Radiological characterization of an ancient Roman tuff-pozzolana cave in Orvieto (Italy)

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Introduction

- Orvieto: 110 km north of Rome, built by Etruscans in the 9th-8th century BC on a tuff cliff (the Rock of Orvieto ~ 1.3 km^2).

- The Rock of Orvieto results from cooling of a high temperature pyroclastic flow.

- Two igneous materials characterize the Rock:
  - *tuff:* marked stony appearance and reddish-yellow colour;
  - *pozzolana:* blackish-grey colour.
  - inhomogeneous distribution.

- Different uses in building construction since Roman age:
  - *tuff*: cut in shape of large brick and used directly in building construction
  - *pozzolana:* component ("Cuma's sand" in the Vitruvius' "*De Architectura*") of a sort of cement, the *opus caementicum*, able to rapidly set also underwater and with a long durability; still in use for hydraulic cement preparation.

Introduction (cont.)

Great availability of these materials

➡ strong extraction activity

➡ large network of caves

Many caves now touristic, lighted and air conditioned.

Introduction (cont. I)

- Study in a pozzolana quarry to characterize the natural radioactivity from the raw materials
- Tuff and pozzolana rich in natural radioactivity and still used as building materials

	A_{Tuff} (Bq kg^{-1})	A_{Pozzol} (Bq kg^{-1})
^{226}Ra	209	164
^{232}Th	349	229
^{40}K	1861	1341

important from RP point of view

Other purpose: test and intercompare different measurement and dose assessment methods in an extreme situation with high Rn/Tn levels and gamma dose rate.

Materials and Methods (1)

- cave under study located under a private dwelling
- large cave: ~ 6 x 3 m^2 with a vault ceiling of ~ 4 m reachable through completely dark small caves and narrow tunnels
- floor and first 50 cm of walls of tuff, upper walls and ceiling of pozzolana.
- two component distribution depending on the material found by ancient miners: when tuff, harder than pozzolana, found excavation direction changed

Materials and Methods (2)

In situ measurements

gamma dose rate, gamma spectrometry, radon and RnDP/TnDP monitoring

- Portable HPGe gamma spectrometers
  - crystal efficiency 70%, nitrogen-cooled, measurement live time = 3 h 20 min
- Portable NaI gamma spectrometer
  - NaI(Tl) spectrometer equipped with dedicated software to calculate gamma dose rate
- Gamma dose rate meter
  - plastic scintillator (3"x3") to intercompare results in terms of gamma dose rate.
- Radon monitoring
  - 0.6 l ionization chamber recording temperature, pressure and relative humidity ~4 h
- RnDP and TnDP activity concentrations in air
  - two independent acquisition units: total airborne RnDP/TnDP and unattached fraction, 1 alpha spectrometer for each unit: sampling = 1 h, measurement ~4 h

Lab measurements

- HPGe 70% gamma spectrometer to measure tuff and pozzolana samples
  - densities of tuff and pozzolana = 1600 kg m^{-3} and 1000 kg m^{-3}; stones crushed, homogenized, put in a 500 cm^3 Marinelli beaker
- ICP-MS: chemical analysis of pozzolana/tuff samples

Results – Gamma *in situ* measurements

Gamma dose rate

huge amount of tuff and pozzolana

- plastic scintillator facing the wall at 0.2 m above the floor: about 780 nGy h^{-1}
- NaI(Tl) detector, two tests:
  - first measurement in 4π geometry = 840 nGy h^{-1}
  - second measurement: detector placed in position similar to the plastic scintillator = 760 nGy h^{-1}

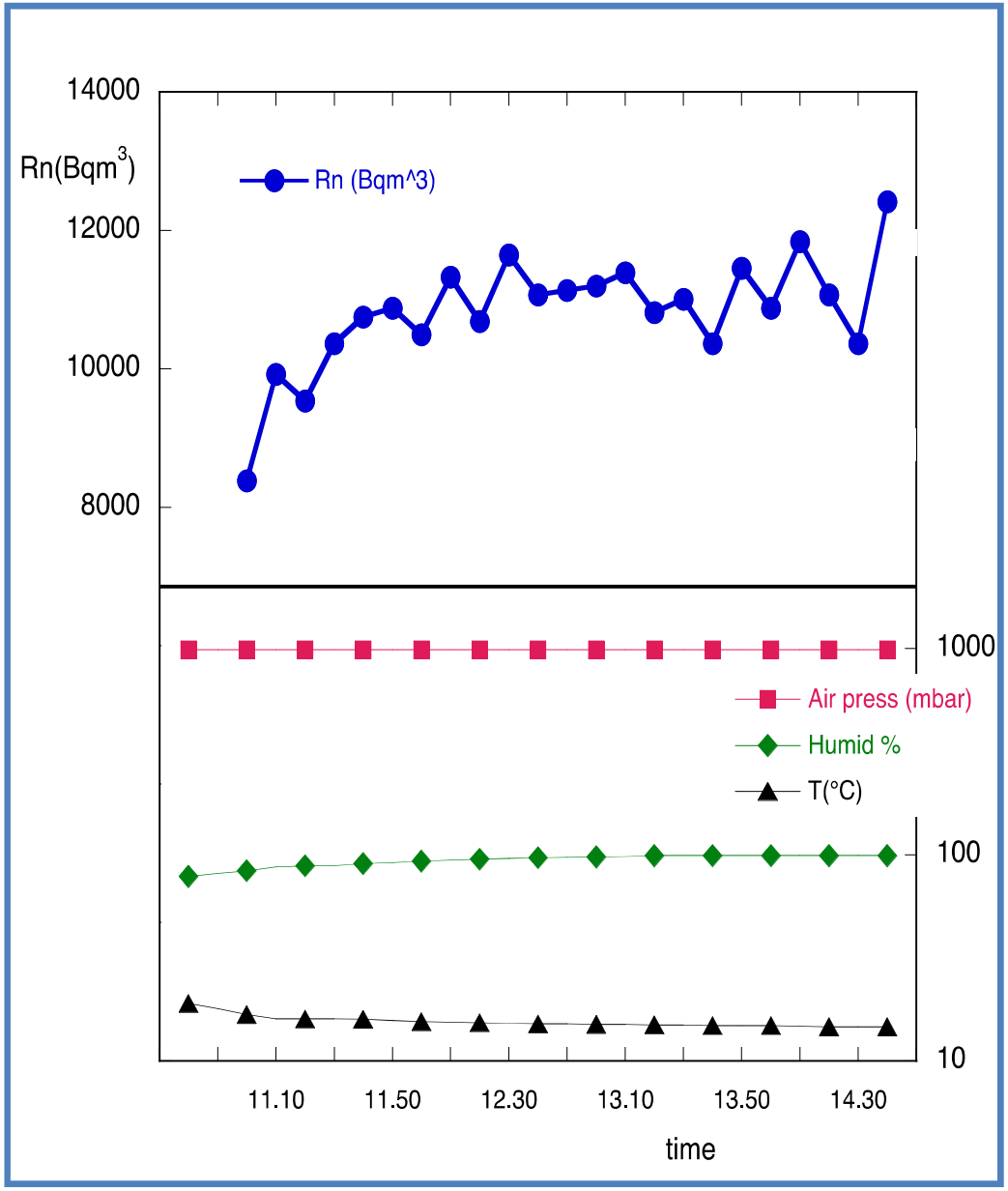
Gamma emitting radionuclide activity concentration

the gamma spectrum recorded in the cave used

- to study *in situ* the ^{226}Ra bulk material disequilibrium of chain and
- to estimate the gamma dose rate contribution from ^{40}K , ^{238}U and ^{232}Th.
- method already used successfully in indoors environments but in the cave 4 π geometry not working accurately

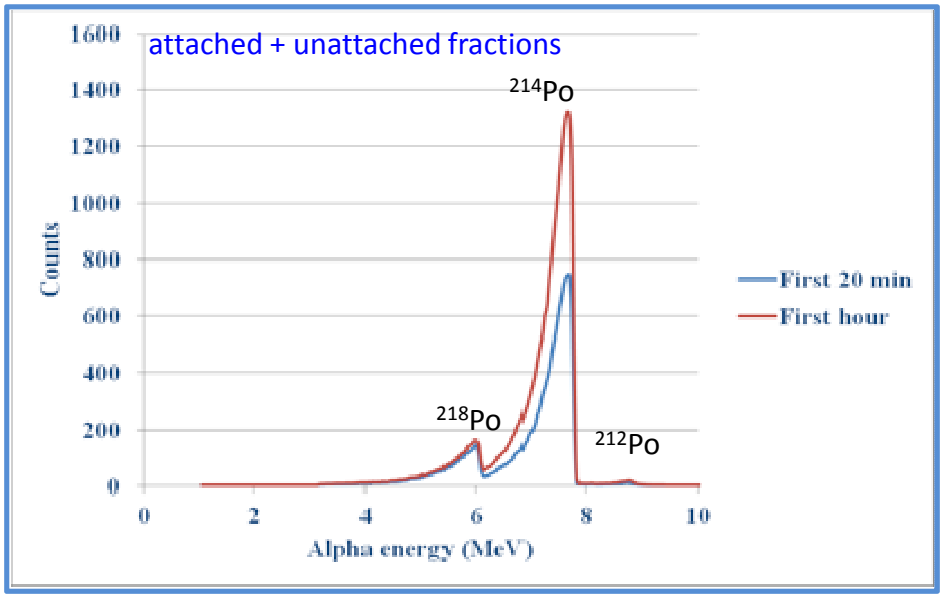
Results - Rn

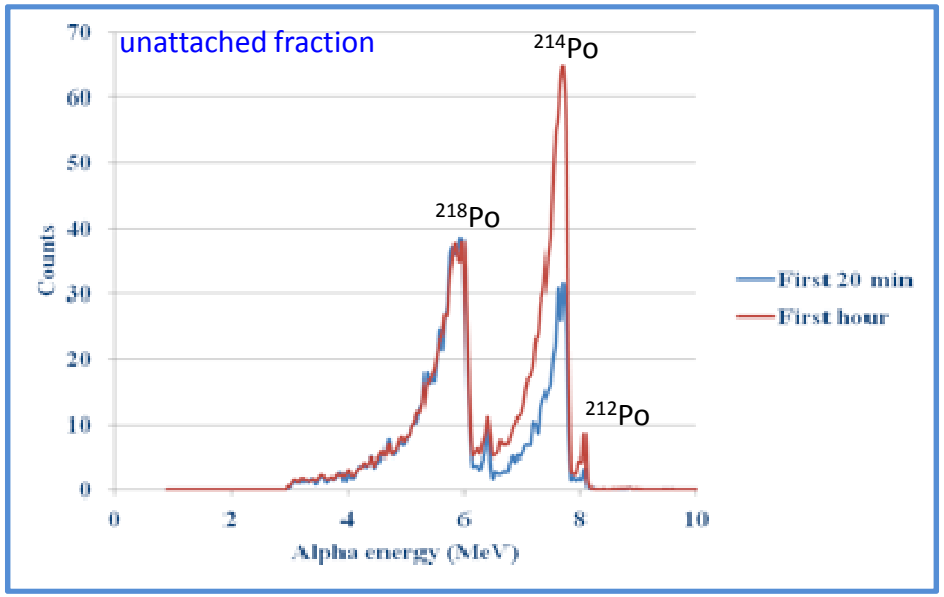
- As expected, high Rn levels during 4 hours of monitoring
- Average Rn activity concentration = 11059 ± 526 Bq m^{-3} .
- Microclimatic parameters:
  - average temperature about 15 °C
  - humidity rate about 99%.
- Good agreement with results in Roman catacombs, underground places similar from a structural point of view and excavated in tuff
  - average Rn activity concentration between 7000 and 38000 Bq m^{-3} in 7 catacombs in Rome



Results - RnDP and TnDP

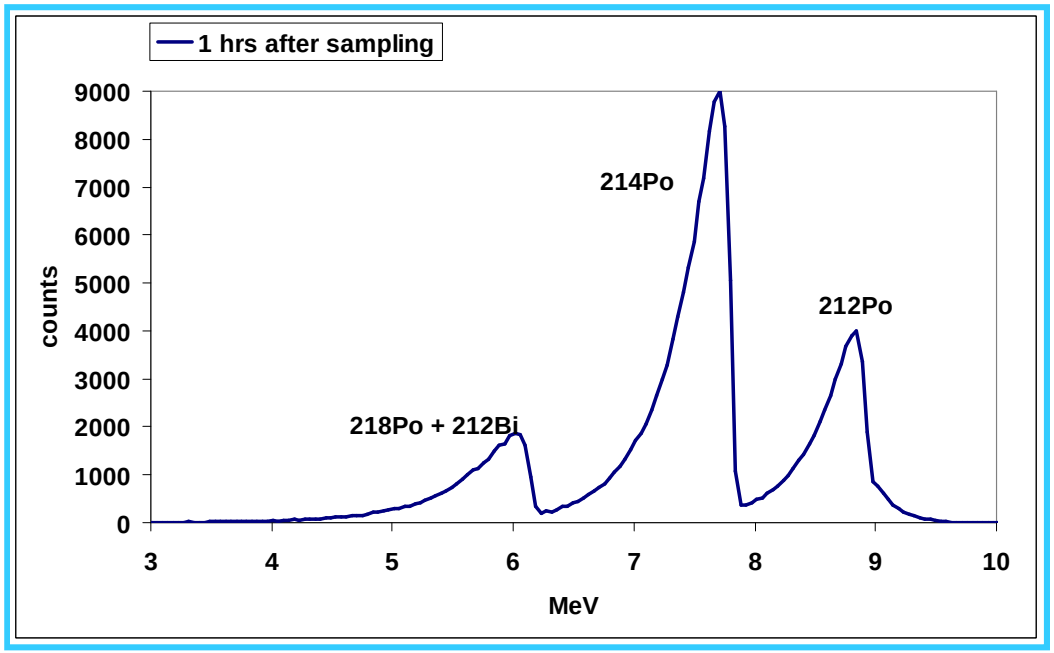
Alpha emission spectra of total airborne and unattached fraction of RnDP



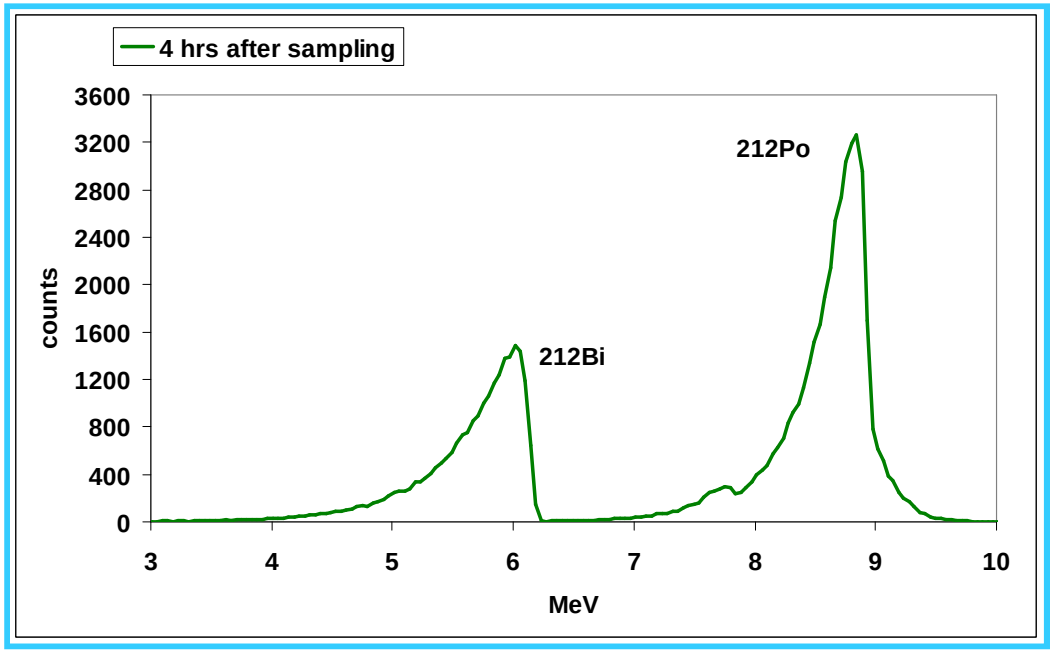


- $PAEC_{Rn}$= 288 MeV cm^{-3}
- EEC_{Rn} = 8300 Bq m^{-3}
- F = 0.75
- f ~7%
- values justified by high airborne particulate concentration and poor ventilation
- similar results in Roman catacombs

New RnDP and TnDP measurement with 30 hours sampling to enhance TnDp signal



1 hrs after sampling both Rn and TnDP detected



4 hrs after sampling only Tn DP detected

Results - Gamma spectrometry in Lab

Lab measurements confirmed:

- tuff and pozzolana rich in natural radioactivity particularly when compared with the world mean value of soil (UNSCEAR)
- $^{232}Th > {}^{238}U$ peculiarity of volcanic materials from Central/Southern Italy

	^{238}U (Bq kg^{-1})	^{232}Th (Bq kg^{-1})	^{40}K (Bq kg^{-1})
Tuff	250±20	370±10	2040±30
Pozzolana	480±30	530±10	2490±40

These data used as input in *the ISS room model*:

840 nGy h^{-1}

Results - Chemical analysis (ICP-MS)

- Very similar chemical composition of tuff/pozzolana fractions
- alumo-silicate material
- standard requirements met for classification as pozzolanic material
  - mineral additive for concrete: minimum total content of silica +alumina + iron oxide at least 75%

	Fine fraction	Coarse fraction
CaO	5.68	5.94
SiO_2	56.0	55.0
Al_2O_3	18.1	18.2
Fe_2O_3	3.87	3.74
MgO	1.07	1.07
TiO_2	0.49	0.48
K_2O	5.87	5.89
Na_2O	2.33	2.42
P_2O_5	0.20	0.16
Mn_2O_3	0.14	0.14
SO_3	0.10	0.20
LOI at 600 ° C	5.15	5.20
LOI at 950 ° C	0.79	0.59
Total LOI	5.94	5.79
H_2O	1.94	3.13

LOI = Loss of ignition

fine fraction = particles < 0.6 mm

Conclusions (1)

- This special environment is a perfect "intercomparison room"
- bulk amount of tuff and pozzolana: high levels of natural radiation
- good agreement between tools and methods to measure gamma dose rate
- lab measurements confirmed the high tuff and pozzolana activity concentration, mainly Th, peculiarity of volcanic materials in Italy
- very high ^{222}Rn activity concentration ~ 10000 Bq m^{-3}
- RnDP: PAEC ~ 288 MeV cm^{-3} (EEC = 8300 Bqm^{-3})
- unattached fraction = 7% and equilibrium factor F ~ 0.75 from low air exchange rate and high aerosol concentration

Conclusions (2)

- ^{218}Po particularly present in the unattached fraction, ^{214}Pb and ^{214}Bi more abundant in the attached fraction: competition between radioactive decay and attachment to aerosol
- short sampling time (1 h): impossible evaluation of TnDp
- new measurements with long sampling time (30 h): significant counts of ^{212}Po, all data under elaboration
- a final remark: environments with extreme conditions and exceptional exposure levels can be very useful for in-field intercomparisons
- Orvieto cave similar to thermal plant of Lurisia, Piedmont (Italy): an intercomparison of radon passive detectors is going to be carried out in Summer 2014

Thank you for your attention!