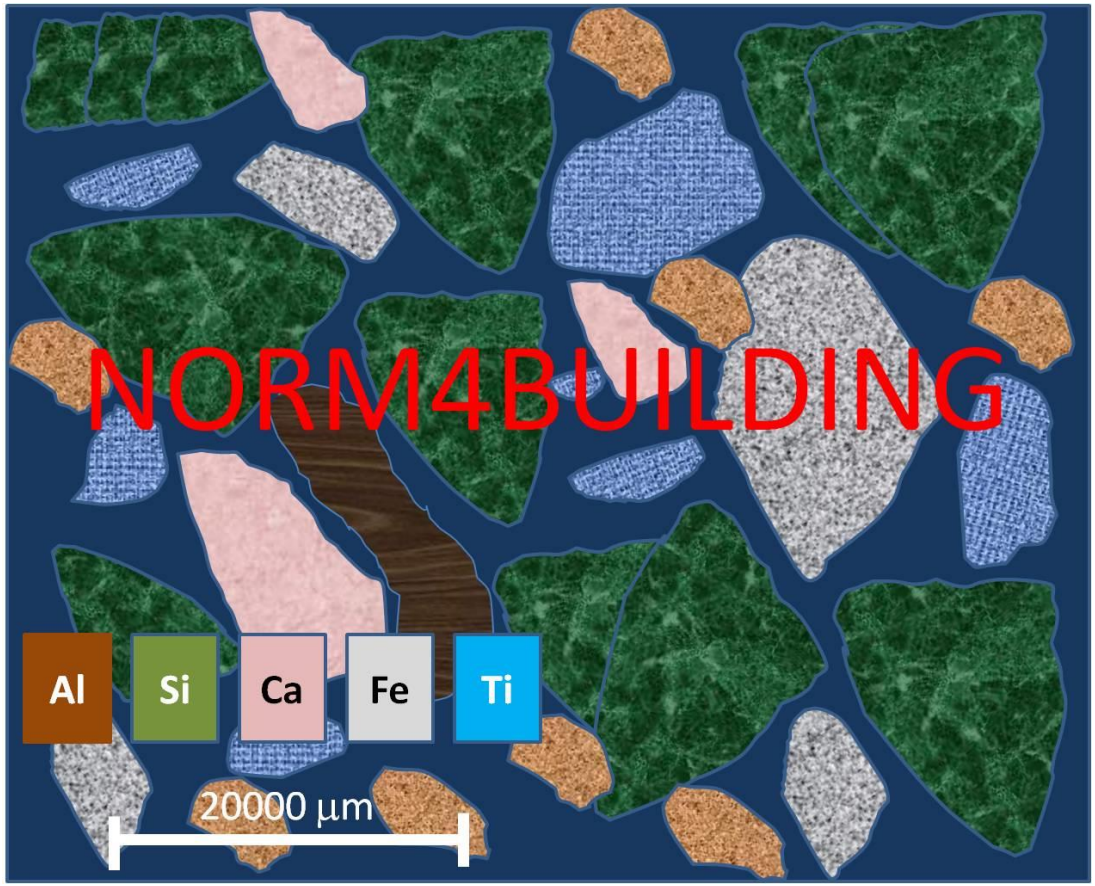
COST Project NORM4BUILDING

On site measurement strategies for NORM and Building materials



IRANOW 15/10/2015 Salerno

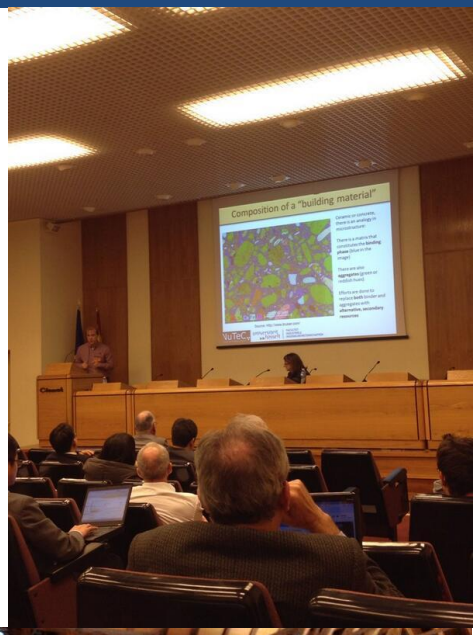
Wouter Schroeyers[1], Gerti Xhixha[2], Niels Vandevenne[1], Fabio Mantovani[2], Tom Croymans[1], Mark Stals[1], Sonja Schreurs[1]

Transport and Urban Development (TUD)

COST – network 'NORM4Building'

(Research and Technological development Framework Program)

- ORGANIZATION OF MEETINGS (2016):
  - Linked to the **Terrestrial Radionuclides in Environment** symposium in **Veszprèm, Hungary (May 17-18, 2016)**
  - Linked to**: Rilem materials-System conference**, **Copenhagen, Denmark (Aug 21-24, 2016)**



- SHORT-TERM SCIENTIFIC MISSIONS
  - New call: February - March 2016

- TRAINING SCHOOLS
  - Athens (September 12-16th, 2016)

- PUBLICATIONS and DISSEMINATION

www.norm4building.org

Main objective 'NORM4BUILDING'

- Exchange of multidisciplinary knowledge and experiences (radiological, technical, economical, legislative, ecological, …)

Stimulate the
reuse of NORM residues in new tailor-made sustainable building materials
(focus on concrete, cement and ceramics)

while
considering exposure to external gamma radiation and the resulting indoor air quality.

Secondary raw materials

NORM residues (with interesting properties for reuse in building materials)	Codification EU-Waste Catalogue?	Estimated production (Milion Tons/year)
coal fly-ash	001 02 or 10 01 16	44 (2003, EU 15) [1]
slag and bottom ash from a coal-fired power plant	10 01 01 or 10 01 14	8 (2003, EU 15) [1]
phosphorous slag from thermal phosphorus production	06 09 02	-
phosphogypsum from phosphoric acid production,	-	180 (2003, World) [2]
red-mud, (bauxite residue), from alumina production	01 03 07	120 (2003, World) [3]
unprocessed slag from primary iron production	10 02 02	260-310 (2011, World) [4]
steel or stainless steel, lead slags	10 04 01	130-210 (2011, World) [4]
copper slags, from primary and secondary production.	10 06 01	24,6 (2009, World) [5]
tin slags from primary and secondary production	-	-
specific residues originating from pyro- and hydro-metallurgies producing platinum group metals or rare earth elements	-	-

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[1] Ecoba - SPECIAL PRINT CPI 04/06
[2] A.B. Parreira, A.R.K. Kobayashi Jr., O.B. Silvestre, J. Environ. Eng. 129 (2003) 956–960
[3] www.redmud.org/Disposal.html
[4] U.S. Geological Survey, Mineral Commodity Summaries, January 2012
[5] S.H. Chew, S.K. Bharati, Proc. Of Int. Symp. On Geoenvironmental Eng., ISGE2009, 705, 2009

NORM processing industries (New EU-BSS ANNEX VI)

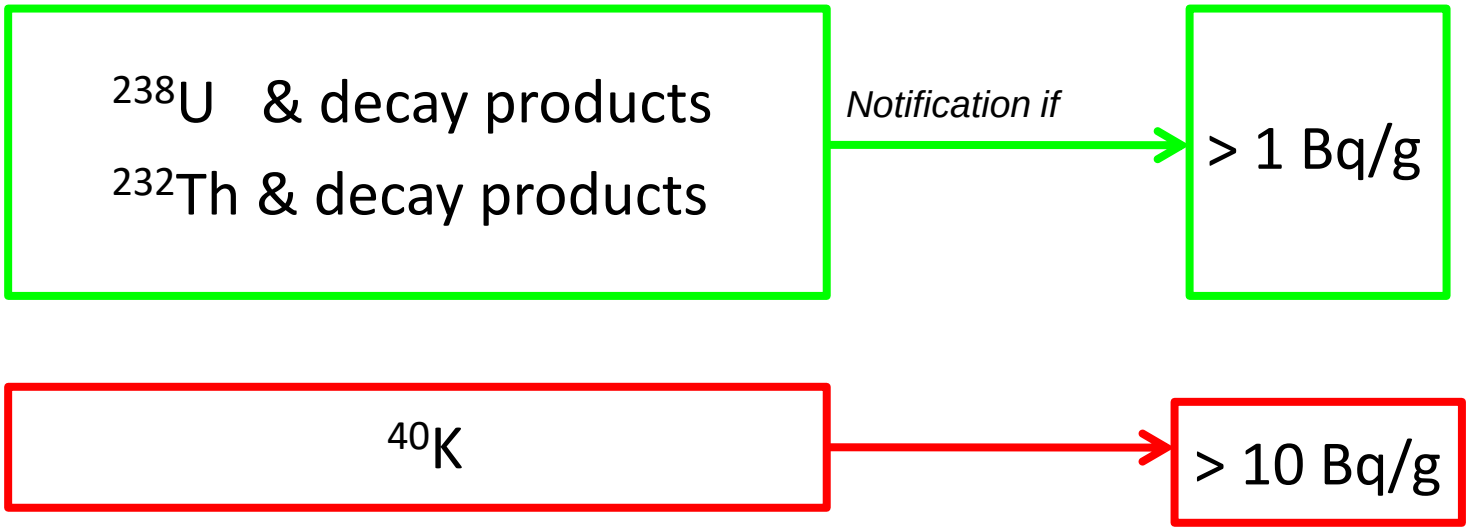
- extraction of rare earths from monazite;
- production of thorium compounds and manufacture of thorium-containing products;
- processing of niobium/tantalum ore;
- oil and gas production;
- geothermal energy production;
- TiO_2 pigment production;
- thermal phosphorus production;
- zircon and zirconium industry;
- production of phosphate fertilisers;
- cement production, maintenance of clinker ovens;
- coal-fired power plants, maintenance of boilers;
- phosphoric acid production;
- primary iron production;
- tin/lead/copper smelting;
- ground water filtration facilities;
- mining of ores other than uranium ore.

Including relevant secondary processes
Member States can add other relevant activities

NORM processing industries

*European list NORM-industries for strict regulation:

Natural occurring radionuclides:



New Euratom-BSS in addition to CPR

Residues of NORM processing industry to be recycled in building materials?

BSS Art. 75.2 + Indicative list in Annex XI + CPR

Yes

$$ACI = \frac{C_{Ra226}}{300}\frac{Bq}{kg} + \frac{C_{Th232}}{200}\frac{Bq}{kg} + \frac{C_{K40}}{3000}\frac{Bq}{kg}$$

C in Bq/kg

Index ≤ 1?

Results may be requested by the regulatory authority

TS 351014 + CPR art. 18 + BSS Annex VII & VIII

Yes

No regulatory control

BSS Art. 26 & 30 and Annex VII Table A part 2

No

BSS Art. 75

Specific national requirements or restrictions taking into account:

Density; thickness of the material; factors relating to materials and intended use of it (bulk or superficial)

(Gamma) Dose **estimate < 1 mSv/year**

"Declaration of performance"

CE marking + classification

to be made available to the consumers

CPR Art. 4, 8.2, 8.3, 11, 24

+ EU harmonized standards

CEN-TG 32

Based on a presentation by Stéphane Calpéna (EC-DG-ENER-D4), EU-NORM 2, Prague (2014)

Radon in dwellings and public buildings

Art. 74.1

National Reference Levels shall not exceed:

300 Bq/m³ → Effective dose 18 mSv/year (ICRP)

Art. 74.2

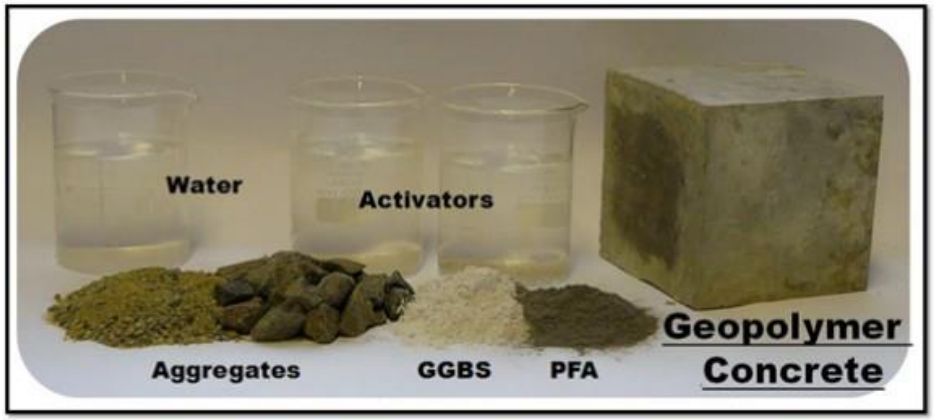
Member States shall promote action to identify dwellings with radon concentration (as an annual average) exceeding the reference level and encourage, where appropriate, by technical or financial means, radon concentration-reducing measures in these dwellings.

Art. 74.3

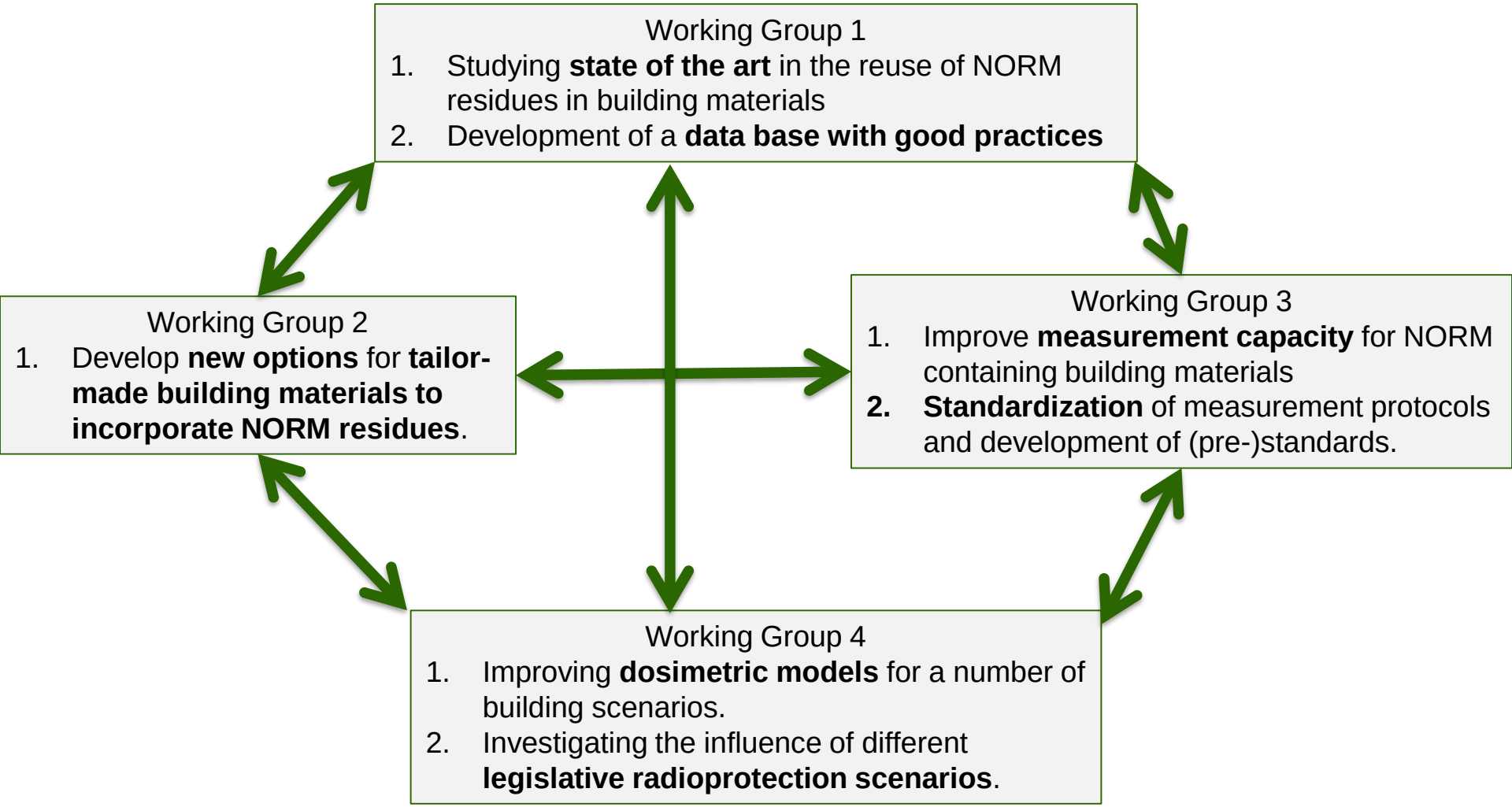
…local and national information to be made available…

NORM in building materials (New European Directives, **Annex XI**)

- Materials including by-products or residues from NORM industries such as
  - fly ash,
  - phosphogypsum,
  - phosphorous slag
  - tin slag
  - copper slag
  - red mud (residue from aluminium production)
  - residues from steel production



Scientific focus working groups



Working Group 1

‘Data base with good practices’

Norm4Building Database

a) **Criteria** for evaluation of practices were set.

b) Gathering information on NORMs currently used for building materials

c) Including **representative national surveys**

Information per entry

General information:

- By-product name; Industrial sector
- Country
- Total amount of by-product [Mt], Number of surveyed samples
- References

Radiological features

- Activity concentration (terrestrial isotopes: Ra-226; Th-232, K-40)
- Activity concentration index
- Emanation and exhalation features

Non-radiological features

*Density	Particle size distribution	H_2O	SiO_2	Al_2O_3	CaO	Fe_2O_3	C	Na_2O	SO_4^{2-}	MgO	Cl	P_2O_5	LOI	pH
[kg/m^3]		[wt%]	[wt%]	[wt%]	[wt%]	[wt%]	[wt%]	[wt%]	[wt%]	[wt%]	[wt%]	[wt%]	loss on ignition [wt%]	

Working Group 1

‘Data base with good practices’

Norm4Building Database

a) **Criteria** for evaluation of practices were set.

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Evaluation of practices

a) Collaboration with industry to **evaluate the application in construction materials**

b) Cost-benefit and SWOT analysis

Output

a) Updating ‘**Activity Concentration Index**’ (ACI) **database**

b) Update information for the **European Waste Catalogue**

c) Dissemination plan

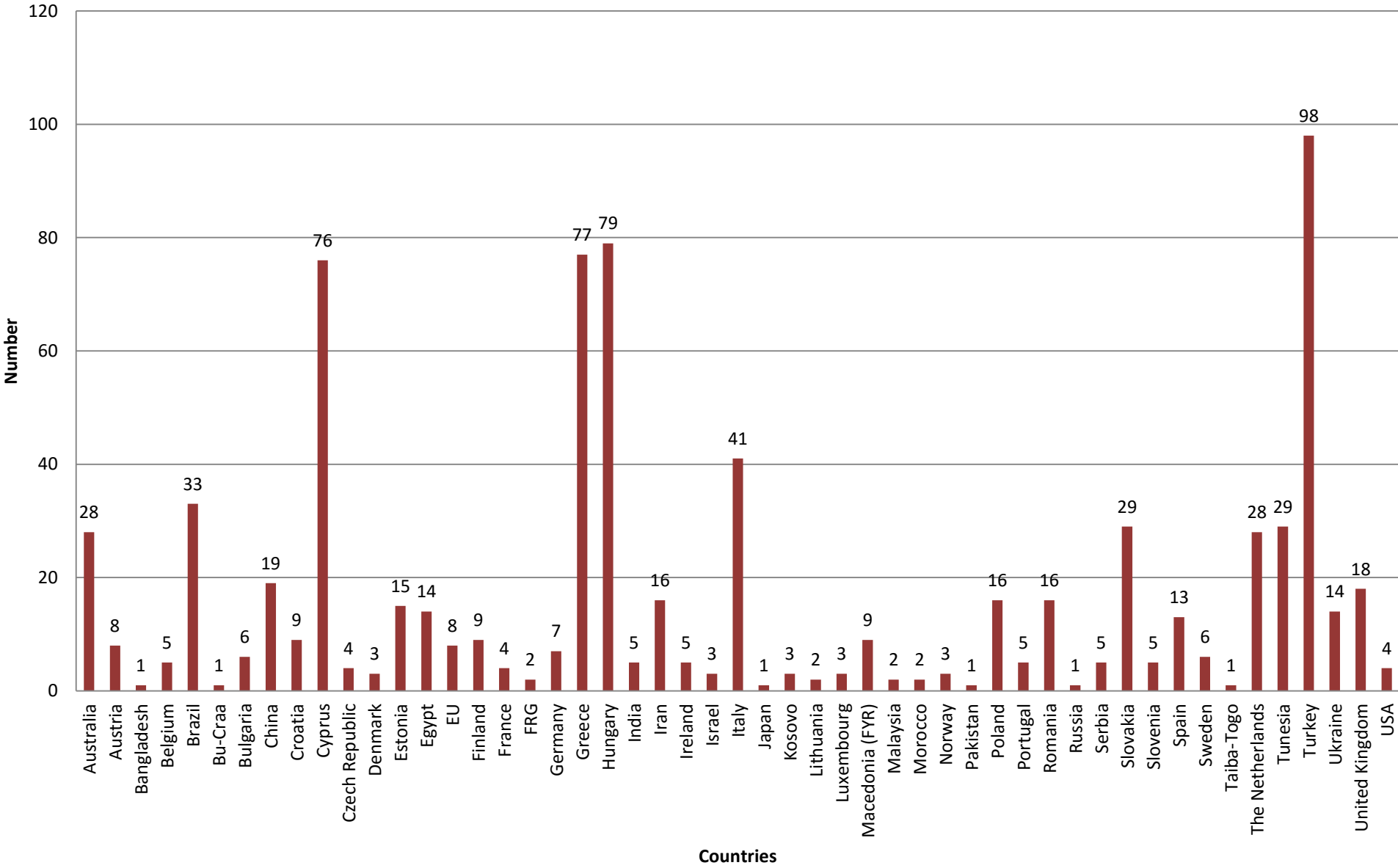
Impact:
-NORM aspects are taken into account for reuse
-’Good practices database’ as guideline for reuse for industry

NORM4BUILDING

cost

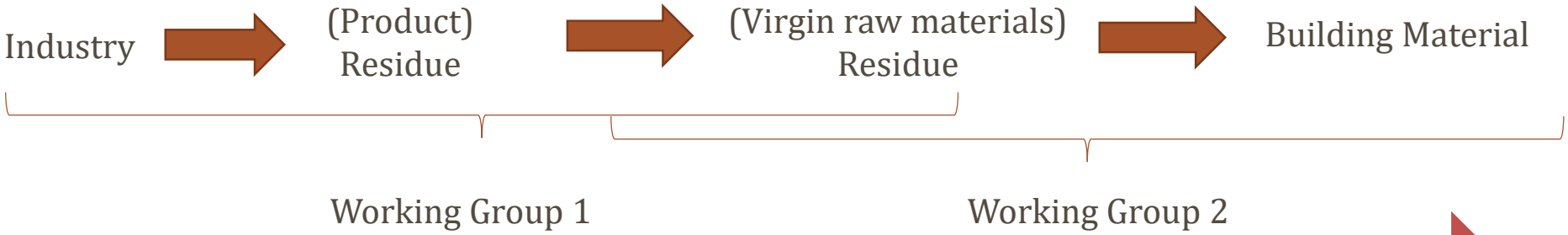
EUROPEAN SCIENCE FOUNDATION

Status database (total 792 entries, 9/10/2015)



Working Group 2

‘Options for new tailor-made building materials’

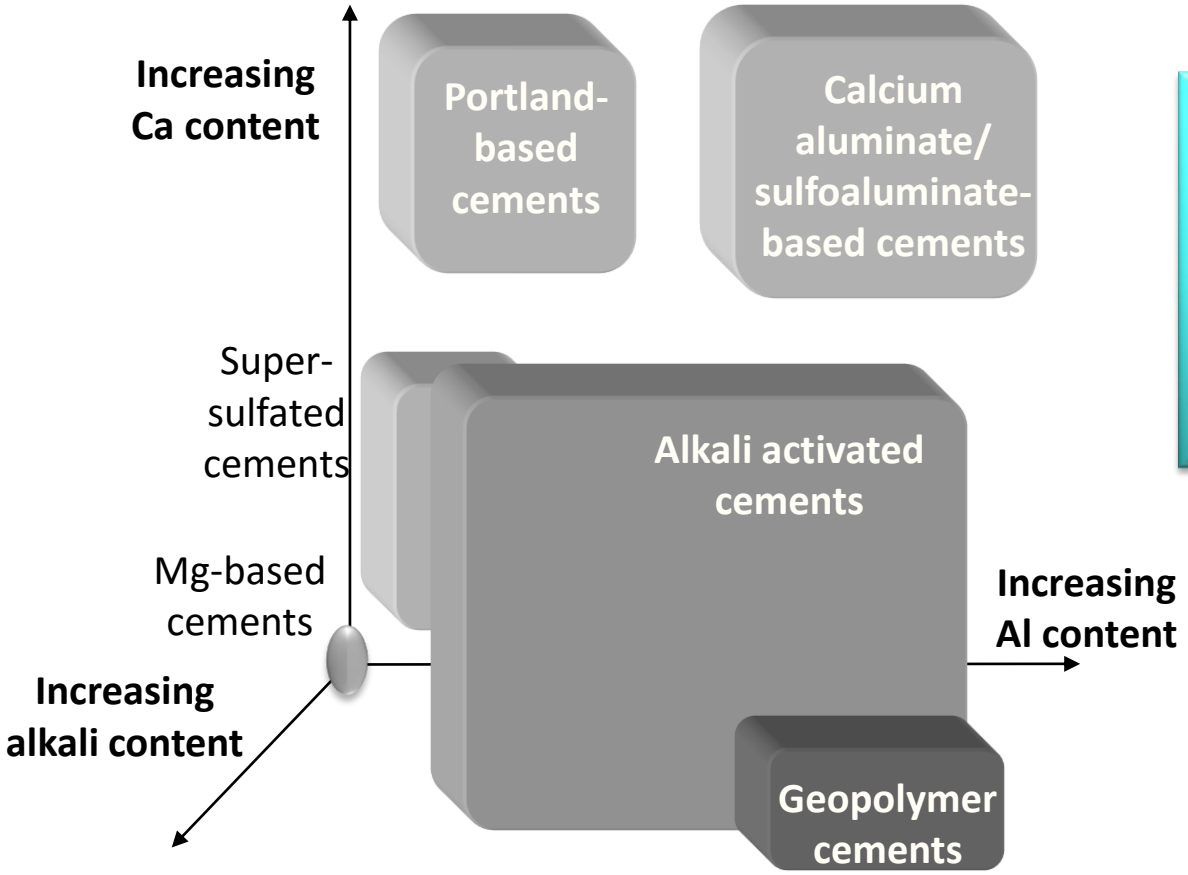


Study new develop options

a) Analytical model with **relation** between **% of virgin raw materials substituted** and **radiological content**

b) Recommendations on use **secondary raw materials** in **alkali activated cements considering the radiological content**.

Traditional and non-traditional cements



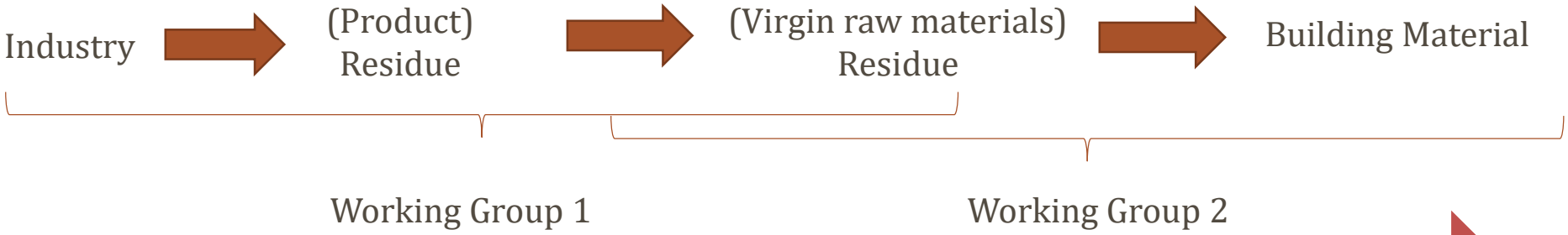
Moving from a single universal cement to an array of cement types

Most important: Designing materials that are fit for purpose!

J.L. Provis & J.S.J. van Deventer (eds.), State of the Art Report of RILEM TC 224-AAM, 2014

Working Group 2

‘Options for new tailor-made building materials’



Study new develop options

a) Analytical model with **relation** between **% of virgin raw materials substituted** and **radiological content**

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Radiological content linked to…

a) **Pretreatment** of the raw materials

b) **Development and application** of the building material

c) Effect of **inherent and engineered properties**

Many aspects determine the eventual reuse of 'byproducts'

- **Size of the byproduct stream**
- Properties of the byproduct
- **Environmental and health issues**
- Potential market and acceptance/**perception** aspects
- **Cost** aspects throughout the chain
- **Competition** with other byproducts
- CE marking and other certification aspects
- …

Working Group 3

‘Improve measurement capacity and standardisation’

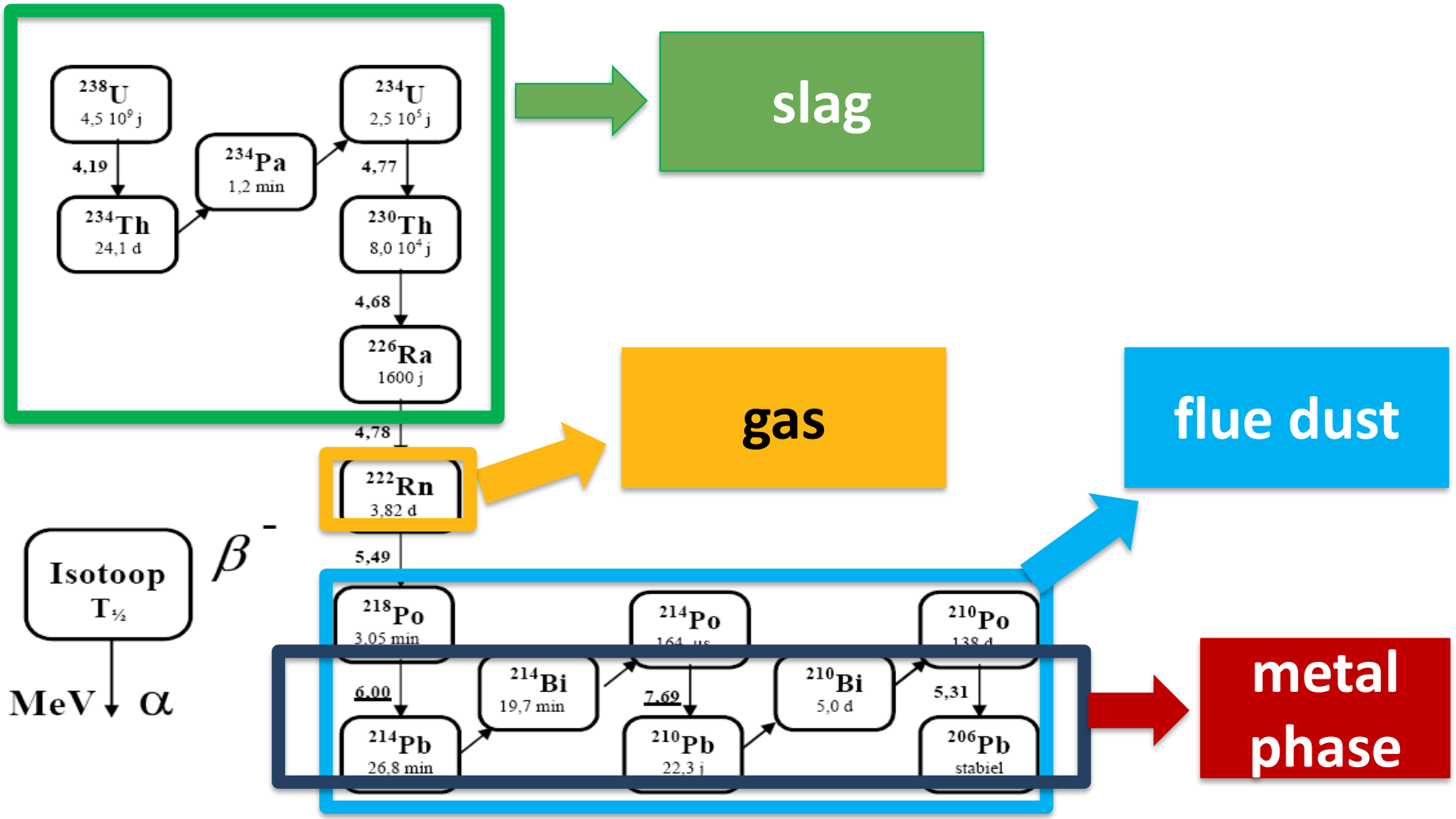
Validated (*on-site*) measurement protocols for

a) Activity Concentration Index

b) Radon (possibly thoron) emanation and exhalation rate

Intercomparisons using several measurements protocols and instruments

Metal processing: how radionuclides (U-238 decay chain) can behave during smelting?



(similar for decay chain of Th-232)

Approaches to in-situ measurements to determine the activity concentration

- conventional approach:
  1. **Spectra analysis calibrating broad spectral windows** during the analysis for the main natural isotopes
     - Activity concentration is determined from the net content of the window around individual peaks
     - Typical energy windows used to estimate the activity concentration for in-situ measurements:

	Radionuclide	Energy (keV)	Window (keV)
Potassium (^{40}K)	^{40}K	1460	1370-1570
Uranium (^{238}U)	^{214}Bi	1765	1660-1860
Thorium (^{232}Th)	^{208}Tl	2614	2410-2810

Approaches to in-situ measurements to determine the activity concentration

1. Spectra analysis calibrating broad spectral windows
   - Problems:
     - Blind to any unexpected signal (anthropic radionuclides).
     - Low accuracy for short time acquisitions
     - Physical restriction of poor intrinsic energetic resolution of the (mostly used) NaI(Tl) detector.
     - Assumption of secular equilibrium

Not valid

Approaches to in-situ measurements to determine the activity concentration

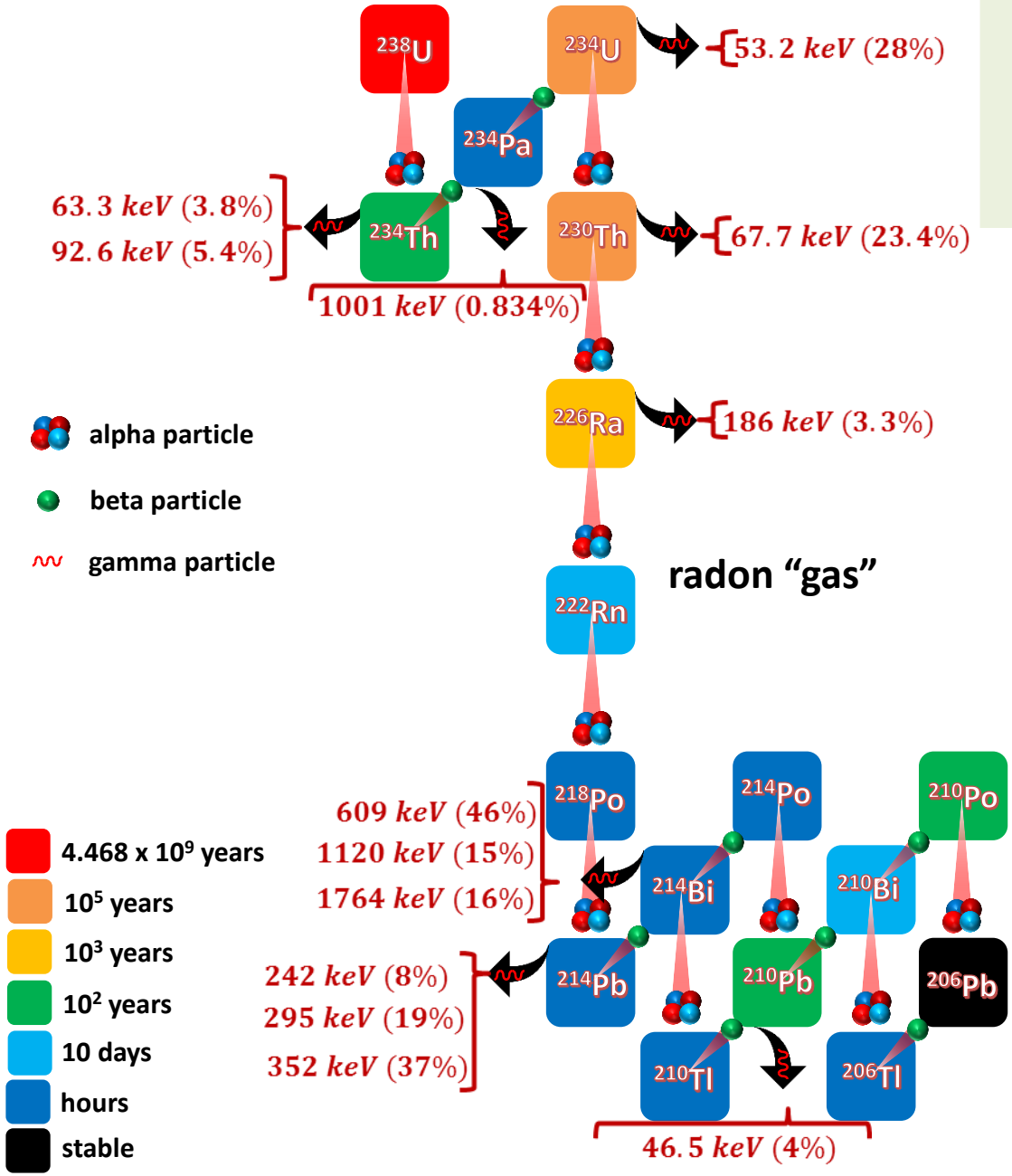
2. Full spectrum analysis method
   - The total spectrum is `unfolded' into the spectra for the individual radionuclides (the so-called standard spectra) and a background spectrum.
   - Standard spectra (of each investigated radionuclide) derived from the calibration procedure.

Hendriks et al. Journal of Environmental Radioactivity 53 (2001) 365-380

Many problems related to the measurement of NORM and NORM containing building materials

- **Disequilibrium** in decay chains
- Big **variation and heterogeneity** of the materials / components / matrices
- **Many radionuclides** (peaks) in one sample (spectrum)
  - Need for corrections for overlapping peaks
  - Some natural radionuclides have no gamma emission or very weak emission probability (e.g. Th-232, U-238, U-234, …)

^{238}U decay chain



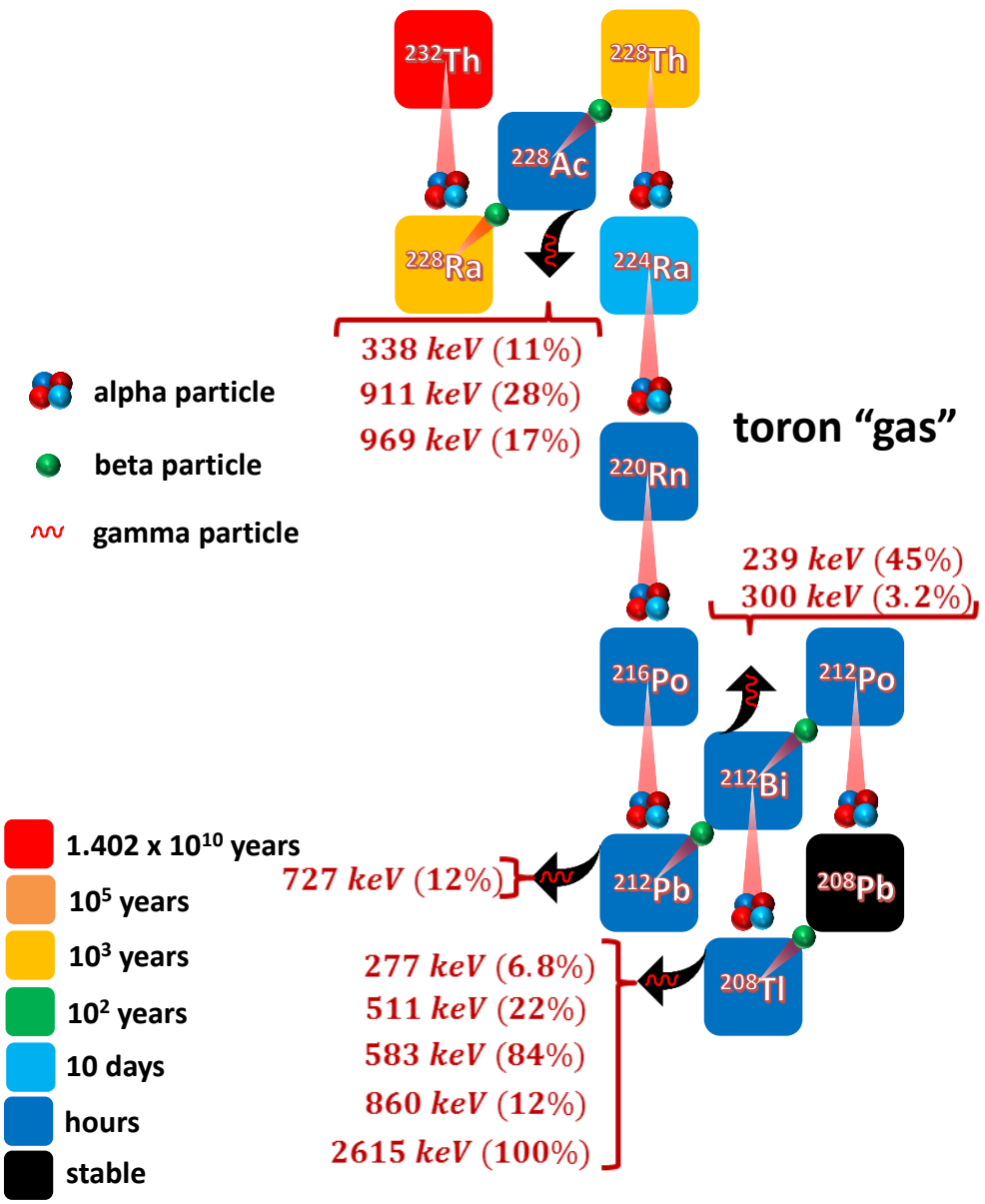
^{238}U decay through a series of alpha and beta decay to ^{206}Pb (stable)

$$^{238}U \rightarrow {}^{206}Pb + 8\,{}^{4}_{2}He + 6e^{-} + 6\bar{\nu}_{e}$$

^{232}Th decay chain

^{232}Th decay through a series of alpha and beta decay to ^{208}Pb (stable)

$$^{232}Th \rightarrow {}^{208}Pb + 6\,{}^{4}_{2}He + 4e^{-} + 4\bar{\nu}_{e}$$



Many problems related to measurement of NORM and NORM containing building materials

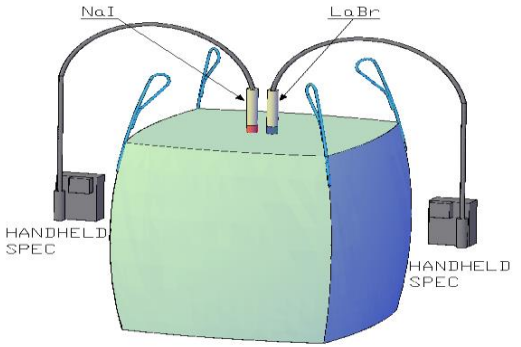
- High level of **expertise** & **instrumentations** required e.g. HPGe, alpha spectrometry
  - **Sample preparation** for alpha spectrometry

- **Sampling uncertainties**

- Need of fast measurement results for taking **fast decisions** e.g. compliance of raw material feed in an industrial process
  - ➔ role of on-site methods as screening tool

Problems related to measurement of NORM and NORM containing building materials

- When can we allow the use of on-site methods?
  - Only use them for specific, well defined applications (**screening tools**)
    - When the measured value (taking into account the uncertainty) is significantly below the screening level
  - Measure material in **well defined geometries** (big bags)
  - If possible measure **larger quantities** of material for a **longer time**
  - Need to **validate** the use of on-site methods
  - Use of **newer scintillation detectors** which generally show better energy resolutions,
    - $LaBr_3$:Ce, $CeBr_3$, BGO (Bismuth germanate), $CdWO_4$, $PbWO_4$



Working Group 3

‘Improve measurement capacity and standardisation’

Validated (*on-site*) measurement protocols for

a) Activity Concentration Index

b) Radon (possibly thoron) emanation and exhalation rate

Intercomparisons using several measurements protocols and instruments

Towards standardisation

a) Proposal for a **calibration procedure**

b) Steps in the development of **pre-standard materials**

Problem: good standards are missing

EURAMET
European Association of National Metrology Institutes

EMRP
European Metrology Research Programme
Programme of EURAMET

The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union

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Towards certification

a) Factsheet for **unified certification procedure of construction materials**.

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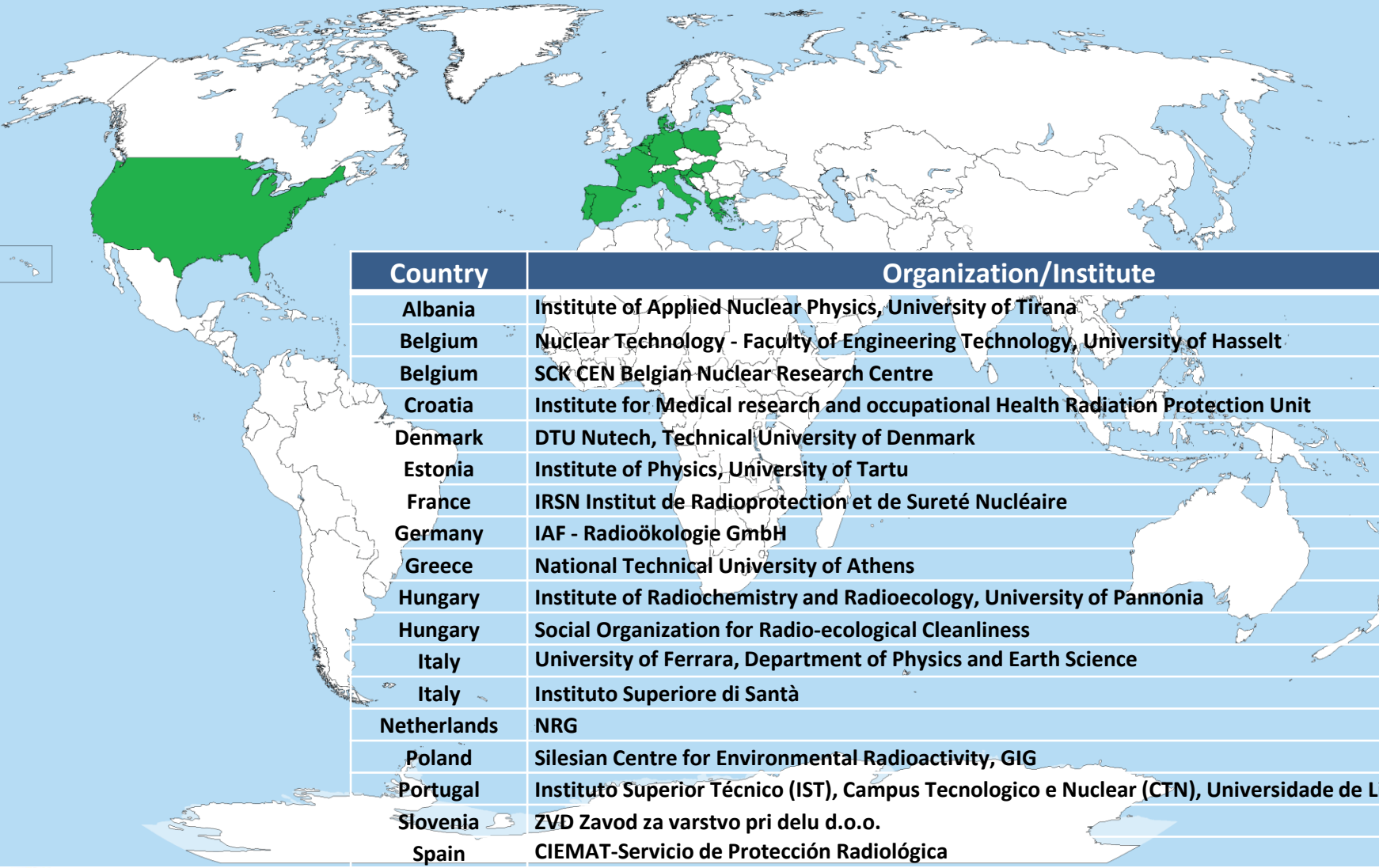
COST network: Intercomparison exercise



Country	Organization/Institute
Albania	Institute of Applied Nuclear Physics, University of Tirana
Belgium	Nuclear Technology - Faculty of Engineering Technology, University of Hasselt
Belgium	SCK CEN Belgian Nuclear Research Centre
Croatia	Institute for Medical research and occupational Health Radiation Protection Unit
Denmark	DTU Nutech, Technical University of Denmark
Estonia	Institute of Physics, University of Tartu
France	IRSN Institut de Radioprotection et de Sureté Nucléaire
Germany	IAF - Radioökologie GmbH
Greece	National Technical University of Athens
Hungary	Institute of Radiochemistry and Radioecology, University of Pannonia
Hungary	Social Organization for Radio-ecological Cleanliness
Italy	University of Ferrara, Department of Physics and Earth Science
Italy	Instituto Superiore di Santà
Netherlands	NRG
Poland	Silesian Centre for Environmental Radioactivity, GIG
Portugal	Instituto Superior Técnico (IST), Campus Tecnologico e Nuclear (CTN), Universidade de Lisboa
Slovenia	ZVD Zavod za varstvo pri delu d.o.o.
Spain	CIEMAT-Servicio de Protección Radiológica
Spain	CIEMAT-Unidad de Radiactividad Ambiental y Vigilancia Radiólogica
Spain	University of Huelva
Spain	University of Salamanca
Spain	University Autonoma de Barcelona
USA	Nuclear Engineering Teaching Lab, University of Texas

16 Countries

23 Labs

Working Group 4

‘Improving dosimetrical models’

Improved (more realistic) dosimetrical models

For use of NORMs in

a) Cement
b) Concrete
c) Ceramics

Specific focus on use of NORMs in **alkali activated cements**.

Working Group 4

‘Improving dosimetrical models’

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Specific focus on use of NORMs in **alkali activated cements**.

Evaluation of implementation on market.

a) **Round table discussions** with all stakeholders
b) **End-of-Life?**
   - Leachability?

WG4: evaluation of reuse of NORM in building materials

- Experimental building material related parameters
  ➔ modeling of impact building materials
  1. Gamma dose modelling
  2. Rn (and Tn) dose modelling
  3. Leachability/breakdown modelling of radiological and chemical impact

Too be published soon: **NORM4Building the book**…

Working Group 4

‘Improving dosimetrical models’

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Evaluation of legislation

a) **Impact EU-BSS**
b) Comparison of alternative national (EU-BSS based) legislative scenarios

And then the Belgians came…

- **FANC Decree of March 2013**

  →Addition treatment of NORM to **work activities**:
  "processing, valorisation and recycling of residues with an activity concentration > RP 122 II"

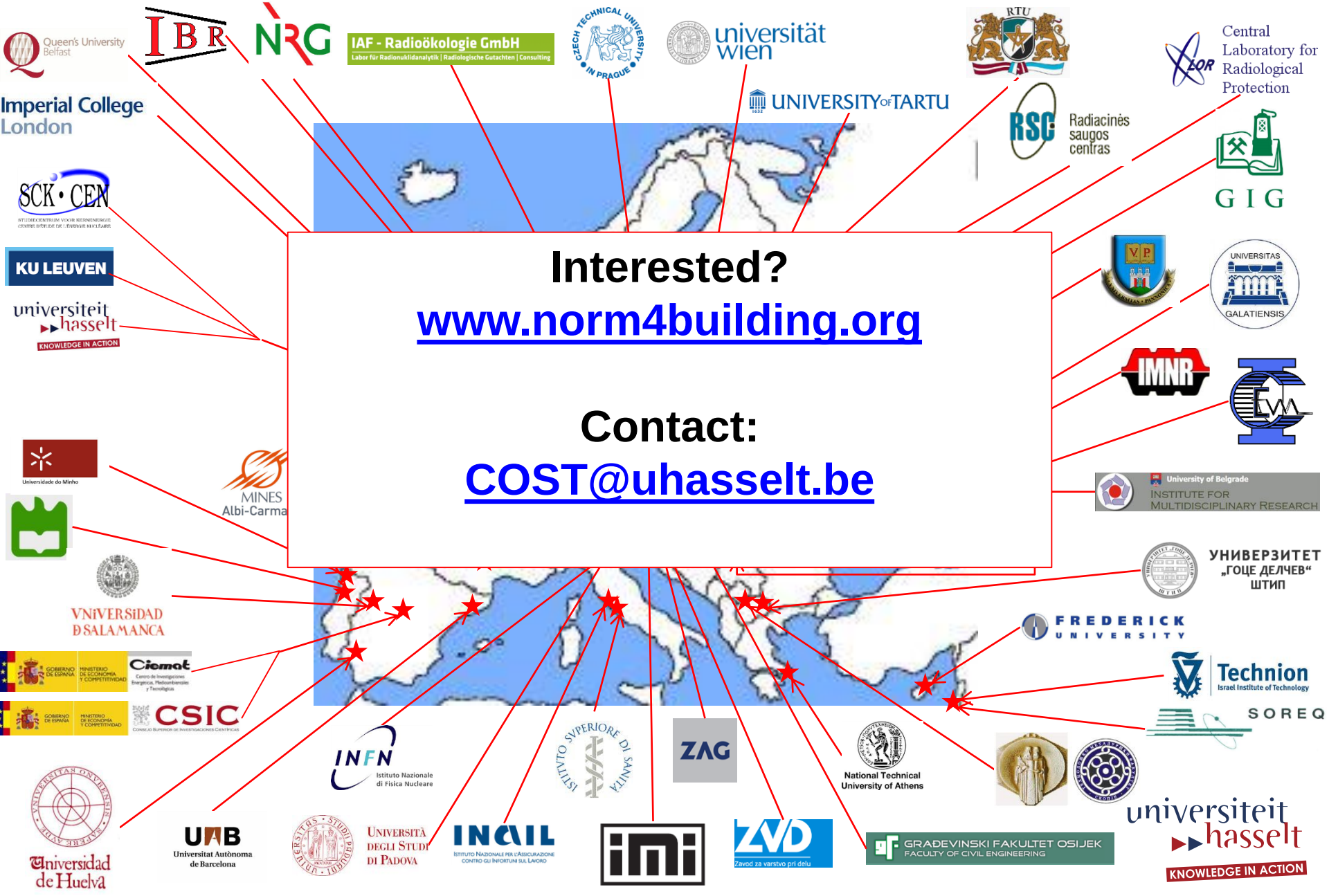
- Submits **NORM residue treatment** facilities to **notification**

Radionuclide	Activity concentration (Bq/g)
U-238sec (incl. U-235sec)	0.5
	0.1 (mono-landfill)
U nat	5
Th-230	10
Ra-226+	0.5
	0.1 (mono-landfill)
Pb-210+	5
Po-210	5
Th-232sec	0.5
	0.1 (mono-landfill)
Th-232	5
Ra-228+	1
Th-228+	0.5
K-40	5

Acceptance Criteria dependent on type of treatment (Belgium, FANC)

Treatment	Activity Concentration		
		Input (Waste producer)	Output (Waste Processer)
Building Materials	$C_{exemption}$	RP 122 II	Monitoring residues - Activity index (buildings) - RP 122 II (roads)
	C_{max}	10 Bq/g	

Current status COST Network; 110 experts 28 countries



Interested?

www.norm4building.org

Contact:

COST@uhasselt.be