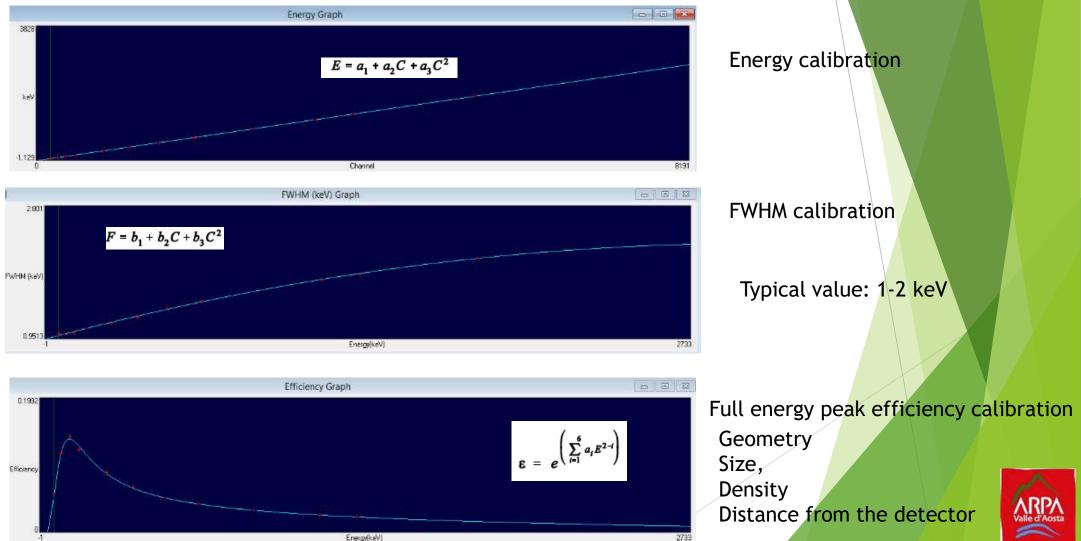
- Energy
- Efficiency
- FWHM

calibration using certified standards (efficiency for different <u>geometries</u>)



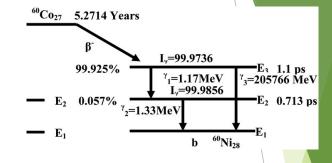
- Quality Assurance procedure (Weekly checks, monthly background measurements)
- Uncertainty budget
- MDA (Minimum Detectable Activity) Currie, ISO 11929 (Detection Limit)
- Corrections for *true coincidence summing* and *self absorption*
- Participation to Proficiency Test (IAEA, EC, ENEA...)





True coincidence summing

This source of error is a consequence of summing of gamma-rays emitted very nearly simultaneously from the nucleus.



It happens in complicated decay scheme (natural and artificial nuclides, QCY standard sources)

It is geometry dependent and errors are particularly severe when sources are positioned very close to the detector

It can be corrected in different ways:

- empirically (increasing distance from the sample to the detector),
- Analitically
- from calibration spectrum (Three curves approach Implemented in GammaVision)
- using Montecarlo (Gespecor, EFFTRAN, Angle, GEANT, ...)



Self absorption

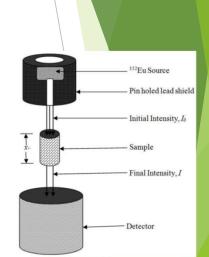
This source of error is a consequence of the interactions of the photons emitted by the sample and its own matrix

It can't be ignored in large sample size

It depends on sample density (E γ > 100 keV) and composition (E γ < 100 keV)

It can be corrected in different ways:

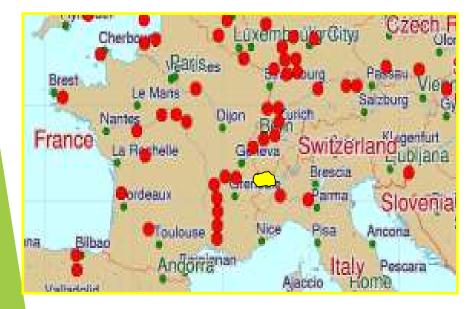
- empirically (measuring a standard source activity with different absorbers),
- analitically (complex integrals)
- using Montecarlo (Gespecor, EFFTRAN, Angle, GEANT, self developed...) → Efficiency Transfer

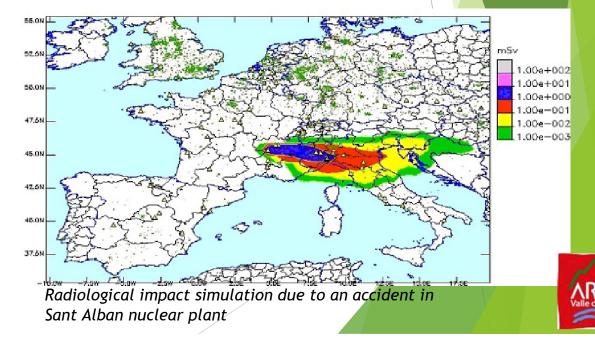




The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

The assessment of the activity concentrations of artificial radionuclides in the environment and foodstuffs makes it possible *to control environmental contamination by radionuclides from diffuse sources of radioactive materials* such as, for example, nuclear fallout from tests or accidents to nuclear facilities (About 20 european nuclear plants are located at less than 200 km from Italian boundaries (in France, Switzerland and Slovenia)).





The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Airborne particulate matter (Total Suspended Particulates)

- Continous sampling (100 l/min) in Saint-Christophe (Aosta);
- Filter: Glass fiber (borosilicate) without binder, 47 mm diameter, pore size 0.7 µm;
- Daily filter, week-end filter;
- Daily analysis: Gamma Ray Spectrometry, Gross alpha (delayed), Gross Beta (delayed);
- Monthly analysis on monthly filter pack: Gamma Ray Spectrometry;
- Searched nuclides by Gamma Ray Spectrometry : Cs137, Cs134, I131







The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Airborne particulate matter (Total Suspended Particulates)

LOW BACKGROUND ALPHA-BETA COUNTING SYSTEM:

- Plastic scintillator covered by a thin layer of zinc sulphide (ZnS)
- The lead shielding lodging the detector is equipped with a holder sledge
- An anti-coincidence guard detector is installed under the alpha-beta detector-
- Alpha and Beta Efficiency (Ratio of net count rate to 2π surface emission rate of circular Am241 (alpha) or Sr90/Y90 (beta) source): 30%





The measurements: artificial radionuclide activity concentration in the environment and foodstuffs Gross alpha activity on TSP

Airborne particulate matter (Total Suspended Particulates)

Gamma-ray spectrometry:

artificial nuclides activity concentration is < MDA (0.01-0.02 mBq/m³

22/Mar 24/Mar 26/Mar 28/Mar

Gross beta activity concentration: ~ 1 mBq/m³

Gross alpha activity concentration: ~ 0.05 mBq/m³

Fukushima accident 11-03-2011



2013

2014

2015

2016

0.25

0.2

0.05

0



Cs did not exceed the detection criterion on daily analysis

Evidence of Cs 137 and Cs 134 in the monthly filter pack of April

Cs 137: 0.016 ± 0,004 mBq/m³ Cs 134: 0.018 ± 0,004 mBq/m³

 $\frac{Cs\ 134}{Cs\ 137}\sim 1.12$

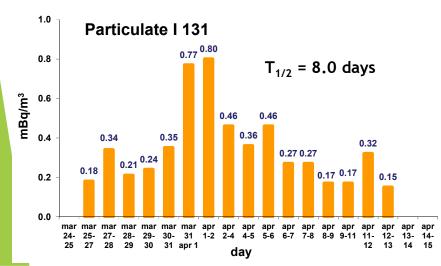


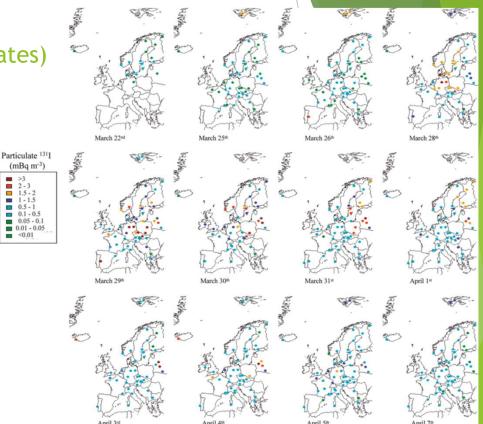
The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Airborne particulate matter (Total Suspended Particulates)

Fukushima accident 11-03- 2011

Releases started on March, 12th. The contaminated air masses entered North America from March 17th. The first European detection of 1131 from Fukushima occurred between March 19th and March 20th in Iceland, between March 19th and March 21st in the northern part of Scandinavia and **between March 23rd and 24th for most of the other European countries.** Increasing levels were generally noticed during the next 10-12 days until March 28th to March 30th for western and central Europe and until April 3rd for the Republic of Belarus.





Contrary to iodine which is mainly found in gaseous form, cesium is rapidly bound to aerosols and thus highly subject to washout removal by rain from the contaminated air masses.



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

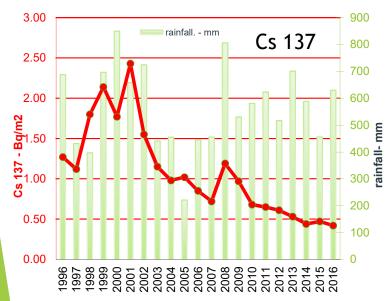
Wet and dry deposition

1 m² collecting area - monthly sampling

Evaporation \rightarrow dry matter (~ 1-8 g)

Gamma ray spectrometry: Cs 137 - Cs 134

Cs 134 : < MDA (0.02-0.03 mBq/m³)





The trend **correlation** of **depositions** of Cs 137 with **rainfall**, very evident in the first years after the Chernobyl accident (1986), is gradually fading

The link between rainfall and cesium concentrations is mediated by the particulate resuspension phenomena: the **resuspension factor** K(t), ie the ratio between the concentration in the air and the ground, is continuously decreased from the years immediately following the accident, indicating that a percentage gradually lower Cs 137 is relieved from the soil into the air

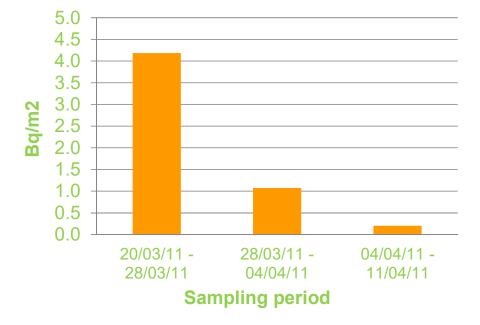


The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Wet and dry deposition

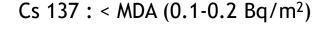
Fukushima accident 11-03- 2011

Analysis not on dry matter in order to avoid I 131 evaporation

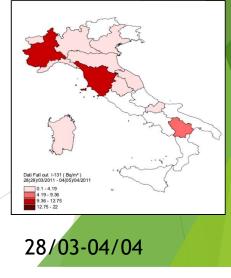


I 131 in FALL-OUT











The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Soil

Ex-situ and *in-situ* measurements of artificial radionuclides (Cs 137)

Ex-situ: different ways of sampling undisturbed soil according to different aims of the measurement:

- Surface (~ few cm) in order to have a 'blank' measurement for future releases;
- To a depth sufficient to sample nearly all the radioactive contamination in order to evaluate the total fall-out;
- At different depths in order to have a vertical profile for studying the transport of the cesium in the soil;

Recent years \rightarrow surface measurements in two sites:

Aosta hill : Avg Cs 137: 13 Bq/kg La Thuile : Avg Cs 137: 15.5 Bq/kg dry matter sieved to 2 mm

Transport of the cesium in the soil, once it is deposited, is affected by many site specific factors including soil type, rainfall, drainage, vegetation and local activities and conditions. Furthermore, local meteorological conditions can have significant impact on the fallout deposition and transport in a given area



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Soil

Ex-situ and in-situ: past years evaluation of total fall-out and determination of vertical profile in <u>medium and high altitude pastures (1000 - 2500 m a.s.l.)</u>

Development of a **combined method** (*) of <u>field gamma ray spectrometry</u> and <u>radiometric analysis in laboratory of</u> <u>soil samples</u>

Site selection :

- undisturbed terrain
- homogeneous radioactive contamination (no 'hot' spots) \rightarrow preliminary dose survey
- plain
- low grass cover
- limited presence of stones;

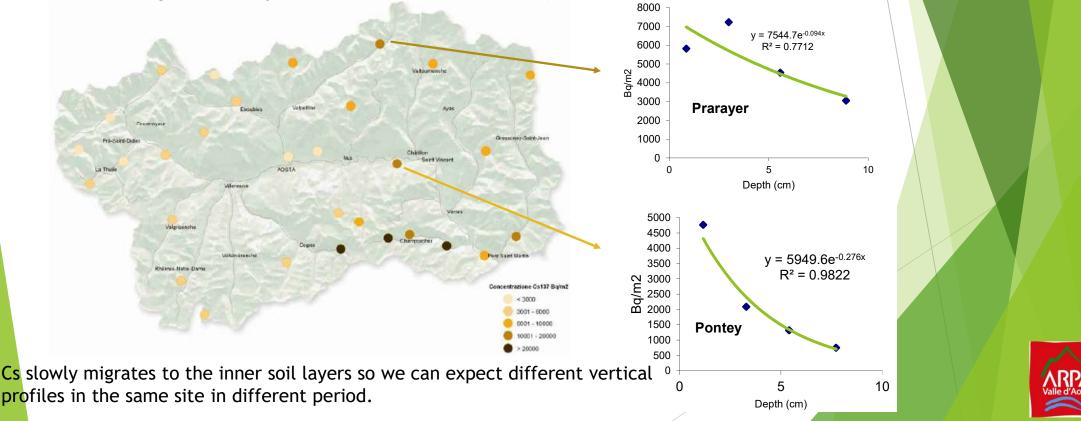
^(*)Agnesod G. et al. , "Accuracy of field spectrometry in estimating 137Cs contamination in high altitude alpine soils". Radiation Protection Dosimetry - Vol. 97, No. 4, pp. 329-332 (2001)



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Soil

Ex-situ and in-situ: past years evaluation of total fall-out and determination of vertical profile in <u>medium and high altitude pastures (1000 - 2500 m a.s.l.)</u>

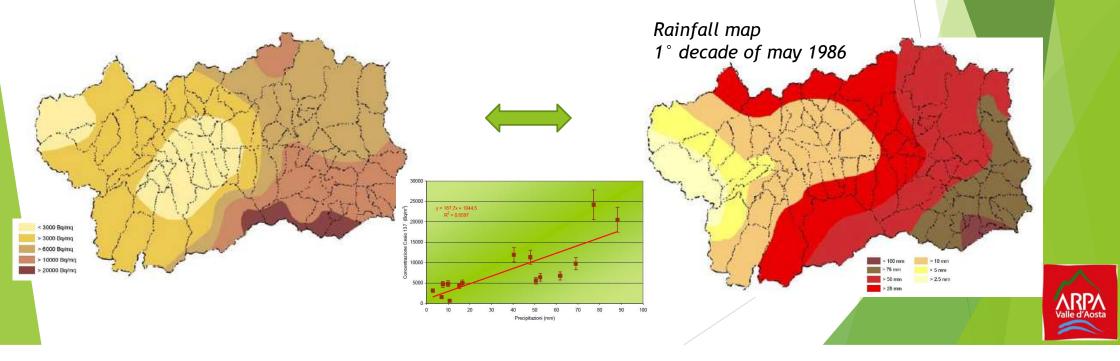


The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Soil

Ex-situ and in-situ: past years evaluation of total fall-out and determination of vertical profile in <u>medium and high altitude pastures (1000 - 2500 m a.s.l.)</u>

By means of geostatistical interpolation techniques we can create a map of Cs 137 distribution in undisturbed soil in Aosta valley



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

SMOD - Sedimentable mineral organic detritus

It's the matter drawn by the water current of a river

Sampler:

- 2 plastic bags, one inside the other, with 3 slits on each side
- 10 by 100 cm PVC strips (rolled inside the bags) which collect the detritus while the water flows inside
- 4 sampler for each sampling site, anchored with rocks
- 10-14 days of sampling time

Treatment and analysis:

- sieving to 1 mm
- supernatant liquid discarded after gravimetric sedimentation
- immediate analysis on wet sample for I 131 evaluation (not for glacial silt)
- Subsequent analysis on dry matter for other radionuclides

Miage (Courmayeur) Glacial silt

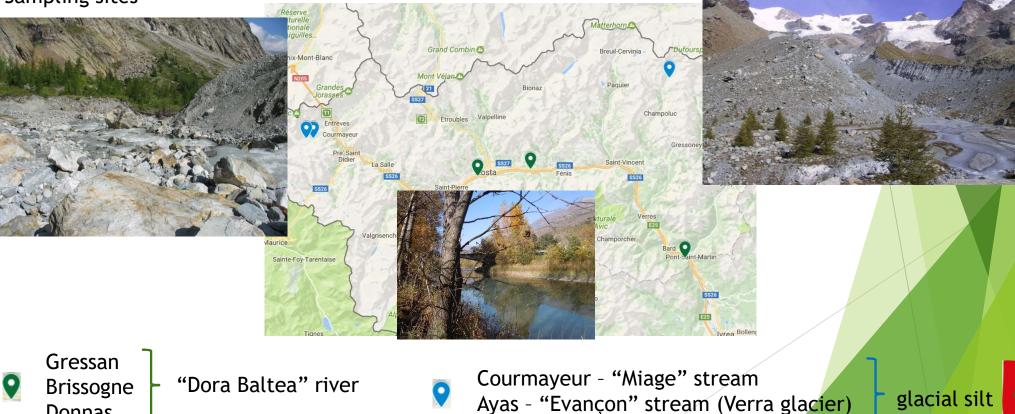


The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

SMOD - Sedimentable mineral organic detritus

Sampling sites

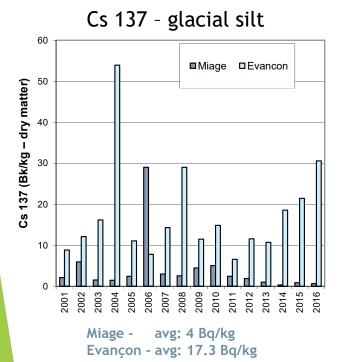
Donnas



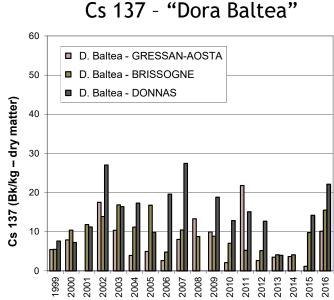


The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

SMOD - Sedimentable mineral organic detritus



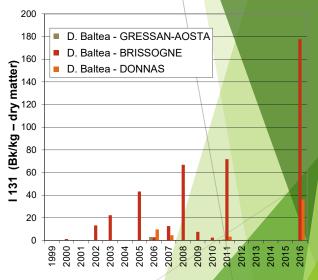
According to the rainfall distribution of the first decade of May 1986, glacier Verra had more Chernobyl fall-out than Miage



Gressan - avg: 7.6 Bq/kg Brissogne - avg: 9.4 Bq/kg Donnas - avg: 15.2 Bq/kg

Following the Dora River, the activity concentration of Cs 137 increases





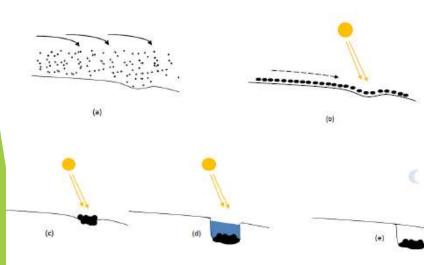
I 131 is used in Nuclear Medicine and the presence in SMOD is due to the patient home excretions. If there are no patients during the sampling period the I 131 is below the MDA (0.5 Bq/kg).



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Cryoconites

Cryoconites are special airborne dust accumulation which is deposited and builds up on the surface of glaciers and snow cover.



- a) airborne dust deposit
- b) accumulation on surface depressions
- c) absorption of solar radiation
- d) melting of the snow or ice beneath the deposit (during warm period)
- e) building up of ice (during cold period)

These dynamics occur in a remote environment

Accumulation sample on decades



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Cryoconites

Sampling sites: ablation zone of glaciers



Cryoconite composition is complex and includes cosmogenic, geogenic, biogenic and anthropogenic material

The properties of the Cryoconite formations depend from the glacier on which they are located: they can take various forms.

Timorion

Cryoconite holes



Spread cryoconite

Indren



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Cryoconites: data of the first surveys

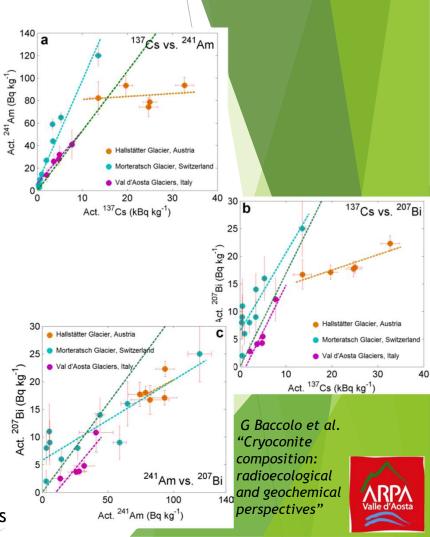
		RUTOR 2010		TIMORION 2010		AROLLA 2010		VENTINA 2011		INDREN 2011		TIMORION 2011		TIMORION 2012		PETIT GRAPILLON 2012		RUTOR 2014	
	Bq/	′kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg	Bq/kg	inc. Bq/Kg
⁷ Be	76	3	50	1212	30	1185	39	1468	28	615	19	1489	28	1033	28	152	14	114	5
40 K	56	9	9	494	18	854	35	511	20	764	17	632	16	281	11	421	9	886	11
¹³⁷ C	56	88	53	2446	36	8894	159	4251	93	5533	110	5693	111	2370	26	2236	23	2930	30
.(41 A r	7	9	5	13.7	6.7	40.8	12.8	25.7	7.4	28.1	6.4	32.2	7.6	15.2	3.3	37	4	27	2
207B	15	.1	0.6	3.2	1.6	13.8	4	4.7	0.6	4.9	0.8	6.2	1.0	3.3	0.7	5.5	0.5	7.7	3.6
¹³⁴ C	5 < M	AR		< MAR		< MAR		1.13	0.81	1.13	0.74	1.62	0.91	< MAR		(1.5)	0.4	< MAR	
⁹⁰ Sr			-	66	105	-	-	68	109	195	312	77	123	-	-	-	-	-	-
239+240	Pu -		-	13.4	1.8	-	-	15.7	2.0	26.6	3.6	19.3	2.4	-	-	-	-	-	-
238Pu			-	0.56	0.10	-	-	0.61	0.08	1.19	0.18	0.83	0.12	-	-	-	-	-	-



The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Cryoconites: comments

- Radionuclides activity concentrations very high compared to the other environmental samples
- Quantification of artificial radionuclides which generally are < MDA
 - Bi 207 → from Nuclear tests in '60s '70s. Major release during a nuclear test in Novaja Semlya (10/10/1961);
 - Pu 238 → from the burning up of the SNAP 9A satellite during re-entry on April 21, 1964;
- Cs 134 from Fukushima accident (absent in sample before 2011) → presence of "recent" dust
 - still present in 2012 in the 'Petit Grapillon' sample, due to the fact that the contamination was led by the Atlantic disturbance
- Am 241 \rightarrow from "global" fall-out, not from Chernobyl ("old" dust)
- Isotopic ratio (Cs137/Am241, Cs137/Bi207,Am241/Bi 207) are quite in agreement with measurements made by other countries on alpine Cryoconites



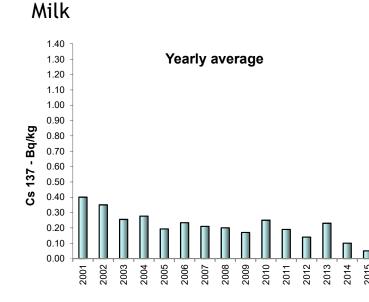
The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Food WHY ?

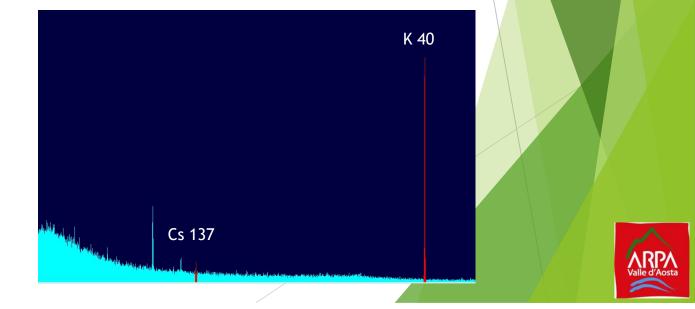
- To give pertinent information on the average extent of dietary contamination
 - foodstuffs and sampling sites on the basis of consumption
 - It is a subset of the regional sampling network with a selection of parameters to allow a picture at national level.
 - milk, cheese, meat, cereal and by-product, complete meal (=mixed diet), vegetable, fruit with different sampling frequency (monthly, three-monthly, six-monthly, seasonal)
 - Sampling held by Regional Health Agency
- To have an environmental information
 - foodstuffs and sampling sites on the basis of production
 - milk, honey, vegetable, fruit (soft fruits), mushrooms, chestnuts

The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Food



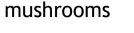
1.4 Bq/kg corresponds to the estimated dose of 1 mSv/year for individuals in the age group 10-17 years old, considering an average consumption of 56 l of milk per year. This age group is the one for which the ratio of dose/concentration is greater.

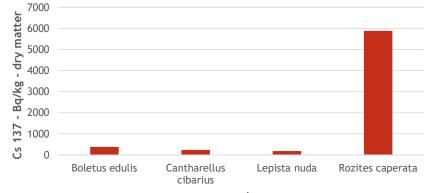


As the potassium concentration in milk in under metabolic control, also the concentration of K40 activity is stable (~50 Bq/kg in bovine milk). The K40 in milk, and in general in food, is a major contributor to the individual average dose for the population

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Food





species

High	Medium	Low
Cantharellus lutescens (M)	Agaricus silvaticus (S)	Amanita rubescens (M)
Cantharellus tubaeformis (M)	Boletus edulis (M)	Armillariella mellea *
Hydnum repandum (M)	Cantharellus cibarius (M)	Calocybe gambosa (S)
Laccaria amethystina (M)	Leccinum aurantiacum (M)	Laccaria laccata (M)
Rozites caperata (M)	Leccinum scabrum (M)	Lepista nuda (S)
Russula cyanoxantha (M)	Russula xerampelina (M)	Lycoperdon perlatum (S)
Suillus variegatus (M)	- F3 - 100	Macrolepiota procera (S)
Xerocomus badius (M)		
Xerocomus chrysenteron (M)		

Nutritional strategy: M ... mycorrhizal, S ... saprotrophic, * mostly parasitic, but with an adaptable strategy

High spatial variability in Cs 137 activity concentration even intra-specie, depending on

- Mycelium habitat and depth
- Forest type
- Fruit body location
- Fall-out
- Soil moisture and microclimate

K 40 : 500- 1500 Bq/kg - dry matter

Selected edible mushroom species with different rates of radiocaesium accumulation



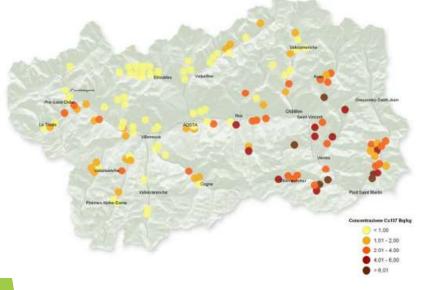
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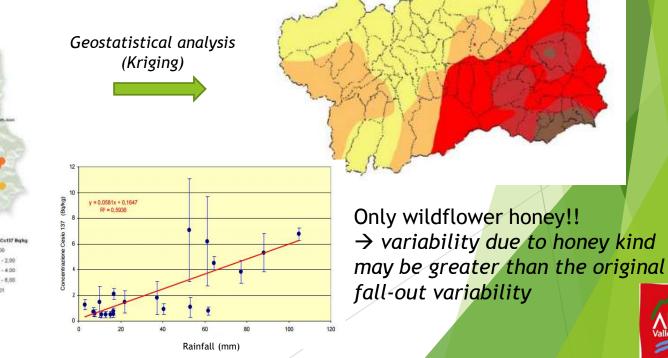
Food

Honey

 \rightarrow spatial integration achieved naturally and in a very fine way , through the methods that the bees have of collecting nectar in the environment

Cs 137 in wildflower honey distribution



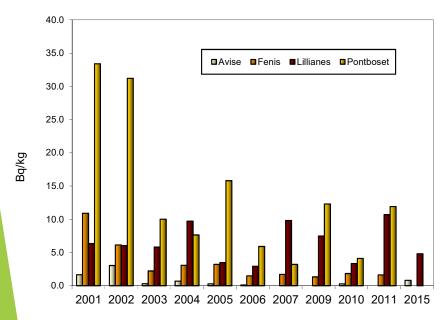


The measurements: artificial radionuclide activity concentration in the environment and foodstuffs

Food

Chestnuts

Chestnuts give information about radiocontamination of the inner layers of the soil where the chestnut tree spreads its roots.



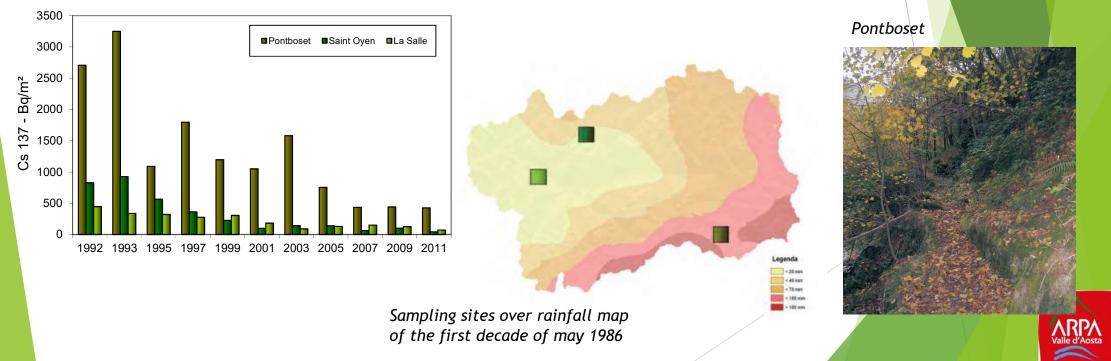
Sampling sites over rainfall map of the first decade of may 1986



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Mosses

Pleurocarpous mosses tend to form spreading carpets: they growth absorbing the water and other nutrients exclusively from the air and from fall-out. They are long living and slow growing plants, so they keep memory of past events in the order of many years.



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EMERGENCY?

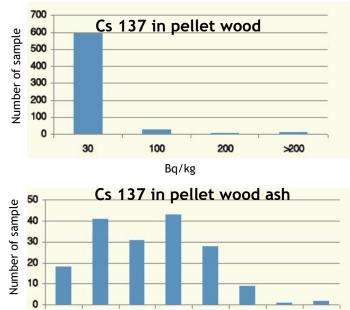
Radioactive wood pellets imported from Lithuania (2009)

Aosta \rightarrow Italy

200

500

1000



2000

3000

Bq/kg

5000

20000 >20000





	Cs-137 (Bq/kg)											
	n° samples	Average	Median	Min	Max	Standard Deviation	99° Percentile					
Pellet	639ª)	22	9.2	0.3	864.4	70.7	314.4					
Ash	173 ^{b)}	1625	900	105	45530	3951	12804					

Radioprotection approach:

- Inhalation
- Irradiation
- Ingestion

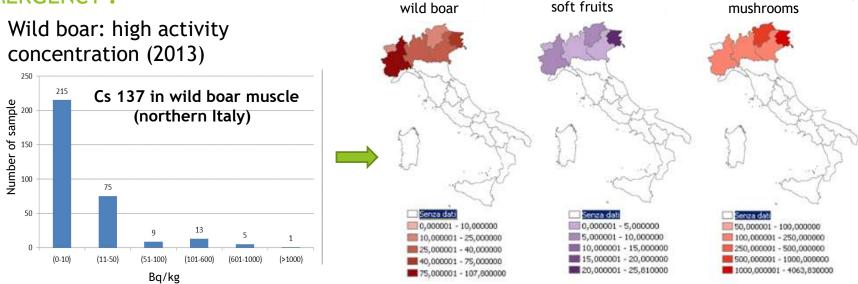
(of vegetable grown on soil fertilized with the ashes of pellets)

~10 µS<mark>v∕y</mark>



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EMERGENCY?





 \rightarrow Due to Chernobyl fall-out: correlation with soil, plants and food (mushrooms, soft fruits)

In order to explain the high contamination of wild boars a radioecological model has been developed by **ARPA Piemonte**. The model contains a soil model and considers the soil to plant transfer, the food intake and the transfer to the wild boar's meat.

Radioprotection analysis \rightarrow < 10µSv/y



Thank you

