# THORIUM-232
# THE LESS KNOWN DECAY CHAIN

*Serena Risica, Francesco Bochicchio and*
*Cristina Nuccetelli*
Istituto Superiore di Sanità
(National Institute of Health)
Rome, Italy

Issues with internal emitters
EU Scientific Seminar, Luxembourg.
November, 23rd 2010

# WHY IS Th CHAIN LESS STUDIED OR UNDERESTIMATED?

- the discovery of radioactivity (→ subsequent research efforts) concerned U series
- nuclear energy production U>>Th
- in environmental matrices, activity concentration of $^{238}U > ^{232}Th$, in general
- Th (and DP) more difficult to measure and trace
- $^{222}Rn$ ($^{238}U$) the most significant for pop. dose
- $^{220}Rn$ ($^{232}Th$) risk believed to be negligible
- no epidemiological data of $^{220}Rn$ (Tn) exposures

ISTITUTO SUPERIORE DI SANITÀ

# CONTENT

## Comparison of couples of radionuclides

- $^{232}Th$ and $^{238}U$*
- $^{228}Ra$ and $^{226}Ra$
- $^{220}Rn$ and $^{222}Rn$

*not isotope but both parent nuclides

**Only some flashes on population exposure**

## $^{232}$Th *versus* $^{238}$U

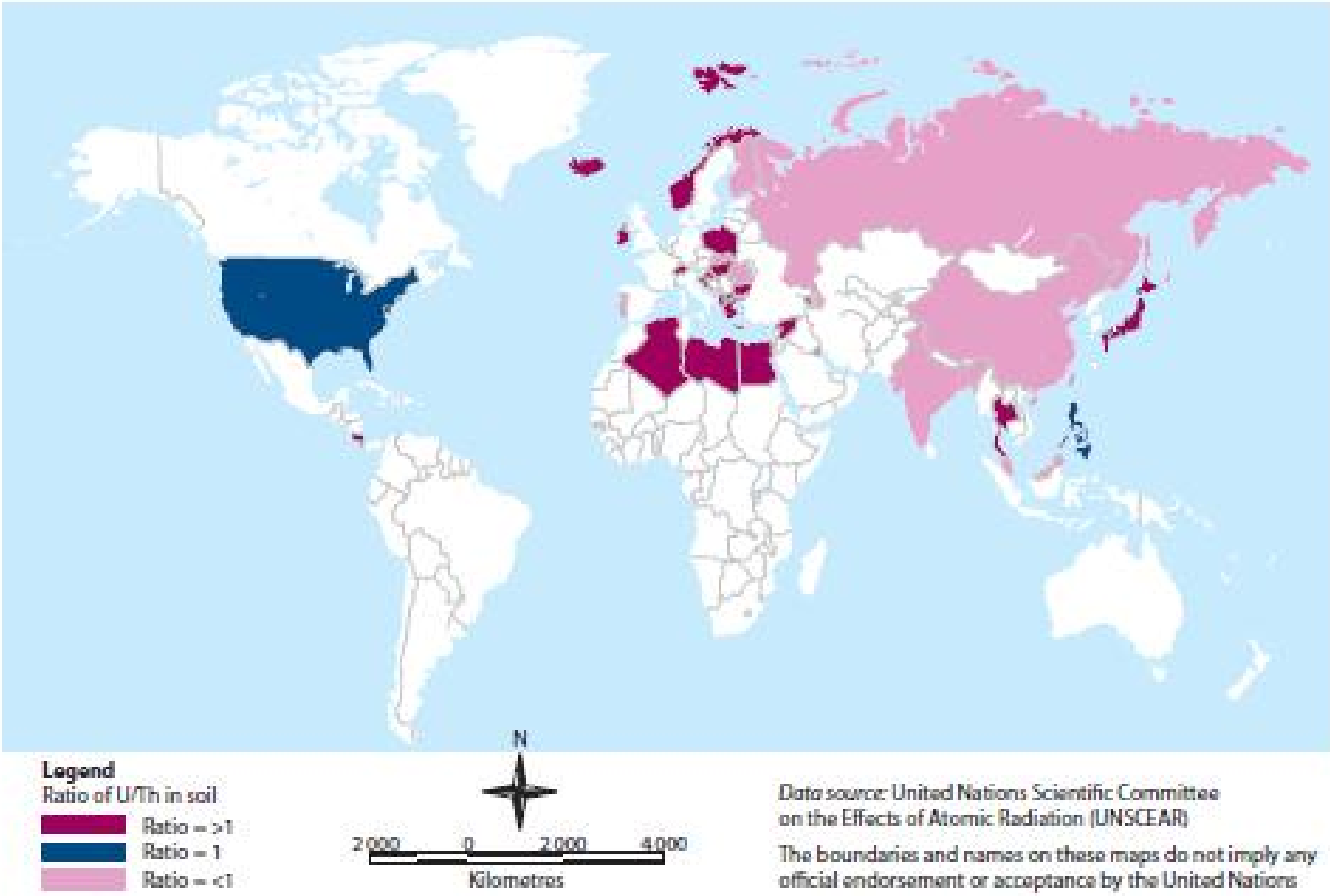
# URANIUM AND THORIUM IN SOIL

- Th/U = 3 in the Earth's crust
- in nature Th occurs almost entirely as $^{232}Th$, U primarily as $^{238}U$
- in soil concentration of $^{232}Th > ^{238}U$, in general
- specific activity of $^{238}U > ^{232}Th$
- in soil activity concentration of $^{238}U > ^{232}Th$

However, $^{232}Th/^{238}U$ *population weighted average activity concentration* in soil is 1.4
(Source: UNSCEAR, 2000, confirmed in UNSCEAR 2008 vol.1)

**Figure IX. Reported ratios of $^{238}U/^{232}Th$ concentrations in soil**

Data from the UNSCEAR Global Survey on Exposures to Natural Radiation Sources




**(Source: UNSCEAR 2008 vol.1)**

# POPULATION EXPOSURE TO $^{232}Th$

external γ irradiation →

intake with the diet →

population dose

# EXTERNAL γ IRRADIATION INDOOR

*various room models*

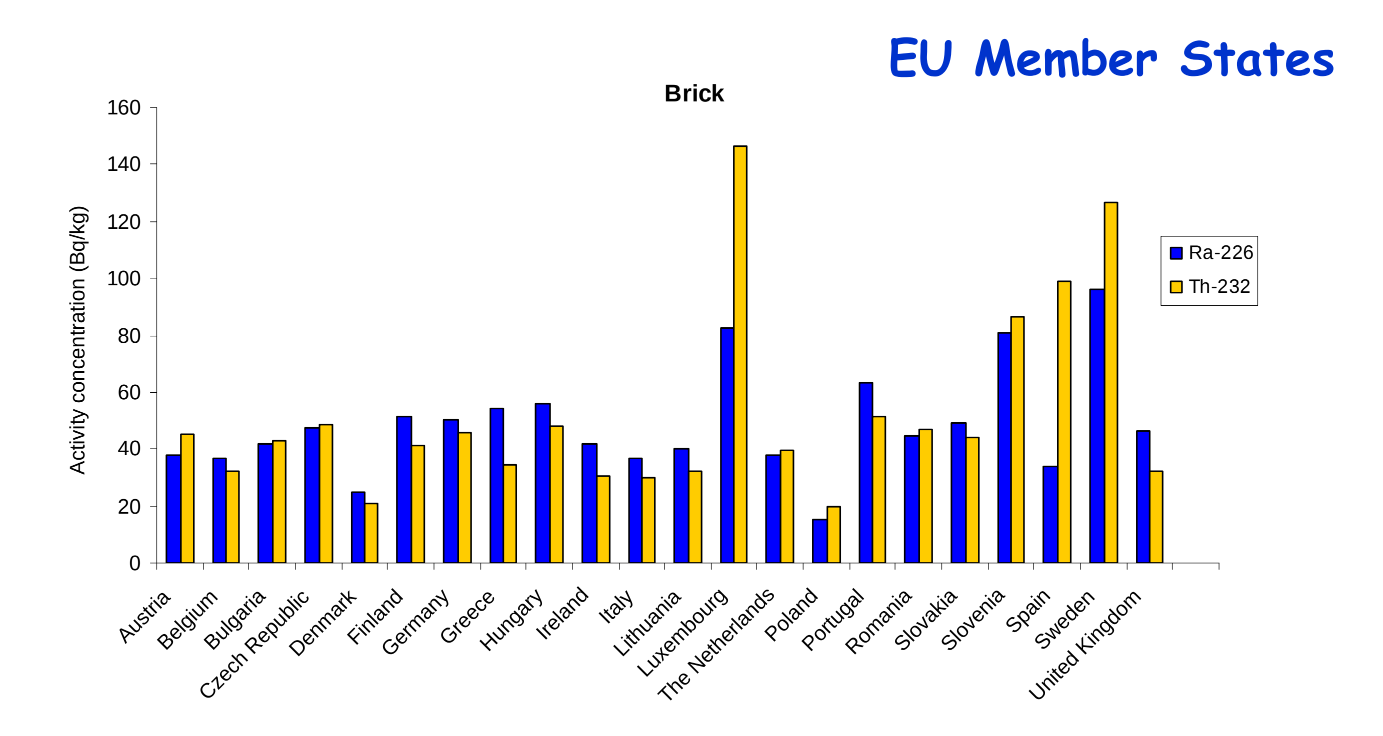
- average specific dose rate (nGy $h^{-1}$ per Bq $kg^{-1}$) $^{232}Th$ is 1.2 x $^{238}U$
- when activity concentration of $^{232}Th$ is > $^{238}U$ → $^{232}Th$ γ dose becomes a high % of the total γ dose

e.g. $^{232}Th$ 1.4 X $^{238}U$ → $^{232}Th$ dose 60% total γ dose

$^{238}U$ 25%

$^{40}K$ 15%

# ACTIVITY CONCENTRATION IN BRICKS




| N. of samples | Activity concentration in bricks (Bq $kg^{-1}$) | | |
|---|---|---|---|
| | $^{226}Ra$ | $^{232}Th$ | $^{40}K$ |
| 1537 | 48 (2 – 200) | 52 (1 – 200) | 619 (12 – 2000) |

**Source:** R. Trevisi, M. D'Alessandro, C. Nuccetelli, S. Risica, Radioactivity in Building Materials: a first Overview of the European Scenario IRPA Conference, Buenos Aires, 2008

# INTAKE WITH THE DIET

- ingestion dose coefficients: $^{232}Th > ^{238}U$ up to one order of magnitude
- scarce data available for $^{238}U$ intake with the diet and even less for $^{232}Th$
- negligible population doses

However, new investigations highly recommended

- possible accumulation phenomena
- importance of natural background values (Chernobyl accident, London $^{210}Po$ poisoning event)

ISTITVTO SVPERIORE DI SANITÀ

# $^{228}Ra$ *versus* $^{226}Ra$

# $^{228}Ra$ POPULATION DOSE

- possible pathways: diet and drinking water
- scarce data → need for national surveys with the aims of
  - detection of possible critical exposures
  - assessment of natural background values

# $^{228}Ra$ INGESTION DOSE

- $^{228}Ra$ dose coefficients
  - > $^{226}Ra$ up to one order of magnitude
  - for adolescents, children and infants: from 5 to > 40 x those for adults →

➢ lower water consumption does not compensate dose coefficient diversity

➢ 1 Bq/l β screening lev. → 0.5 mSv/y (adults)
→ 7.6 mSv/y (infants)

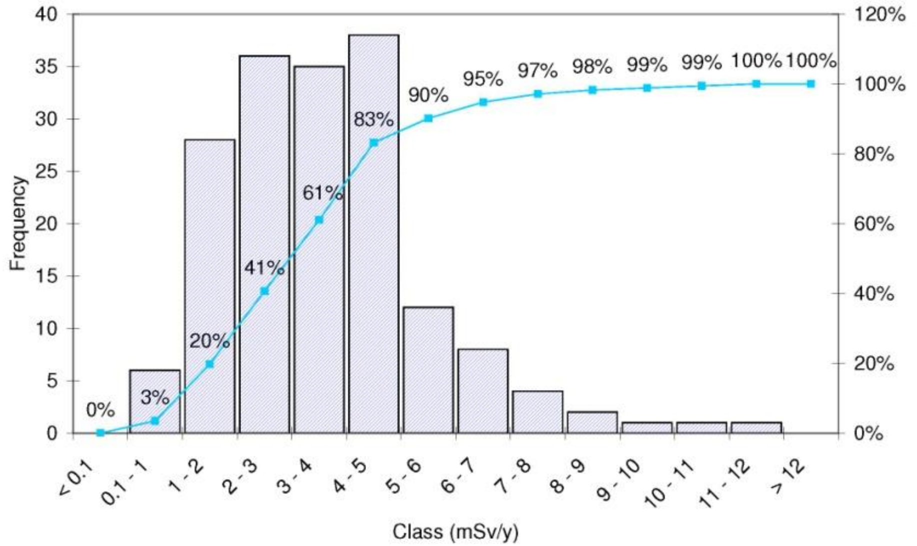
| Nuclide | Committed effective dose per unit intake (Sv/Bq) | | | | | |
|---|---|---|---|---|---|---|
| | Age class (y) | | | | | |
| | ≤ 1 | 1 - 2 | 2 - 7 | 7 - 12 | 12 - 17 | > 17 |
| $^{226}Ra$ | $4.7\ 10^{-6}$ | $9.6\ 10^{-7}$ | $6.2\ 10^{-7}$ | $8.0\ 10^{-7}$ | $1.5\ 10^{-6}$ | $2.8\ 10^{-7}$ |
| $^{228}Ra$ | $3.0\ 10^{-5}$ | $5.7\ 10^{-6}$ | $3.4\ 10^{-6}$ | $3.9\ 10^{-6}$ | $5.3\ 10^{-6}$ | $6.9\ 10^{-7}$ |

(Source: ICRP Publication 72, 1996)

ISTITVTO SVPERIORE DI SANITÀ

# RADIUM IN DRINKING WATER

## an example from the Cambrian-Vendian aquifers (Estonia)




Committed effective dose for infants

**Source:**

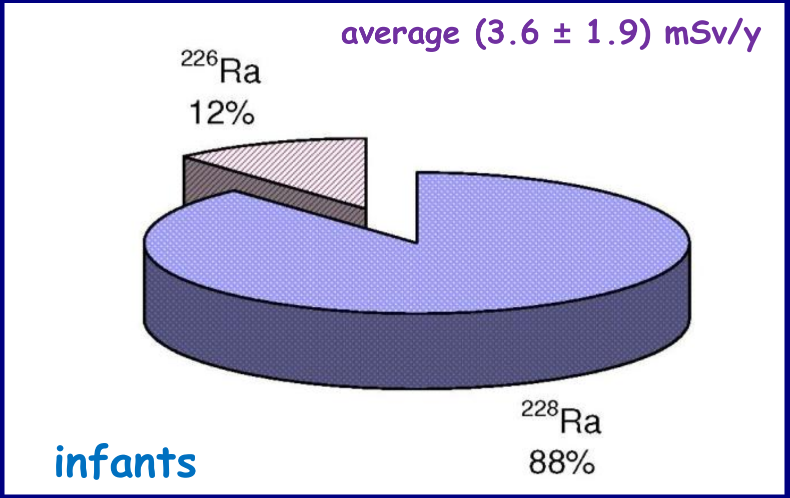
Radium isotopes in Estonian groundwater: measurements, analytical correlations, population dose and a proposal for a monitoring strategy

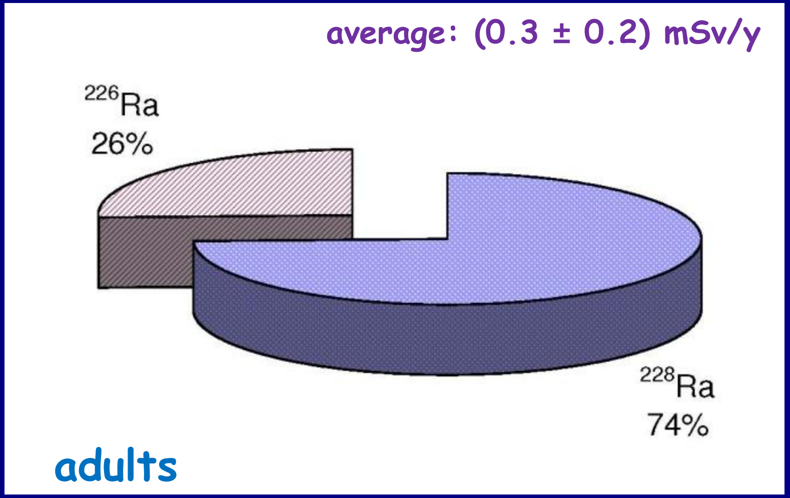
M Forte, L Bagnato, E Caldognetto, S Risica, F Trotti and R Rusconi

in press in Journal of Radiological Protection

# RADIUM IN DRINKING WATER (cont.)
## an example from the Cambrian-Vendian aquifers (Estonia)

**Average relative contributions of $^{228}Ra$ and $^{226}Ra$ to committed effective dose**




**Source:**

Radium isotopes in Estonian groundwater: measurements, analytical correlations, population dose and a proposal for a monitoring strategy
M Forte, L Bagnato, E Caldognetto, S Risica, F Trotti and R Rusconi
in press in Journal of Radiological Protection

# $^{228}Ra$ IN DRINKING WATER
## legislative aspects

- *WHO Guidelines for drinking water* account for adult exposure, only
- *Council Directive 98/93 98/83/EC on the quality of water intended for human consumption* not as yet implemented
- caution with tap and mineral water, especially with infants and adolescents
- for infant dose/mineral water: no international limit values, Italy and Germany pioneers
- caution with β screening level of 1 Bq/l

ISTITUTO SUPERIORE DI SANITÀ

# $^{220}Rn$ (Tn) *versus* $^{222}Rn$

ISTITVTO SVPERIORE DI SANITÀ

# THE TWO ISOTOPES AND SUBCHAINS

| $^{222}Rn$ and Decay Products (DPs) | | | |
|---|---|---|---|
| nuclide | half-life | decay | energy (MeV) |
| $^{222}Rn$ | 3.82 d | α | 5.5 |
| $^{218}Po$ | 3.09 min | α | 6.0 |
| $^{214}Pb$ | 26.8 min | β,γ | |
| $^{214}Bi$ | 19.9 min | β,γ | |
| $^{214}Po$ | 162 μs | α | 7.7 |
| $^{210}Pb$ | 22.2 y | β,γ | |

| $^{220}Rn$ and Decay Products (DPs) | | | |
|---|---|---|---|
| nuclide | half-life | decay | energy (MeV) |
| $^{220}Rn$ | 55.8 s | α | 6.3 |
| $^{216}Po$ | 0.15 s | α | 6.8 |
| $^{212}Pb$ | 10.6 h | β,γ | |
| $^{212}Bi$ | 60.5 min | β,γ | |
| $^{212}Po$ | 0.3 μs | α | 8.8 |
| $^{208}Pb$ | stable | | |

*Recommended data* from Decay Data Evaluation Project
(http://www.nucleide.org/DDEP.htm)

ISTITVTO SVPERIORE DI SANITÀ

# SOURCES OF $^{220}Rn$ EXPOSURE

- significant $^{220}Rn$ indoor concentration for
  - population exposure: where soil or building material, or both, are rich in $^{232}Th$
  - occupational exposure: where Th-enriched sands/ores are handled or Th welding rods are used

ISTITVTO SVPERIORE DI SANITÀ

# EXPOSURE TO $^{220}$Rn *vs* $^{222}$Rn

- $^{220}$Rn concentration cannot be predicted from $^{222}$Rn measurements
- like for $^{222}$Rn, DPs are significant for the dose
- unlike $^{222}$Rn, effects of exposure to $^{220}$Rn and DPs are not available from epi studies
- " $^{220}$Rn can be a source of error in residential radon studies that do not distinguish the two contributions to exposure. Future measurements studies should therefore consider the contribution of both $^{222}$Rn and $^{220}$Rn "
(Source: UNSCEAR 2006)

ISTITVTO SVPERIORE DI SANITÀ

# MONITORING OF $^{220}Rn$ *vs* $^{222}Rn$

- measurement techniques for $^{220}Rn$ DPs, particularly with passive dosemeters, developed mainly in the last years

- as for calibration and QA of measurements:

  – few high quality reference chambers (still significant discrepancies)

  – 2008/2009, Japan, NIRS,
  I international intercomparison of detectors
  (9 participants, 3 from EU, 6 sent back results)

  – 2009, Germany, PTB, I primary standard of $^{220}Rn$, published in 2010

# $^{220}$Rn DOSE COEFFICIENT

- the latest ICRP dosimetric approach (ICRP 50, 1987) is based on old dosimetric models (1983)

- proposal of a *comparative* dosimetric approach (C.Nuccetelli and F.Bochicchio, Radiat. Prot. Dosim. 78,1998), supported by dosimetric calculations (J.W.Marsh and A.Birchall, Radiat. Prot. Dosim. 81, 1999)

- UNSCEAR 2000: similar *comparative* approach
  → $^{220}$Rn $DCF_{dwell.}$(EEC): 40 nSv/(Bq h m$^{-3}$)

EEC($^{220}$Rn Tn) = 0.91 ($^{212}$Pb) + 0.087($^{212}$Bi)

# $^{220}$Rn DOSE COEFFICIENT (cont.)

## More recent estimates:

- **UNSCEAR 2006 (Annex E):**

  confirmed the UNSCEAR 2000 estimate

- **T.Ishikawa et al. 2007**

  $^{220}$Rn $DCF_{dwell.}$(EEC): → 116 nSv/(Bq h m$^{-3}$) dosim.
  → 7 nSv/(Bq h m$^{-3}$) comp.

- **G.M.Kendall and A.W.Phipps 2007**
  - $^{220}$Rn DCF (Sv/Bq): (2-3) X UNSCEAR estimate
  - DCFs for children are rather larger but compensated by lower breathing rate

# $^{220}Rn$ REGULATION

- Title VII of EURATOM 96/29 Directive suggested to limit $^{220}Rn$ at workplaces, but proposed no limit or recommended values
- no further decision in the draft Directive
- draft IAEA BSS provide no suggestion for $^{220}Rn$ indoors
- no national limitation has been issued so for

Should monitoring and dosimetric difficulties be solved first?

# UNSCEAR 2006 (ANNEX E)

"In the past, exposures to Tn and its decay products were often ignored...it has become increasingly clear that the exposure to Tn and its decay products cannot be ignored in some environments (both workplaces and residential) as it contributes to the risks otherwise assigned solely to Rn and its decay products.

... data collected for the present study indicate that the levels of Tn (and hence doses from exposure to Tn and its decay products) are highly variable and that Tn may provide a larger contribution to natural background dose than previously thought. Doses from Rn and Tn represent approximately half of the estimated dose from exposure to all natural sources of ionizing radiation."

ISTITUTO SUPERIORE DI SANITÀ

# CONCLUSIONS

## Research needs for Th and decay products

- investigate sources in diet and indoor/outdoor environment, workplaces included

- improve measuring techniques:
  - new detection techniques?
  - traceable standard
  - reference materials
  - intercomparison runs

... an interdisciplinary effort!

- improve $^{222}$Rn/$^{220}$Rn dosimetry. Emerging problem: $^{220}$Rn contribution to $^{222}$Rn measurements in epi studies

- improve assessments of $^{220}$Rn dose coefficients

# CONCLUSIONS (cont.)

## Policy implications for Th and decay products

- Attention be paid to
  - lower age classes particularly for $^{228}Ra$ in drinking water
  - screening levels in case of $^{228}Ra$
  - mineral and spa water

- Is time ripe to propose regulations for limiting $^{220}Rn$ concentration indoors? In workplaces first?

- Should environmental monitoring (EU Recommen.2000) be extended to major natural radionuclides in order to
  - know natural background values?
  - highlight critical exposures ?

ISTITVTO SVPERIORE DI SANITÀ

Thank you
for your attention