### Strasbourg

### 27th June 2022

### 30<sup>th</sup> June 2022

Radiobiological Effects, Epidemiology & Health Impacts Risk assessment from low dose/rate chronic exposures Nuclear Power Plant Accidents & Risk Communication Handling of Human Exposures and Responsibilities Novel Radiation Protection Philosophy & Concepts Education Standards & Standards Education (ESSE) **Environmental Monitoring & Dose Assessment** Environmental Modelling for Radiation Protection High Level Natural Background Radiation Areas Radon, Thoron & Decay Products Measurements **Regulatory Control and Responsibilities** NORM & TENORM

10th ICHLERA

International Conference on High Level Environmental Radiation Areas M. Sohrabi, ICHLERA R. Barillon, Strasbourg University A. Nourreddine, Strasbourg University

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**İPHC** 

Faculté de physique et ingénierie Université de Strasbourg





# 10<sup>th</sup> International Conference on

## **High Level Environmental Radiation Areas**

27 – 30 June 2022

Strasbourg - France

### **Book of Abstracts and program**

Conference venue :

Amphi Grunwald Institut Pluridisciplinaire Hubert Curien (IPHC) CRONENBOURG campus 23 rue de Loess 67200 Strasbourg

### CONTENTS

Overview & welcome	P2
Organizing Committee	P4
International Committee	P5
Conference Program	P6
Abstracts	
Session 1 : Novel Radiation Protection Philosophy and Concepts	p13
Session 2 : Handling of Human Exposures and Responsibilities	P19
Session 3 : Impact of Environmental Exposure	P25
Session 4 : Environmental Measurements and Analysis	P31
Session 5 : Environmental Monitoring for Radiation Protection	P39
Session 6 : NORM & TENORM	p45
Special Olko Anniversary	
Session Ok 1 : From Natural core breeding to the Oklo discovery	P51
Session Ok 2 : From Oklo samples to natural core simulations	P57
Session 7 : Radon, Thoron & Decay Products Measurements	P63
Session 8 : Nuclear Power Plant Accidents & Risk Communication	P69
Session 9 : Radiobiological Effects, Epidemiology & Health Impacts	P79
Onsite Poster Session	P87
List of participants	P97

### **Overview & Welcome**

This Conference is jointly organized by the International Committee on High Level Environmental Radiation Areas (ICHLERA) and the Strasbourg University, which is also supported by the CNRS/IN2P3 (Institut National de Physique Nucléaire et de Physique des Particules), IPHC (Institut Pluridisciplinaire Hubert Curien) and Faculty Physique et Ingénierie of Strasbourg.

Historically, a chain of "International Conferences on High Levels of Natural Radiation and Radon Areas" has been held in the past in Brazil (1977), India (1981), Iran (1990), China (1996), Germany (2000), Japan (2004), India (2010), and Czech Republic (2014). These conferences have been on high levels of natural radiation and radon areas with themes to estimate risks of radiation-induced health-effects on public living in such areas. The International Committee on High Levels of Natural Radiation and Radon Areas (IC-HLNRRA) has been highly instrumental to facilitate, regularize and organize such periodic chain of conferences in cooperation with different countries for enhancing new radiological, radiobiological, and epidemiological data and novel radiation protection philosophies and concepts in order to better disseminate state-of-the-art information and to harmonize future studies for obtaining consistent and reliable radiation-induced health risk data needed in radiation protection.

At the 8<sup>th</sup> IC-HLNRRAs in Prague, Czech Republic (2014), it was decided to expand the scope of this chain of conferences for gaining broader information on the areas with environments contaminated due to natural and/or man-made radionuclides and installations forming exiting, planned and emergency exposure situations with emphasis on human health effects, epidemiology, and social impacts. In this context, the scope of such conferences was expanded to "International Conference on High Level Environmental Radiation Areas (IC-HLERA). Accordingly, the 9<sup>th</sup> IC-HLERAs was the first conference of this kind successfully held at the Hirosaki University, Japan (2018), for Understanding Chronic Low-Dose-Rate Radiation Exposure Health Effects and Social Impacts. At the last general assembly, it was decided to choose the Strasbourg University to host the 10<sup>th</sup> IC-HLERA. After two successive postponements in 2020 and 2021 due to the COVID-19 epidemic, we e are pleased to welcome you to Strasbourg and to the conference to be held from June 27 to 30, 2022 at IPHC-Cronenbourg Campus. This Book contains about 63 abstracts from 20 nationalities reporting on recent achievements on novel radiations, in particular radon and progeny, on health and environmental measurements with eminent Keynote Lecture, Invited Talks, as well as Oral and Poster Presentations under nine major topics :

- Novel Radiation Protection Philosophy and Concepts,
- Handling of Human Exposures and Responsibilities,
- Impact of Environmental Exposure,
- Environmental Measurements and Analysis,
- Environmental Monitoring for Radiation Protection,
- NORM & TENORM,
- Radon, Thoron & Decay Products Measurements,
- Nuclear Power Plant Accidents & Risk Communication, and
- Radiobiological Effects, Epidemiology & Health Impacts.

Moreover, on June 7<sup>th</sup> 1972, the first observation of an anomalous sample from the Oklo (Gabon) mine revealed one of the greatest secrets of Nature. This was a clear witness of the existence of sustained fission reactions in Nature. We will celebrate this year the 50 years of research about what we call it the "Oklo Phenomenon". For this reason, on June 28<sup>th</sup> afternoon, a special session in ICHLERA-10 will be dedicated to celebrate the Oklo'50<sup>th</sup> anniversary.

Chair of the ICHLERA Chair of the Organizing Committee Secretary of the Organizing Committee *June 2022* 

### **Organizing Committee**

#### **Executive Committee**

Prof. Abdel-Mjid NOURREDDINE	IPHC-Strasbourg University	Chairman
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Prof. Benoît GALL	IPHC-Strasbourg University	Oklo anniversary
Prof. Mehdi SOHRABI	Amirkabir University of Technology, Tehran	Chair of the ICHLERA

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S. TOKONAMI	Hirosaki University	Japan
T. TOMMASINO	Italian National Agency of Environmental Protection	Italy
A. WOJCIK	Stockholm University	Sweden

### **Conference Program**

### Monday 27<sup>th</sup> June 2022

08h00 – 09h00	Registration
09h00 - 9h15	Opening address :
	A. NOURREDDINE., Chair of organizing committee
	R. BARILLON, Vice President Strasbourg University
	S. COURTIN, Director IPHC Strasbourg
	M. SOHRABI, ICHLERA President
Session 1: Novel	Radiation Protection Philosophy and Concepts - <b>Chair :</b> <i>L. TOMMASINO</i>
9h15 - 10h00	Keynote lecture : M. SOHRABI (Tehran, Iran)
	Modern universal standardized trends in worker and public exposure
	monitoring and control in 21 $^{ m st}$ century by Sohrabi URPS-based hypothesis
10h00 - 10h15	Invited talk 1.1: F. SHANNOUN (UNSCEAR, Vienna, Austria) *
	The United Nations Scientific Committee on the Effects of Atomic Radiation
	current programme of work and projects related to low dose and low-dose
	rate
10h15 - 10h45	Invited talk 1.2: S. BOUFFLER (UKHSA, London, United Kingdom) *
	UNSCEAR 2020/2021 Report on Biological mechanisms relevant for the
	inference of cancer risks from low-dose and low-dose-rate radiation
10h45 - 11h15	Coffee Break + Onsite Poster Session
Session 2 : Handlin	ng of Human Exposures and Responsibilities - Chair: M. BALCAZAR
11h15 - 11h45	Invited talk 2: J. Mc LAUGHLIN (Dublin, Ireland)
	Public and expert perceptions of risks from radiation
11h45 - 12h00	Talk 2.1 : L. LUZZI (Roma, Italy)
	Inventory of NORM-related industrial sectors in Italy: preliminary results
12h00 - 12h15	Talk 2.2 : P. BOSSEW (Berlin, Germany)
	True and spurious anomalies in ambient dose rate monitoring
12h15 - 12h30	Talk 2.3 : N. MARTIN-BURTART (Mississauga, Canada)
	Calibration of radiation portal Monitors for characterization of historic low-
	level radioactive waste
12h30 - 14h00	Lunch
Session 3 : Impact	of Environmental Exposure – <b>Chair :</b> <i>M. SOHRABI</i>
14h00 - 14h30	Invited talk 3: V. BRETON (Clermont-Ferrand, France)
	Impact of natural radioactivity on microorganisms
14h30 - 14h45	Talk 3.1 : A. NADHIYA (Hulhumail, Maldives) *
	Determination of heavy metal level in Tuna fishes from Laccadive sea and
	their associated health risk
14h45 - 15h00	Talk 3.2 : G. HOLUB (Bordeaux, France)
	Bacterial diversity characterization of naturally radioactive mineral springs:
	Role of these bacterial communities in uranium biogeochemistry

15h00 - 15h15	<b>Talk 3.3 :</b> <i>S. SINGH (Amritsar, India)</i> High prevalence of uranium in groundwater of Punjab: A comparative study
15h15 - 15h30	<b>Talk 3.4 :</b> <i>S. KOLOVI</i> (Clermont-Ferrand, <i>France</i> )         Radioactive mineral springs: A first approach of modelling the radiation         exposure on microorganisms
15h30 - 15h50	Coffee Break + Onsite Poster Session
Session 4 : Enviro	nmental Measurements and Analysis - <b>Chair :</b> J. Mc LAUGHLIN
15h50 - 16h20	Invited talk 4: M. DEL NERO (Strasbourg, France)
	Linking speciation and soil-water-plant transfers of natural radionuclides in a
	wetland impacted by a former U mine: recent advances from the pluri-
	disciplinary and multi-partners project INSPECT-NEEDS
16h20 - 16h35	Talk 4.1: G.D. TEBA (Jena, Germany)
	Biogeochemical behaviour of tellurium (Te) and caesium (Cs) at the continent- ocean interface: preliminary scenarios for accidental radionuclide releases
16h35 - 16h50	Talk 4.2 : S. GUO (Strasbourg, France)
	In situ ATR - FTIR study of uranyl sorption at illite - solution interface in the presence of phosphate ions
16h50 - 17h05	Talk 4.3 : B. HERNÁNDEZ MÉNDEZ (Edo. de México, Mexico) *
	Environmental radiological baseline for unconventional oil and gas zones in Mexic
17h05 - 17h20	Talk 4.4 : B. SALAS (Mexico City, Mexico) *
	Fraudulent exams in the training of reactor supervisors at the Laguna Verde
	Nuclear Power Plant- México
17h20 - 17h25	P 4.1 : Radioactive waste management without adherence to standards at the
	Laguna Verde Nuclear Power Plant, Mexico
18h30 -	Welcome reception at the Hotel de Ville

#### Tuesday 28th June 2022

Session 5 : Environmental Monitoring for Radiation Protection – Chair : V. BRETON

nvited talk 5: L. TOMMASINO (Roma, Italy)
Radon measurements indoors, in soil and in water by track detectors and/or
by Geiger-Muller counters
Talk 5.1: H. HASHIMOTO (Hirosaki, Japan) *
Development of a portable Type personal dosimeter for internal and external
lose assessments
<b>Falk 5.2 :</b> Y. TAMAKUMA (Nagasaki, Japan) *
Site-specific dose conversion factors for radon progeny based on actual
aerosol size distributions at various environments
Talk 5.3 : VEERASAMY (Tokyo, Japan) *
Determination of 226Ra/238U and 228Ra/232Th disequilibrium in surface soils
from HBRA, Odisha, India and its radiological implications

10h15 - 10h30	<b>Talk 5.4 :</b> <i>F. LEONARDI (Roma, Italy)</i> Radon spatial and seasonal variations in University's buildings located in an Italian karst region
10h30 - 10h45	Coffee Break + Onsite Poster Session
Session 6: NORM	& TENORM – <b>Chair :</b> S. <i>TOKONAMI</i> (Hirosaki, Japan)
10h45 - 11h15	<b>Invited talk 6 :</b> <i>M.U. KHANDAKER (Selangor, Malaysia)</i> * NORMs in cultivated honey in Malaysia and concomitant dose to the consumers
11h15 - 11h45	<b>Talk 6.1:</b> R.C. RAMOLA (Tehri Garhwal, India) *Estimation of radiation dose due to Thoron and progeny inhalation in highBackground natural radiation area of Eastern Coastal Area of Odisha, India
11'45-1200	<b>Talk 6.2 :</b> <i>M. BALCAZAR (Ocoyoacac, Mexico)</i> Good RAD7 performance for high radon levels
11'45-1200	<b>Talk 6.3 :</b> <i>S. RATNAYAKE</i> (Pitakotte, Sri Lanka) Radiation Dose Assessment and Mobiliy Studies of Thorium in Soil from a High Level Natural Background Radiation Area in Sri Lanka
12h -	
Session Ok 1: Fro	m Natural core breeding to the Oklo discovery - <b>Chair :</b> B. GALL
14h00 - 14h20	<b>Talk Ok 1.1:</b> F. GAUTHIER-LAFAYE (Strasbourg, France)The Oklo phenomenon, discovery, first questions, first answers
14h20 - 14h40	<b>Talk Ok 1.2 :</b> J.F. DOZOL (Cadarache, France)From routine sample measurements in CEA to the Oklo phenomenon
14h40 - 15h00	Talk Ok 1.3 : J.C. NIMAL (Paris, France) * Historical simulations of Oklo cores
15h00 - 15h20	<b>Talk Ok 1.4 :</b> <i>D. LOUVAT (Paris, France)</i> Oklo natural analogue of radioactive waste disposal, summary of European Commission projects'results (1991-1999)
15h20 - 15h40	<b>Talk Ok 1.5 :</b> <i>S.E. BENTRIDI (Khemis-Miliana Algeria)</i> Influence of Initial poisons and clays on the criticality of Oklo natural nuclear reactors
15h40 - 16h10	Coffee Break + Onsite Poster Session
Session Ok 2 : Fro	m Oklo samples to natural core simulations - <b>Chair :</b> F. GAUTHIER-LAFAYE
16h10 - 16h30	<b>Talk Ok 2.1 :</b> A. EL ALBANI (Poitiers, France) The Gabonionta: Great Oxidation Event, reactors and Life
16h30 - 16h50	<b>Talk Ok 2.2 :</b> A. MESHIK (Washington, USA) Natural Nuclear Reactors: prediction, search, discovery, operation and implications
16h50 - 17h10	<b>Talk Ok 2.3 :</b> <i>H. HIDAKA (Nagoya, Japan)</i> Evidences of fast neutron operating in Oklo
17h10 - 17h30	<b>Talk Ok 2.4 :</b> E.D. DAVIS (Kimberley South Africa) *
17h30 - 17h50	<b>Talk Ok 2.5 :</b> <i>B. GALL (Strasbourg, France)</i> Parallel between natural Oklo cores and industrial reactors operating

### Wednesday 29th June 2022

Session 7 : Radon, Thoron & Decay Products Measurements

9h00 - 9h30	<b>Invited talk 7.1:</b> <i>S. TOKONAMI (Hirosaki, Japan)</i> Up-to-date dose conversion factors for radon isotopes and their historical
9h30 - 10h00	<b>Invited talk 7.2:</b> <i>L. FONT GUITERAS (Barcelona, Spain)</i> On the use of numerical models to predict/mitigate indoor radon levels in highly contaminated areas
10h00 - 10h15	Talk 7.1: C. DI CARLO (Roma, Italy)         Proposal of an affordable method to estimate indoor thoron concentration         close to the walls using active radon monitors
10h15 - 10h30	Talk 7.2: M. SOHRABI (Tehran, Iran)         Some Applications of novel polycarbonate/ACF radon detectors for personal and environmental Monitoring
10h30 - 10h50	Coffee Break + Onsite Poster Session
Session 8 : Nuclea	r Power Plant Accidents & Risk Communication - <b>Chair :</b> M. DEL NERO
10h50 - 11h20	Invited talk 8: F. BOCHICCHO (Roma, Italy)
	Exposure of people living around nuclear power plants and other nuclear facilities
11h20 - 11h35	Talk 8.1:       G. DOUGNIAUX (Paris, France)
	Detection limit variation against coarse aerosol during airborne contamination measurement with CAM
11h35 - 11h50	Talk 8.2: M. JEBBAD (Safi, Morocco) *
	Grain size effect on radon exhalation rate and uranium activity of Moroccan sediment
11h50 - 12h05	Talk 8.3 : N. ARBOR (Strasbourg, France)
	A drone-borne gamma spectrometry system for environmental radioactivity monitoring
12h05 - 12h20	Talk 8.4 : L. STUTTGE (Strasbourg, France)
	Development of a new material for neutron detection
12h20 -12h25	<b>P8.1:</b> <i>K. IWAUKA</i> ( <i>Chiba, Japan</i> ) <sup>a</sup> Calculation tool for iodine 131 biodistribution depending on the aerosol size distribution
12h25 -12h30	P 8.2 : C. KRANROD (Hirosaki, Japan) *
	Preliminary study on radon level, ambient aerosol, and external gamma radiation dose rate at lung cancer area in Northern. Thailand
12h30 – 12h35	P 8.3: Y. OMORI (Hirosaki, Japan) *
	Evaluation method of natural gamma dose rates using an Nal(Tl) spectrometer under coexistence of radioceasium and natural radionuclides
12h35 – 12h40	
12h30 -	Lunch
15h45 - 17h30	Guided Boat Tour (Strasbourg city)
20h00 - 23h00	Gala dinner (Maison Kammerzell)

#### Thursday 30th June 2022

Session 9: Radiobiological Effects, Epidemiology & Health Impacts - Chair: L. FONT GUITERAS

9h00 - 9h15	Talk 9.1: P. BEAUDIER (Bordeaux, France)
	Towards the characterization of single-cell molecular response to ionizing
	radiation
9h15 - 9h30	Talk 9.2 : B FRANGIONE (Ottawa, Canada)
	Maternal and paternal exposure to low dose radiation and adverse birth
	outcomes: preliminary findings from a systematic review and meta-analysis
9h30 - 9h45	Talk 9.3 : M. IDLIL (Rabat, Morocco)
	Low dose rate radiation exposure effects among medical workers from the
	knowledge of epidemiologic studies
9h45 - 10h00	Talk 9.4: C. ELHASSANE (Algiers, Algeria) *
	Ionising radiation exposure of Medical staff: risks and prevention
10h00 - 10h15	Talk 9.5 : Q. RAFFY (Strasbourg, France)
	Radiolysis of concentrated myoglobin by accelerated ions
10h15 - 10h30	Talk 9.6: A DANVIN (Strasbourg, France)
	Effects of the dose-rate on the radiolysis of water and small protein
	biomolecules
10h30 - 10h45	Coffee Break + Onsite Poster Session
10h45 - 12h00	General assembly
12h00	Closing remarks

#### **Onsite Poster Session**

**P1:** A. ARNONE (Strasbourg, France)

Radiolysis of a small peptide in solution by accelerated ions, electrons and X-rays

**P 2 :** A. BELAFRITES (Jijel, Algeria)

Assessment of radioactivity levels and radiological hazard indices in phosphate samples from Algeria

**P3:** S. CHEFSON (Strasbourg, France)

Comparison of experimental and Geant4-DNA simulated data of yields of water radiolysis species

**P4:** S. FERRERES (Strasbourg, France)

Study of the mobility of uranium (VI) and organic matter of a soil from an enhanced natural radioactivity area.

**P 5 :** C. GALINDO (Strasbourg, France)

Mechanistic study of Myoglobin and Apo-Myoglobin radiolysis by accelerated ions

**P 6 :** S. GEORG (Strasbourg, France)

Study of the speciation and mobility of uranium (VI) in natural waters: Effect of organic matter

P7: F. LEONARDI (Roma, Italy)

Spatial variability in large buildings of university campus: a case-study to identify the best measurement protocols

#### **P9:** Y. OMORI (Hirosaki, Japan)

Evaluation method of natural gamma dose rates using an NaI(TI) spectrometer under coexistence of radioceasium and natural radionuclides

**P8:** J. RIFFAUD (Strasbourg, France)

Assessment of Potential Activation in Food Products Irradiated with High Energy X-Rays: Experimental and Modelling Approach

### Session 1

### Novel Radiation Protection Philosophy and Concepts

**Chair** Luigi TOMMASINO (Roma, Italy)

#### **Keynote Lecture**

#### Modern Universal Standardized Trends in Worker and Public Exposure Monitoring and Control in 21<sup>st</sup> Century by Sohrabi URPS-based Hypothesis

#### Mehdi SOHRABI

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The Universal Radiation Protection System (URPS) has been hypothesized by Sohrabi in order to address the many deficiencies existing in current worker and public exposure monitoring and control<sup>[1-3]</sup>. This paper while briefly presents the philosophy, concepts, and methodologies of the URPS hypothesis, it emphasizes and proposes a standardized global worker and public exposure monitoring system. A worker is committed lifetime since birth to non-occupational exposures in particular from unfractionated natural background (NBG) radiation as a member of public. According to URPS, a "worker" is a member of public committed to also to non-occupational exposure lifetime plus occupational exposure from radiation work"<sup>[1,2]</sup>. Accordingly, a worker and public currently suffer from lacking standardized trends on; (a) risk estimates and accepted risk model, (b) dose limit, (c) correct personal dosimetry, (d) occupational fractionated exposures, and (e) integrated dose system. The URPS hypothesizes equal human heath-effect risks per unit radiation dose either from natural or man-made sources; integrates all individual doses; considers worker as member of public; conserves cause and effects for risk estimation; uses fractionation weighting factors; proposes a "URPS Model" for bridging "linear no-threshold and hormesis models, formulates an example dose limit for worker and public, and provides new exposure definitions<sup>[1-9]</sup>. By considering these concepts and methodologies and in order to standardize workers and public exposure towards integration of doses, individual exposure monitoring and control philosophy, concepts and methodologies are proposed and formulated compared to those of ICRP<sup>[10]</sup>. The proposed standardized methods resolve current individual exposure monitoring and control deficiencies to better protect human beings in 21<sup>st</sup> century worldwide, independent of sources of radiation and country of origin.

#### References

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- [2]. Sohrabi, M., "Universal Radiation Protection System (URPS); A Natural Global Standardized Trend for Human Exposure Control in 21<sup>th</sup> Century", Radiat. Prot. Dosimetry. May (2019), doi:10.1093/rpd/ncz097.
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- [4]. Sohrabi, M., "On Dose Reconstruction for the Million Worker Study: Status and Guidelines", Health Phys. 109, 327-329 (2015).
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- [6]. Sohrabi, M., Editorial, "Dose Fractionation Concept in Radiation Protection to Standardize Risk/Dose Limits and Epidemiology Studies", J. Epidemiol. Public Health Rev. 2(4) (2017).
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- [8]. Sohrabi, M., "Bridging the LNT and Hormesis Radiation Protection Models", Nuclear News, pp.37-42, June (2018).

- [9]. Sohrabi, M. Eyes Should be Washed for Global Change in Radiation Protection of 21<sup>st</sup> Century. Health Physics 120 (4), 455-458) (2021).
- [10]. International Commission on Radiological Protection. The 2007 recommendations of the international commission on radiological protection. Publication 103, Ann. ICRP 37(2–4), Elsevier (2007).

#### **Invited Talk 1.1**

The United Nations Scientific Committee on the Effects of Atomic Radiation current programme of work and projects related to low dose and low-dose rate

#### Borislava BATANDJIEVA-METCALF and <u>Ferid SHANNOUN\*</u>

United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), Vienna International Centre, 5 Wagramerstrasse, 1400 Vienna, Austria

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In 1955, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) was established by the United Nations General Assembly to assess and report on the levels, effects and risks of exposure to ionizing radiation for humans and the environment. The Committee is composed today of scientists from 31 Member States of the United Nations.

Its reviews are the scientific basis for national and international recommendations and standards for the protection of people and the environment from ionizing radiation.

The Committee's programme of work is endorsed by the United Nations General Assembly. The Committee compiles regularly relevant data submitted by United Nations Member States, international organizations and non-governmental organizations, as well as in the peer-reviewed scientific literature, and engages specialists to analyze those data, to study relevant scientific topics and to produce scientific evaluations. These authoritative reviews are published as scientific annexes to the Committee's report to the United Nations General Assembly.

The Committee's current programme of work envisages the publication of evaluations on medical, occupational and public exposure. The medical and occupational exposure evaluation are recently finalized, while in 2020, the Committee launched its public exposure evaluation including exposure from natural and artificial sources. This evaluation is intended to be completed by 2024. Also recently, the Committee has finalized its evaluation on biological mechanisms relevant for the inference of cancer risks at low doses and low dose-rates. The outcomes are presented in this conference in a dedicated paper, while this paper will focus on the Committees' current programme of work and future plans with regard to projects dealing with low dose and low dose rate radiation.

For further information please consult the new published UNSCEAR website [1].

#### References

[1]. https://www.unscear.org/unscear/en/index.html

#### **Invited Talk 1.2**

#### UNSCEAR 2020/2021 Report on Biological mechanisms relevant for the inference of cancer risks from low-dose and low-dose-rate radiation

#### <u>Simon BOUFFLER<sup>1</sup></u>, Serge CANDÉIAS<sup>2</sup>, Markus EIDEMÜLLER<sup>3</sup>, Prakash HANDE<sup>4</sup>, Leon MULLENDERS<sup>5</sup>, Gayle WOLOSCHAK<sup>6</sup>

on behalf of UNSCEAR <sup>1</sup>UK Health Security Agency, UK; <sup>2</sup>Commissariat à l'énergie atomique et aux énergies alternatives, France; <sup>3</sup>Helmholtz Zentrum München, Germany; <sup>4</sup>National University of Singapore, Singapore; <sup>5</sup>Leiden University Medical Centre, The Netherlands; <sup>6</sup>Northwestern University, USA.

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In 2016, the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) established an Expert Group on biological mechanisms relevant for the inference of cancer risks at low doses and low dose-rates. The Committee defines low doses as those of 100 mGy (low LET radiation) or less and low dose-rates, those of 0.1 mGy/minute (low LET radiation) or less. The Expert Group undertook a systematic review of the relevant literature to identify relevant studies, each study was evaluated in terms of is relevance to the report and quality. Specific mechanisms/endpoints considered in the report include DNA damage; DNA damage signalling, chromatin remodelling and epigenetics; effects on other signal transduction pathways; gene and protein expression; DNA repair and effects on somatic cells; genomic instability, bystander effects, damage/effects on non-nuclear cellular components, adaptive response and hyper-radiosensitivity; stem cells and target cell populations for radiation carcinogenesis; effects at the whole organism level, including effects on the immune system. The report concludes that, while complete understanding of the mechanisms and modulators of carcinogenesis following low-dose and low-dose-rate radiation exposures is not yet available, little in the way of robust data could be identified that would prompt the need to change the current approach taken for low-dose radiation cancer risk inferenceas used for radiation protection purposes. The potential contributions of phenomena such as transmissible genomic instability, bystander phenomena and adaptive response remain unclear. Some studies indicate that low-dose and low-dose-rate exposures can extend lifespan and possibly reduce tumour burdens in experimental animals; however generally, there is insufficient mechanistic understanding of these observations. There is evidence emerging that low dose exposures can stimulate tumour vascularisation. Overall, the Committee concluded that there remains good justification for the use of a non-threshold model for risk inference given the robust knowledge on the role of mutation and chromosomal aberrations in carcinogenesis. Looking to the future, the Committee recommended an approach that combines mechanistic understanding of low dose radiation carcinogenesis with epidemiological studies through the use of mathematical modelling integrating data from experimental systems. Furthermore, the UNSCEAR 2020/2021 Report, Annex C [1] includes an appendix setting out quality criteria to be taken into account in evaluations of experimental studies of radiation exposure.

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### Session 2

### Handling of Human Exposures and Responsibilities

Chair

Miguel BALCAZAR (Ocoyoacac, Mexico)

#### **Invited Talk 2**

#### Public and Expert Perceptions of Risks from Radiation

#### James Mc LAUGHLIN

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As shown by public reactions to nuclear accidents in the past, such as occurred in Chernobyl in 1986 and in Fukushima in 2011, there are considerable differences or gaps between how the public and how radiation experts perceive the risks from radiation. The reasons for these differences are complex and involve not only technical issues but also sociological ,ethical and behavioural aspects of society. Some of these reasons are discussed in this account as well as the influence they can have on the effectiveness of radiation risk communication strategies targeted at the public.

Even amongst radiation experts differences in radiation risk perceptions can be present. The belated modifications to the safety features of the Windscale nuclear reactor in the UK during its construction in the early 1950s is a good example of this. When the reactor went on fire in 1957 these belated safety modifications fortunately ensured that atmospheric releases of radionuclides to the environment and their health impact were much lower than would have occurred without them. This and other examples of importance to radiation protection such as differences between the way some radiation protection authorities dealt with environmental contamination following the Chernobyl accident are also discussed in this account.

#### Talk 2.1

Inventory of NORM-related industrial sectors in Italy: preliminary results

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A research project is ongoing in order to develop technical and scientific tools for supporting Italian stakeholders of the NORM-related industrial sectors in order to comply the provisions of the Italian transposition [1] of the EU BSS [2].

In the framework of the project, a task is focused on the collection of information concerning the Italian NORMrelated industrial sectors: indeed, the creation of a national NORM-related industry inventory is a good starting point for the evaluation of the effectiveness of the new legislation for the radiological protection of workers and members of the public. A first Italian inventory of NORM-related industries was carried out in 2014 [3]: now, based on that experience, a revision is in progress accounting for the EU BSS provisions and those given by new Italian Decree issued in 2020. Moreover, an updated inventory is a useful tool to improve the national strategy about the management of NORM residues.

In particular, for each NORM-related industrial sector, a data sheet is prepared in order to collect information about:

- Number of companies and factories, their distribution on the national territory
- Number of workers (if available)
- Description of the industrial process
- Most significant exposure scenarios
- Exposure pathways (external gamma radiation/intake of radionuclides by inhalation/accidental ingestion)
- Raw materials; description, radiological characterization
- Residues: description, radiological characterization
- Doses to workers
- Doses to members of the public

It is worth noting that the Italian Decree, if compared to the former legislation, considers a greater number of NORM-related industrial sectors to take into account the list of industrial sectors given in the EU BSS, such as geothermal energy production plants and coal power plants. Preliminary results are presented and discussed in this work.

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#### Talk 2.2

True and spurious anomalies in ambient dose rate monitoring

#### Peter BOSSEW<sup>1</sup>, Petr KUČA<sup>2</sup>, Jan HELEBRANT<sup>2</sup>

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A simple way of areal monitoring of ambient dose rate (ADR) is ground surveying carrying a dose rate meter coupled to GPS which logs measured ADR values together with geographical coordinates and date/time. This is the method used by the Citizen Science project *Safecast* [1], devoted, among other, to ADR mapping. The project has been founded shortly after the Fukushima accident 2011 in Japan and soon spread world-wide. Several thousand citizens contribute as volunteers, carrying detectors and sending data to Safecast.org, where they are presented in a map [1]. The project uses a standard device called *bGeigie Nano* (based on a GM detector), but in principle also other monitors of similar type can be used. For the sake of usability, such instruments are small and therefore sensitivity is relatively low.

Citizen monitoring involves a number of particular quality assurance (QA) issues, apart from conventional metrological QA. The former aspect has been addressed in [2], the latter in [3], both in [4] together with a statistical evaluation of ADR results from cities world-wide.

Usually, the detectors are carried in pockets, back bags, on bicycles or in cars near windows. Depending on the travelling speed and measurement period (5s for the *bGeigie Nano*), ADR anomalies of certain spatial extension can be detected. In order to generate not too noisy ADR maps, measurement periods are aggregated (12 periods for the *bGeigie Nano*), because individual periods usually contain only few counts, which however reduces spatial resolution.

This algorithm gives rise to spurious anomalies, i.e. ones generated by the measurement process. With some frequency, GM tubes can generate extreme count numbers per period, which denote count numbers largely exceeding what can be expected from Poisson count statistics. Such phenomenon is represented by isolated high-count periods, but aggregation generates seemingly extended anomalies, which we call spurious. In *Safecast* maps, the effect can pretend an ADR anomaly where in fact none exists. Only analysis of the raw log files allows identifying spurious effects. As an example, we discuss dose rate maps of Berlin and Vienna, parts of which have been monitored extensively with a *bGeigie Nano* device mainly carried on bicycle. Closer inspection of detected anomalies reveals some as real, some as spurious. We also discuss possible physical reasons for spurious signals.

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#### Talk 2.3

#### Calibration of Radiation Portal

Monitors for Characterization of Historic Low-level Radioactive Waste

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Radiation portal monitors are large volume radiation detectors typically positioned along roadways, railways, and pedestrian portals. They are typically used for the detection of radioactive material where no such material is expected. Applications include monitoring vehicles and personnel exiting nuclear facilities, at the entrance to steel and scrap metal facilities, and at ports of entry, such as international borders and ports.

The Port Hope Project involves the cleanup of approximately 1.2 million cubic metres of historic low-level radioactive waste from various sites in Port Hope. The waste is a consequence of past practices involving the refining of radium and uranium by a former federal Crown corporation, Eldorado Nuclear Limited, and its private-sector predecessors. It involves the construction of a new Long-Term Waste Management Facility at the site of an existing, closed low-level radioactive waste management facility located in the Municipality of Port Hope, on the shoreline of Lake Ontario.

The waste is transported from its current location to the new facility using trucks. Each truck passes through a radiation portal monitor coupled with a scale when travelling between the waste location and waste storage facility (both directions; empty and full). The radiation portal monitors have been calibrated to provide an estimate of the total radioactivity being deposited into the new waste facility. This paper describes the methodology developed for such a project, where multiple truck sizes and operational considerations were taken into account. Radioactivity concentration ranges from 0 to 5 Bq.g-1 above natural background. A first calibration was done at a smaller site, representing approximately 450,000 cubic meters worth of waste and located 700 m away from the definitive site [1]. This site uses 2 portals, both of them having 3 overhead PVTs.

The methodology has since been used at 3 more sites, including 5 overhead PVTs and with the capability of running trucks in both directions.

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### Session 3

### Impact of Environmental Exposure

**Chair** Mehdi SOHRABI (Tehran, Iran)

#### **Invited Talk 3**

#### Impact of natural radioactivity on microorganisms

#### Vincent BRETON<sup>1\*</sup>, Aude BEAUGER<sup>2</sup>

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Radiation is present everywhere in the universe and on earth. The role it has played in the emergence and evolution of life is still a completely open question. Within the framework of the ZATU Long-Term Socio-Ecological Research observatory, we have started to explore how microorganisms were impacted by natural radioactivity. In order to suppress cosmic rays, we have first conducted long term evolution experiments in the Modane Underground Laboratory (LSM) at the frontier between France and Italy. These experiments have shown that microorganisms displayed the same evolutionary path when natural radioactivity was reduced by a factor 7 (figure 1)[1].

In a second step, we are now studying microbial biodiversity in radioactive mineral springs which are peculiar ecosystems where physico-chemical and radiological parameters are significantly different from their surroundings and extremely stable over very long periods of time. We recently observed that a significant fraction of the microscopic algae (diatoms) colonizing the most radioactive springs of the Auvergne region (La Montagne spring) displayed stress response through deformation of their exoskeleton (figure 2) [2]. The roles played by both chemical and radiological stresses are currently under investigation in 30 mineral springs in Auvergne and beyond.



Figure 1: Bacteria evolved for 500 generations in Clermont-Ferrand (LPC) and Modane Undergound Laboratory (LSM) have similar fitness increases. [1]



Figure 2 : time variation of the deformation rate (in %) of diatom exoskeletons in two radioactive springs of the Auvergne region [2]. La Montagne radon activity reaches up to 4000 Bq/l

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Determination of heavy metal level in tuna fishes from Laccadive sea and their associated health risk

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The concentration of heavy metals in Yellowfin tuna and Skipjack tuna from the coastal waters of Laccadive sea were determined by ICP-OES to evaluate the probable toxic effects to the Maldivian. The samples were collected from different atolls of Maldives to ensure a good representation of sample distribution. The metal concentration of tuna fishes lies within the maximum tolerable limit set by different health organizations. The EDI of each metal was much lower than their respective TDI/PTMI/PTWI. The THQ values for individual metal were well below 1, indicating no health risk for humans due to the intake of individual metal. The maximum targeted cancer risk value for inorganic As was 10-4, indicating low carcinogenic risk from consumption of tuna fish from the Maldives. According to these results, the consumption of tuna in the Laccadive sea is safe for human health.

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### Bacterial diversity characterization of naturally radioactive mineral springs: Role of these bacterial communities in uranium biogeochemistry

#### <u>Guillaume HOLUB<sup>1\*</sup></u> for the TIRAMISU collaboration, Claire SERGEANT<sup>1</sup>, Marie-Hélène VESVRES<sup>1</sup>, Clarisse MALLET<sup>4</sup>, Lory-Anne BAKER<sup>2-4</sup>, Sofia KOLOVI<sup>5</sup> Aude BEAUGER<sup>2</sup>, David BIRON<sup>4</sup>, Patrick CHARDON<sup>5</sup>, Lydia MAIGNE<sup>5</sup>, Didier MIALLIER<sup>5</sup>, Hervé MICHEL<sup>3</sup>, Gilles MONTAVON<sup>6</sup>, Vincent BRETON<sup>5</sup>

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Microorganisms are the first living beings to have appeared on Earth, and populate many habitats, including the most extreme. Among these environments, some are characterized by naturally high levels of radiation where microorganisms have developed over thousands of years by adopting various strategies to respond to the stresses induced by ionizing radiation [1]. Naturally radioactive mineral springs are therefore ecosystems where ionizing radiation could constitute an abiotic driver impacting the diversity and structure of microbial communities.

The primary objective of the TIRAMISU collaboration, within the "Zone Atelier Territoires Uranifères (ZATU)", is to understand the effects of radioelements (radiation / chemical toxicity) on microbial communities from a diverse range of naturally radioactive mineral springs in the French Massif Central (Auvergne). The Massif Central is a region made up of uranium-rich geological formations (granite massif), well known for its naturally radioactive mineral sources. Microbial inhabitants of natural uranium enriched environment exhibit various adaptive features to support their growth and survival, giving rise to uranium tolerant populations in such sites [2]. The objective of our study is to identify and characterize the bacterial communities in these mineral sources, and to understand their role in uranium biogeochemistry.

In this study, we focused on the study of samples from six naturally radioactive mineral springs of varying radioelement concentration gradient (<3.71 ppb U) in the Massif Central sampled in autumn and spring. The bacterial biodiversity of these samples was characterized by next generation sequencing (Illumina Miseq), targeting the 16S rRNA gene. In addition, the influence of the different physico-chemical parameters of these sources on these bacterial communities was studied. On a second hand, culturing on non-selective medium (TSB) was performed for samples from these six mineral springs and a seventh with a higher concentration of uranium (15.91 ppb U). A collection of 625 aerobically cultivable bacterial isolates, corresponding to 295 OTUs, was thus constituted via Sanger sequencing of the 16S rRNA gene. Subsequently, close relatives to known radiotolerant species were screened in uranium resistance experiments. The most resistant strains were further selected and characterized genetically and metabolically to understand the mechanisms behind their tolerance towards radioelements. These preliminary results will be discussed in the context of understanding the evolution, functionality and survival of microbial communities and their interactions with uranium in naturally radioactive sources.

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#### High prevalence of uranium in groundwater of Punjab: A comparative study of its distribution, potential source & health risk assessment

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The present study has been undertaken to assess the distribution of the uranium in groundwater along with health risks associated with its ingestion, covering South-West (SW) and Northern (N) regions of Punjab state, India. The uranium concentration in groundwater of SW and N regions of Punjab has been observed to be varying from 1.3 to 541.8  $\mu$ gL<sup>-1</sup> and BDL to 68.6 $\mu$ gL<sup>-1</sup>, with mean values of 93.6 and 8.9 $\mu$ gL<sup>-1</sup>, respectively. It has been observed that 78% of analyzed samples particularly from the SW-Punjab, exceeded the maximum permissible limit sets by WHO(2011) of 30µqL<sup>-1</sup> and even 52% of the samples exceeded 60µqL<sup>-1</sup>, the limit recommended by AERB (2004) of India.The measured annual effective dose ranged from 9.9 to 1084.0 μSv y<sup>-1</sup> and 0 to 106.7 μSv  $v^{-1}$  for SW and N Punjab, respectively. The chemical toxicity has been found to be in the range of 0.44 to 48.65  $\mu$ g Kg<sup>-1</sup>day<sup>-1</sup> with an average of 6.38µg Kg<sup>-1</sup>day<sup>-1</sup>for SW-Punjab, which is guite higher than AERB recommended limit of 4.53 μg Kg<sup>-1</sup>daγ<sup>-1</sup>. The HQ for SW-Punjab was found to be greater than 1, indicating significant risk due to intake of uranium contaminated water from the SW region. However, the uranium concentration with its both radiological and chemical risks in all the analyzed groundwater samples from N-Punjab was observed to be well below the international and national recommended safe limits. The guite high levels of TDS, salinity and conductivity have also been observed particularly in SW region of Punjab, which might have increased the leaching of uranium through the formation of carbonates and bicarbonates. Maximum uranium concentration in groundwater has also been observed only at shallow depths. In SW-Punjab, it is being recommended to use the canal water for drinking purposes and domestic uses by urban and rural populations than groundwater sources.

Radioactive mineral springs: A first approach of modelling the radiation exposure on microorganisms

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The TIRAMISU collaboration within the ZATU in France focuses on the analysis of the response to natural radioactivity of microorganisms living in mineral springs.

For the current study, water and sediment samples from 27 mineral springs in the volcanic Auvergne region (Massif Central, France) have been collected and analyzed for their radiological and microbial content. High levels of <sup>222</sup>Rn activities in water (up to 4000 Bq/L) and concentrations of <sup>226</sup>Ra (up to 50 Bq/gr) in the sediments were measured by  $\gamma$  -spectroscopy, while the characterization was supplemented with  $\alpha$ -spectrometry and ICP-MS-HR analyses. Among the microorganisms present, diatoms (microalgae), widely used as environmental indicators of water quality, have shown to display an exceptional abundance of teratogenic forms in the most radioactive springs studied [1].

A first estimation of the radiological risk to freshwater biota using the ERICA tool [2] showed that most of the sampled mineral springs are highly above the risk threshold of  $10 \mu$ Gy/h due to the large concentrations of radium in the sediments.

The radiological data from the analysis of the water and sediment samples are used as inputs to Monte Carlo simulations to evaluate the dose received by the diatoms and the potential radio-induced damages. Microdosimetric (GATE) [3] and nanodosimetric (Geant4-DNA) [4] approaches are coupled to model the surrounding environment, the diatom and the nucleus, in order to assess the damages on the diatom DNA. Results on radiation exposure and DNA Strand Breaks due to direct energy deposition will be discussed for different porosities and sediment compositions around the diatoms.

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### Session 4

### Environmental Measurements and Analysis

**Chair** James Mc LAUGHLIN (Dublin, Ireland)

#### **Invited Talk 4**

Linking speciation and soil-water-plant transfers of natural radionuclides in a wetland impacted by a former U mine: recent advances from the pluri-disciplinary and multi-partners project INSPECT-NEEDS

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Main sources of radioactivity in the environment are natural radionuclides (RNs) which are ubiquitous in all Earth's surface ecosystems, i.e., in air, soils, waters, and living organisms including Humans, as terrestrial RNs (uranium-238, uranium-235, and thorium-232 from geochemical background and their radioactive daughters) or cosmogenic RNs (e.g. carbon-14). However, during last decades, numerous human activities in the fields of civil or military nuclear industries, or medicine, for example, have led to the production of technologically - enhanced natural radioactive materials (TE-NORM) as well as of artificial RNs that have been released or emitted in natural surface media. TE-NORM may occur typically in the Earth's "critical zone" which is the near-surface environment where interactions between rock, soil, water, air and living organisms control natural habitats and life-sustaining resources.

RNs are thus a main environmental issue owing to their increasing rate of release in aquatic and terrestrial biotopes, their potential radio-/chemical toxicity, and the complexity of the bio-physicochemical mechanisms that control their fate and eco-toxicity. For example, vegetated soils and / or wetlands that are located in vicinity of former uranium mines or U-mill tailing storage area are potentially long-term sources for increased transfers to surface waters and to biosphere of cocktails of potentially toxic RNs, which may undergo a bio-magnification along trophic chains. Understanding and predicting the transfers of the RNs' cocktails and assessing their direct or indirect effects is mandatory and addresses major societal challenges of the 21st century: reduction of anthropogenic risks, sustainability of resources, and preservation of ecosystem's and Human's health.

When a RN is released into a soil-water-plant system, its transfers, bio-availability, and eco-toxicity is controlled by its chemical speciation (chemical states and forms), which depends on its ability to interact and to form stable chemical species with components within the aqueous, (nano-)mineral and (micro)biological compartments of the ecosystem or at the compartments' interfaces. The concept of chemical speciation is at the heart of the studies in environmental chemistry, where the priority is no longer to only measure concentrations of RNs but also to develop predictive speciation models allowing to apprehend transfers –and thereby eco-toxicity- of RNs in ecosystems. In order to develop these models, it is of crucial importance to make direct determinations of the speciation of RNs existing at trace levels in complex natural systems and to elucidate the relations existing between speciation, lability, mobility and transfers to trophic chains (bioavailability). To this regard, challenging and emerging research topics deal with elucidating the effects of mineral-solution interfaces, microorganisms, and complex natural organic matter. Meeting these challenges requires the use of powerful and advanced spectrometric, spectroscopic, and analytical techniques in order to carry out molecular-level investigations of the speciation of RNs, as well as to quantify transfers of trace RNs at the ecosystem scale by analysis of environmental and biological matrices.

This presentation will highlight new perspectives of innovative and pluri-disciplinary approaches developed at the interface between chemistry and biology within the frame of the NEEDS-INSPECT project in order to elucidate the mechanisms of transfers of natural RNs in very complex systems, namely the soil-water-plants continuum of wetlands. Decisive and recent advances on our understanding of mechanisms of the bio-geochemical transfers of RNs in a wetland impacted by a former U mine in France (Rophin), which were obtained by mobilizing complementary expertise of scientists of eleven teams of CNRS, University, CEA and IRSN within the INSPECT project<sup>[e.g., 1]</sup>, will be reviewed.

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#### Talk 4.1

#### Biogeochemical behaviour of tellurium (Te) and caesium (Cs) at the continent - ocean interface: preliminary scenarios for accidental radionuclide releases

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Radioactive tellurium (Te) and caesium (Cs) are fission products in nuclear power plants (NPP) emitted during accidental events (i.e., Chernobyl-1986, Fukushima-2011)[1]. Post-accidental models describing their fate and dispersion in natural environments require the understanding of the biogeochemical behaviour of their stable homologues. However, stable Te and Cs are poorly studied in aquatic ecosystems due to analytical challenges (i.e., ultra-trace levels of Te), and low research interest in non-radiological isotopes of Cs[2]. This work presents (i) long-term records (2014-2017) of dissolved and particulate Te and Cs concentrations along a major European fluvial-estuarine system, the Gironde Estuary, and (ii) historical records of Cs and Te bioaccumulation in oysters living at the estuary mouth (RNO/ROCCH, 1984-2017). All samples were analyzed with the new generation ICP-MS-MS (Thermo® iCAP TQ). Riverine monitoring showed a synchronous seasonal cycle of concentrations for dissolved and particulate Te, resulting in constant log<sub>10</sub> Kd along the year (~4.75 L kg<sup>-1</sup>)[3]. Estuarine reactivity in flood and drought conditions suggests a reactive behaviour of Te, potentially dominated by mineralization and re-adsorption processes. In contrast, the fluvial log<sub>10</sub> Kd of Cs increased downstream from

~4.54 to 6.17 L.kg<sup>-1</sup>, showing conservative behaviour along the estuarine salinity and turbidity

gradients. A time series of bioaccumulation of Te and Cs in wild oysters showed no clear long- term trends (average 2.08  $\mu$ g kg<sup>-1</sup> and 114  $\mu$ g.kg<sup>-1</sup> dry weight, respectively)[3], and bioaccumulation factors (BAFs) of one to two orders of magnitude below those observed for essential elements (BAF Te = ~30 à 70 L.kg<sup>-1</sup>; BAF Cs = ~ 386 à 1220 L.kg<sup>-1</sup>). This study provides the first conceptual scenarios for environmental fate of Te and Cs radionuclides in case of hypothetical accidental releases from the Blayais NPP located in the Gironde Estuary. Based on the dynamics of the Maximum Turbidity Zone (MTZ; >1g. L<sup>-1</sup> of suspended particulate matter, SPM) and SPM estuarine residence times, accidental releases during flood conditions would cause the highest sorption of Te and Cs radionuclides in the MTZ. It is expected that during the following dry season, desorption of particulate Cs (MTZ acting as a secondary source of radionuclides to the ocean) and transport towards the coastal area may occur, whereas radioactive Te particles may be transported upstream towards the city of Bordeaux.

Keywords: Technology Critical Element, temporal series, environmental risk assessment

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## Talk 4.2

# In situ ATR - FTIR study of uranyl sorption at illite - solution interface in the presence of phosphate ions

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Introduction: Gaining insights into mechanisms and species of uranyl sorption onto illite, at a realistic and low concentration of uranyl in the micromole range per liter, is highly needed for providing refined mechanistic process understanding of radionuclide retention in clay systems. Moreover, studying uranyl sorption processes due to the presence of aqueous ligands (such as phosphate) is mandatory for the transferability of experimental results on radionuclide retention to "the real system".Materials and methods: Na-homo-ionic form illite du Puy (NaldP) < 75 $\mu$ m was used as "received illite" material. Batch experiments were performed under atmospheric conditions at pH 3 - 8, solid-to-liquid ratios ( $R_{S/L}$ ): 0.5g - 3g/L, [UO<sub>2</sub><sup>2+</sup>]: 1-10 $\mu$ M and [PO<sub>4</sub><sup>3-</sup>]: 20-100 $\mu$ M. We monitored by ATR-FTIR spectroscopy the dynamics of uranyl-phosphate species at the NaldP-solution interface during sorption process. We have focused the IR spectra on the frequency region characteristics of phosphate ions (900-1200cm<sup>-1</sup>).

Phosphate sorption at NaIdP-Solution interface: Sorption isotherms of phosphate at pH 4 showed an increasing amount of sorbed phosphate with increasing initial aqueous phosphate concentration, until reaching a plateau. The result of sorption edge showed that the amount of removed phosphate remained almost constant at acidic pH and decreased at near-neutral pH. Examining changes in IR spectra during phosphate sorption, suggested the existence of two types of sorption mechanisms. Resolving IR spectra of phosphate–NaIdP interface showed an increase of IR bands at 1075 and 1157 cm<sup>-1</sup>, with increasing initial phosphate concentrations, as well as appearance of bands at 1009 and 1040 cm<sup>-1</sup> at high phosphate concentrations. This suggested formation of an outer sphere surface complex of phosphate and an increasing contribution of an inner-sphere phosphate surface complex forming at high concentrations. It is hypothesized that the latter is probably a bidentate phosphate surface complex formed at aluminol / ferrinol sites present at edges of illite.

Uranyl sorption at NaIdP-Solution interface: Sorption edge of uranyl indicated that: (1) the amount of removed uranyl increased with the increasing acidic pH and reached a plateau at higher pH; (2) the presence of phosphate ions increased the removal of uranyl at acidic pH, then reached a plateau and showed no significant differences comparing to that in the absence of phosphates ions at higher pH. Blank experiments (without NaIdP) showed that uranyl containing colloidal phases were predominantly present in the solution at higher pH. This suggested that the removed uranyl mainly presented as uranyl-colloidal and uranyl-phosphate-colloidal phases at higher pH. Sorption isotherms of uranyl at pH 4 showed that: (1) the amount of removed uranyl increased with increasing initial concentration of uranyl until reaching a plateau, and (2) the presence of phosphate ions increased the uranyl adsorption. Resolving IR spectra of uranyl-phosphate (co)sorption at NaIdP-solution interface showed an increase of IR band at 1050 cm<sup>-1</sup> with increasing initial concentration of uranyl while a decrease of IR bands at 1075 and 1160 cm<sup>-1</sup>. This suggested an outer sphere surface complex of phosphate decreasing with an increasing contribution of an uranyl-phosphate inner-sphere surface complex or an uranyl-phosphate surface precipitation at higher concentrations of uranyl. An increas of IR bands at 992, 1081 and 1114 cm<sup>-1</sup> was also showed during "a long time" sorption process which suggested an autunite-like phase surface precipitation.

### Talk 4.3

#### Environmental Radiological Baseline for Unconventional Oil and Gas Zones in Mexico

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The natural radiation level in a given site can be changed by NORM material generated by the oil, natural gas, copper, silver, uranium and lithium mining industries, among others. In Mexico there is no baseline of natural radiological levels but there is exploitation of energy resources by the industry and there is a possible extraction of unconventional hydrocarbons (UHC). This work deals with a radiological baseline study for four sites within the oil province of Tampico, which comprises 58% of the Mexican UHC reserves, the study is performed prior to the possible massive development of UHC extraction wells using the fracking technique.

Three of the evaluated sites (A, B and C) are located in areas characterized by important hydrological reservoirs, grassland and seasonal agricultural crops for human consumption; the fourth site (D) is located in the Geological Basin of Burgos, relevant for its oil and natural gas extraction for the last 70 years and it is considered as one of the most important in the world. In each of the four sites, the environmental equivalent dose rate was determined for an area of 1 km<sup>2</sup>, measured in five transects of 200 m each one. The results showed value bands, in mSv/year, of 0.99-2.18, 0.57-0.78, 0.61-0.74, and 0.13-0.88 for sites A, B, C and D, respectively. The bands of low values clearly show the non-impact of the zones by industrial activities. The radiological maps obtained were interpolated using the geospatial kriging method and are discussed in terms of vegetation, soil type, population and crops data layers for the study areas. The highest values of the equivalent dose rate are located in grassland areas and close to a population of approximately 181 inhabitants at 1.5 km distance. Additionally, groundwater flows were identified, where the presence of environmental <sup>226</sup>Ra and decay products in water samples were assessed. These results are the first of this type for Mexico, which will serve for the energy sector as a reference for future comparisons and/or restrictive limits in the case of massive development of fracking or other extractive industries that can increase doses to the baseline of the site.

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# Talk 4.4

# Fraudulent exams in the training of reactor supervisors at the Laguna Verde Nuclear Power Plant- México

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In both the nuclear and aeronautical industries, operator training is necessarily rigorous and permanent due to the risks of an accident and its consequences. However, at the Laguna Verde Nuclear Power Plant (LVNPP), this statement was questioned after a group of five candidates presented an exam for Reactor Supervisor, and it was discovered that one candidate left on a computer where the exam would be applied, an electronic storage device (USB) containing the same test. The details are described in the Official Letter entitled "Special Inspection IE-01/15-LV1"[1], which was obtained through the National Institute of Access to Public Information and was prepared by the institutions involved: the National Commission of Nuclear Safety and Safeguards and the Federal Commission of Electricity, which operates the LVNPP. In this paper we discuss the contents of the "Special Inspection IE-01/15-LV1", which also revealed the vulnerabilities of the LVNPP. This paper aims to draw attention and bring up for discussion the need of international monitoring over this workplace in order to eliminate possible unsafe conditions in LVNPP that could have consequences for the integrity of the population, its workers and the Pepartment of Physics at the Faculty of Sciences of the National Autonomous University of Mexico.

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#### Poster 4.1

# Radioactive waste management without adherence to standards at the Laguna Verde Nuclear Power Plant, Mexico.

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Radioactive Waste Management at the Laguna Verde Nuclear Power Plant (LVNPP) has been investigated and reprehended by the National Commission of Nuclear Safety and Safeguards (NCNSS)-the regulatory body on nuclear matters in Mexico. The NCNSS, after performing the "Inspection of Activities Related to the Radioactive Waste Management from the Laguna Verde Nuclear Power Plant 1 and 2" and the activity titled: "Verification the isotopic composition of the batches prior to initiating the process of solidification", noticed anomalies in the implementation of the working procedures that establish how these activities must be carried out. The statements above are based on the Official Letter No. A00.130 / 017/2015[1], dated 14 April 2015, issued by the NCNSS with the subject: "Administrative Admonition with Warning". This paper discusses the recommendations issued by the NCNSS to overcome these irregularities, and additionally other incidents are mentioned related to Radioactive Waste Management, which aims to ensure the safety of the staff, public and the environment. "Emphasis is placed on that the possession of an Operating License from a Nuclear Power Plant, granted by the Ministry of Energy of the Mexican government, gives the LVNPP the responsibility to perform with the highest safety standards, fulfilling all and each of the regulatory requirements and license conditions which apply, always staying vigilant to ensure the safety of the personnel, the public and the environment.". It is intended to point out and eliminate the Bad Practices of Radiological Protection in the LVNPP. This paper was written at the Department of Physics at the Faculty of Sciences of the National Autonomous University of Mexico.

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# Session 5

# Environmental Monitoring for Radiation Protection

**Chair** *Vincent BRETON* (Clermont-Ferrand, France)

# **Invited Talk 5**

Radon measurements indoors, in soil and in water by track detectors and/or by Geiger-Muller counters

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The large radon concentrations of the home's indoor air can be caused essentially by two sources, the soil or the water supply. A thoroughly investigation of the indoor radon may require the measurement of radon indoors, in soil, and in water respectively. With existing technologies, these radon measurements require a variety of different devices and sampling procedures. Finally, a new generation of passive radon monitors has been developed, which makes it possible to obtain a multitude of different types of measurements indoors, in water, and in soil with a single compact device. This device can be referred to as a radon film-badge, which is formed by a radon-sorption plastic-film facing an alpha-particle detector. This radon film badge, once enclosed in a radon permeation-bag, makes it finally possible to use the same device for Rn-measurements indoors, in soil, and in water for different exposure durations from a few days to one year.

The radon film badge is based on the detection of tracks induced by alpha particles from a radiator-absorbed radon and its decay-products. Finally, it has been proved that it is possible to measure the radon without using the track detector. In this case, the radiations emitted from the radon (and its decays) from the radiator are counted directly by a Geiger-Muller counter.

In order to prove the unique characteristics of these two novel radon monitoring techniques, they are used for the measurement of very different radon concentrations, which may vary from a few tens of  $B_0/m^3$  for indoor radon to the extremely large radon concentrations, encountered in water wells and/or in the thermal waters of a SPA. These radon concentrations can be larger than 10000 KB $_0/m^3$ , as those encountered in the Lurisia SPA from Italy, the thermal waters of which were first studied for their large radioactivity by Madame Curie (Ciardi, 2017).

Reference : M. Ciardi. Marie Curie-La signora dei mondi invisibili. Hoepli Ed., Milano (2017)

Development of a Portable Type Personal Dosimeter for internal and external dose assessments

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In the Fukushima-Daiichi Nuclear Power Station (FDNPS) accident that occurred in March 2011, existing monitors for radioactive materials in the atmosphere are expensive and often large, so they could not be deployed in a wide range of locations after the accident. Thus, there are few data on internal exposure doses of residents after the FDNPS accident. On the other hand, small and inexpensive radiation measuring instruments for assessing external exposure doses are available on the market, but there have been many cases where the indicated values differ depending on the manufacturer even when measured at the same location, or where it takes time to obtain the indicated values. In this study, we have developed an instrument that can evaluate not only ambient dose equivalent rate but also airborne radioactivity concentrations which are corresponding to internal exposure doses quickly, easily, and precisely with a single measuring instrument under an emergency situation. The structure of the monitor developed in this study is shown in Figure 1. For internal exposure dose assessment, radioactive materials in the atmosphere are collected on a filter, and their radiations are detected by a silicon semiconductor detector placed in front of the filter. The detection and collection sections are designed so that the alpha particles on the filter can be detected efficiently. In emergency situations, the purpose is to detect plutonium and uranium, and the exposure dose is evaluated by the gross count rate of detected alpha particles. The performance of the system was evaluated by conducting continuous measurements in the radioactive aerosol chamber installed at the Institute of Radiation Emergency Medicine (IREM), Hirosaki University. A GAGG scintillator was used as a detector to evaluate the ambient dose equivalent rate. Cs-137 gamma-ray source was used for evaluation of the energy resolution and energy calibration of the GAGG scintillation detector. The integrated measuring instrument for simultaneous assessment of ambient dose equivalent rate and atmospheric radionuclides concentration developed in this study was significantly smaller and lighter than the devices available on the market so far. On the other hand, there is still room for improvement at present. If this issue is resolved, the device is expected to be utilized for environmental radiation monitoring around nuclear facilities and for radiation control in hospitals, universities, laboratories, and other institutions that handle radioactive materials.

This study is supported by the Strategic Core Technology Advancement Program Grant Number 220202020 (the Ministry of Economy, Trade and Industry of Japan).

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# Site-specific dose conversion factors for radon progeny based on actual aerosol size distributions at various environments

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The ICRP recommended a new dose conversion factor for radon, which is two times higher than the value by the UNSCEAR[1,2]. Therefore, an inhalation dose assessment for radon and its short-lived progenies gets more essential recently. The ICRP also mentioned the importance of the aerosol size distribution in the inhalation dose assessment. Many researchers reported that the inhaled aerosol size would determine the sites of deposition in the respiratory tract. In addition, each region of the respiratory tract has different radiosensitivity. Therefore, the inhalation dose strongly depends on the aerosol size distribution. The ICRP also mentioned that site-specific dose conversion factors can be used if sufficient, reliable aerosol information is available and estimated doses warrant more detailed consideration. In this study, aerosol size distributions were measured under various situations and the site-specific dose conversion factors were evaluated and compared with values of other reports.

The aerosol size distributions were measured at a tourist cave by a Portable Aerosol Mobility Sizer (Model3310, Kanomax Inc.), which can measure the aerosol size distribution in the range from approximately 15 to 800 nm. Also, activity size distributions of radon and thoron progeny were measured using Micro-Oriffice Uniform Deposit Impactor (Model 110, MSP) and alpha counters (Model 43-10, Ludlum Measurements, Inc.) at the outdoor environment in Hirosaki. In addition, 4-stage impactor was used with Solid-State Nuclear Track Detector (CR-39, BARYOTRAK, Fukuvi Chemical Industry Co., Ltd.). Based on the results obtained, the site-specific dose conversion factors were evaluated using Integrated Modules for Bioassay Analysis (IMBA Professional, Health Protection Agency) under several assumptions.

The results of the measurements and calculations will be presented in the presentation

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# Determination of <sup>226</sup>Ra/<sup>238</sup>U and <sup>228</sup>Ra/<sup>232</sup>Th disequilibrium in surface soils from HBRA, Odisha, India and its radiological implications

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Naturally occurring radionuclides of terrestrial origin are ubiquitous in the Earth's crust. The radionuclides which are responsible for the enhanced natural background radioactivity are <sup>238</sup>U, <sup>232</sup>Th and their decay products. Therefore, it is necessary to monitor the environmental behavior of the naturally occurring radionuclides and radiation patterns in the HBRAs to increase awareness with public health risks. Radiological investigations have been carried out in the eastern coastal area of Odisha, India to measure gamma dose rate and natural radionuclide concentration in selected surface soil samples. <sup>238</sup>U and <sup>232</sup>Th were measured using inductively coupled plasma mass spectrometry (ICP-MS) and high purity germanium (HPGe) gamma spectroscopy was used for <sup>226</sup>Ra, <sup>228</sup>Ra and  ${}^{40}$ K. The high concentration of  ${}^{232}$ Th in soil increased the absorbed dose rate in air to the maximum of 748 nGy/h. The ratios of <sup>226</sup>Ra/<sup>238</sup>U and <sup>228</sup>Ra/<sup>232</sup>Th in soil at radioactive secular equilibrium are 1. In this study <sup>226</sup>Ra/<sup>238</sup>U and <sup>228</sup>Ra/<sup>232</sup>Th ratios ranged from 2.9 to 5.6 and 0.8 to 2 respectively. The ratios of <sup>228</sup>Ra/<sup>232</sup>Th in soil show radioactive secular equilibrium, whereas <sup>226</sup>Ra/<sup>238</sup>U ratios are > 1 with an absolute difference 2.9. The ratios of <sup>226</sup>Ra/<sup>238</sup>U clearly exhibiting the radioactive diseguilibrium in soil. In order to understand the environmental behavior of the radionuclides, plots were performed between <sup>228</sup>Ra/<sup>238</sup>U and <sup>226</sup>Ra/<sup>238</sup>U (Fig 1(A)); <sup>226</sup>Ra/<sup>238</sup>U and total labile  $^{238}$ U (Fig 1(B)). Fig 1(A) shows strong positive correlation R<sup>2</sup> = 0.90 and Fig 1(B) shows correlation R<sup>2</sup> = 0.42. This suggests that geochemical nature of U is highly mobile or it is intensively leaching from the surface soil than <sup>226</sup>Ra. High values of <sup>226</sup>Ra/<sup>238</sup>U diseguilibrium are associated with depletion of <sup>238</sup>U. Therefore, the radioactive disequilibrium data between <sup>226</sup>Ra and <sup>238</sup>U is crucial to predict the geochemical mechanism such as the dissolution or migration of <sup>238</sup>U from surface soil. For detailed understanding of this phenomenon physicochemical characteristics of soils has also been carried out.



Fig. 1 (A)  $^{226}$ Ra/ $^{238}$ U between  $^{228}$ Ra/ $^{232}$ U; (B)  $^{226}$ Ra/ $^{238}$ U between Total labile  $^{238}$ U

Radon spatial and seasonal variations in University's buildings located in an Italian karst region

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In the framework of a collaboration between INAIL and University of Salento (UniSalento in the following) a radon survey in 54 buildings belonging to the UniSalento's campus was performed. The buildings are located mostly in a restricted area presenting a morphology characterized principally by marls, calcareous marls and calcarenites belonging to several Pleistocene sedimentary cycles (karst area).

The survey was performed monitoring for two consecutive semesters corresponding to spring/summer (SS) and autumn/winter (AW) about 900 rooms located at different floors (see tab. 1). Moreover, in a restricted sample of about 250 rooms another radon monitoring was performed for six-months corresponding to winter-spring season (WS) in order to better characterize radon seasonal variations and to evaluate if the indoor radon concentration in a certain season could be assumed as representative of a one year radon level. For radon monitoring, passive devices with SSNTD were used; more information about laboratory technique and procedures are given elsewhere (1–3).

The analysis of radon level distribution respect to floors highlighted not negligible radon levels also in rooms located at upper floors, in particular in historical buildings (figure 1). This situation requires particular attention in choosing and testing the proper remedial actions. The estimated annual Radon concentration are very similar to the ones calculated for the WS period (annaul\_Rn/ WS\_Rn =1.1). Regarding the ratio between SS and AW an inverted seasonal factor was found out: AW/SS=0.85. This phenomenon was also found in other karst areas (4), although generally it is not common to find higher radon concentration during SS than in AW.

Floor	N. of rooms		
Ground floor	668		
Basement	145		
1 <sup>st</sup> floor	197		
$2^{nd}$ and upper floors <sup>*</sup>	90		



Table 1. Spatial distribution of the monitored rooms

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# Session 6

# NORM & TENORM

# Chair

*Shinji TOKONAMI* (Hirosaki, Japan)

# **Invited Talk 6**

NORMs in cultivated honey in Malaysia and concomitant dose to the consumers

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Food consumption is one of the main pathways that lead to the radiation exposure to human body. Honey is well known for its nutritional value, thus finds great demand by the world communities including by Malaysian. This study assess the concentrations of natural and artificial radionuclides (especially <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>40</sup>K and <sup>137</sup>Cs) in 8 category of honey produced in Malaysia via High Purity Germanium  $\gamma$ -ray spectrometry. Activity concentration of <sup>226</sup>Ra, <sup>228</sup>Ra and <sup>40</sup>K radionuclides in honey samples found to be in the range of 3.5±0.4-4.5±0.4 Bqkg<sup>-1</sup>, 1.0±0.4-1.7±0.4 Bqkg<sup>-1</sup> and 41.4±3.3-105±7 Bqkg<sup>-1</sup>, respectively. Present study show no data for <sup>137</sup>Cs or it was below the detection limit. The average committed effective dose via consumption of honey are estimated to be <1 µSv/y, which is much lower than the permissible limit 290 µSv/y (for all sources of natural radiation) given by the UNSCEAR. Such a lower dose is attributed to the very little consumption of honey by Malaysian population. This study shows no significant radiological threat to human health and safe to consume in daily life.

**Keywords:** Honey; HPGe  $\gamma$ -ray spectrometry; Natural and fall out radionuclides; Committed effective dose

# Talk 6.1

# Estimation of Radiation Dose due to Thoron and Progeny inhalation in high Background natural radiation area of Eastern Coastal Area of Odisha, India

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It is an establish fact that exposure of high radon is one of the causative factors of human lung cancer. The presence of thoron, <sup>220</sup>Rn, was often neglected because it was considered that the quantity of thoron in the environment is less than that of radon. However, recent studies have shown that the dose due to exposure to <sup>220</sup>Rn and its progeny can equal or several times exceed that of <sup>222</sup>Rn and its progeny. The results of thoron and its progeny measurements in the houses of normal and high background radiation areas (HBRA) of India using both active and passive techniques in different types of houses are presented here. A comparison between the results obtained with various techniques is presented in this paper. Thoron concentration was found relatively higher in the houses of the study area. The effectiveness of various thoron and progeny measurement techniques in estimating the dose to general public are discussed in details.

# Talk 6.2

#### Good RAD7 performance for high radon levels

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A good performance of six RAD7 electronic devices was confirmed for detecting a wide range of Radon concentrations (32 to 280,000) Bq m<sup>-3</sup>, in air, soil and water. Two types of experiments were performed. Firstly the RAD7 detectors were tested in the laboratory using a high radon source from a confined uranium mineral inside a small sealed container, from which radon is diluted to other experimental chambers of different sizes. Evaluation of each radon concentration in the experimental chambers is achieved by means of a Ge (Hp) detector. In accordance with the reported sensitivity by the manufacturer of 0.4 cpm/PCi/L, a general expression was derived which permits to set criteria for choosing the expected counting errors in terms of time-counting intervals and radon concentrations. Secondly, the RAD7 performance was successful tested in a uranium mine under high environmental properties; where radon concentrations in air was up to 93 kBq m<sup>-3</sup> and up to 285 kBq m<sup>-3</sup> in soil. Short time-counting intervals of 5 minutes were chosen because the relative humidity inside the detector nearly reached values above 20% that may reduces the efficiency of the detector and increase the risk of damage it; temperature was also extreme slightly above 40 °C.

## **Invited Talk 6.3**

# Radiation Dose Assessment and Mobiliy Studies of Thorium in Soil from a High Level Natural Background Radiation Area in Sri Lanka

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Sri Lankan coastal areas are well-known for their high background radiation levels, mostly due to Th-bearing minerals, identified not more than a decade ago [1,2]. The radioactivity of Th-232 in Sri Lanka ranges between 9 and 1166 Bq kg<sup>-1</sup> (average 138 Bq kg<sup>-1</sup>) [3] while global natural Th-232 content in soil ranges between 11 and 64 Bq kg<sup>-1</sup> (average 30 Bq kg<sup>-1</sup>) [4]. However, higher inland natural background radiation levels were discovered in 2015 in the Kawudupelella area (Matale district) while exploring the availability of natural radio-minerals under a collaborative project between the Sri Lanka Atomic Energy Board (SLAEB) and the Geological Survey and Mines Bureau (GSMB) of Sri Lanka [5]. The radioactivity data of this location raised concern related to the risk of public radiation exposure since the screened area comprised a school playground.

This study presents both on-site radioactivity measurements and results from leaching experiments using soil samples from the school playground in order to better understand the environmental risk in the area. Gamma spectrometry from the soil samples clearly showed the dominant contribution of Th-232 to total activity (ranging between 4000 and 7600 Bq kq<sup>-1</sup>). In-situ measurements carried out using a radiation survey meter equipped with a Nal scintillation detecor and built-in GM counter, yielded background radiation levels at one-meter height above the ground of 2.5  $\pm$  1.2 µSv h<sup>-1</sup> (max. 21.6  $\pm$  10.9 mSv yr<sup>-1</sup>). The calculated absorbed dose rates in air, 3000 – 4600 nGy  $h^{-1}$ , exceed the world averages (18 – 93 nGy  $h^{-1}$ ) and are similar to those registered in monazite-bearing sands in coastal areas of Kerala and Madras (200 - 4000 nGy h<sup>-1</sup>) [4]. The potential mobility (i.e. solubility and leaching behavior) of radioactive Th was extensively studied using appropriate selective extraction schemes (sequential and single batch extractions) and column experiments. Both sequential and single extractions showed comparable results (i.e., no significant losses of Th during sequential extractions) indicating that the maximum extractions (8 wt.% of total Th) occurred from carbonate, organic and amorphous iron/manganese (hydr)oxide fractions, i.e. not related to the major Th-containing mineral phases (oxide, phosphate and silicate phases). Batch extractions and column leaching experiments with simulated rainwater showed much lower leachability of Th compared to the chemical extractions. These results imply that >90 wt.% of the radioactive Th is mainly present in the residual fraction (assumed to be non-reactive) and that environmental processes may mobilize less than ~500 Bq kq<sup>-1</sup> (still non-negligible). This study suggests important public, on-site exposure to radioactive Thcontaining particles, marking the onset for future work concerning radioactive risk assessment in Sri Lanka.

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# Session 1: Special Olko Anniversary

# From Natural core breeding to the Oklo discovery

Chair

Benoît GALL (Strasbourg, France)

#### The Oklo phenomenon, discovery, first questions, first answers

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The discovery in 1972 of the natural nuclear reactors at Oklo provoked many questions among scientists, physicists and geologists. Under the leadership of R. Naudet, the CEA launched the "Franceville Project" which brought together researchers from around the world to answer the many questions raised by this discovery. The initial answers were compiled in two proceedings of two symposia held in Libreville and Paris in 1976 and 1978. This project was then followed by two European projects led by the CEA and the IRSN in 1990 and 1996.

My presentation will quickly recall the scientific adventure of the first years of work on the Oklo site: what were the hypotheses and misunderstandings to finally propose a first coherent but still incomplete scenario in 1978.

The reactors are described with emphasis on the only reactor that has been preserved in its natural state, the other 15 having been exploited. Information is given on the thousands of samples taken.

The history of the reactors is reconstructed in time and the geological context on the basis of the most recent data [1]. In particular, we show that the conditions that led to the functioning of nuclear reactions in a geological series more than 2100 million years old and having had turbulent history are quite exceptional and why they never occurred again [2].

But the great advantage of the Oklo reactors is that they can teach us a lot about the behaviour of fission products and actinides buried for two billion years in a clay environment rich in bitumen. Oklo has already shown that it is a remarkable project in this respect. Much more can be learned, but Oklo's first lesson is to show that we still have a lot of work to do to understand and master a system for trapping nuclear waste in a natural environment.

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## From routine sample measurements in CEA to the Oklo phenomenon

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In 1972, before shipping natural uranium to the USSR for enrichment operations in <sup>235</sup>U, the analysts at Pierrelatte plant noted a slight deficit in <sup>235</sup>U: 0.7171 instead of 0.7202. The Direction des Productions of the CEA launched a vast campaign of analyses for the different mines exploited, at all stages of the elaboration of uranium: analyses on the ore, then on the yellow cake, on the uranium oxides issued of the yellow cake transformation, then on the UF<sub>4</sub> and on the UF<sub>6</sub>.

For this analysis campaign, the Direction des Productions relied on the analytical laboratory of the Pierrelatte plant and on the Central Analytical Laboratory of the CEA, managed by Michele Neuilly, where I was in charge of analyses by mass spectrometry.

The numerous chemical and isotopic analyses of uranium lead to Gabon and more precisely to the Oklo mine. Indeed, the closer one gets to this site, the higher the uranium content and the higher the <sup>235</sup>U depletion.

At a meeting held at the CEA headquarters, it was decided that the laboratories at Pierrelatte and Cadarache would continue to carry out the analysis campaign, and that the laboratory at Cadarache would analyze the samples from Oklo.

COMUF, the company that operated the mines in Gabon, sent two samples of magnesium uranate and two samples of ore to Cadarache. After the analyses of the U and <sup>235</sup>U content, there were enough samples left that I decided to analyze them on a spark mass spectrometer, which provides a panoramic analysis of all the isotopes present in the analyzed product. I discovered on the photo plate of the mass spectrometer, isotopic anomalies, in particular the absence of <sup>149</sup>Sm, whereas <sup>147</sup>Sm was present. The next step was isotopic analyses of some elements, including neodymium and samarium after chemical separation. They revealed that the isotopic composition of these two elements was completely different from that of the natural elements. The results of these analyses were transmitted to the CEA Directorate which sent them to the neutron specialists at Saclay. Their conclusion was as follows, the isotopic compositions of the Oklo ores are identical to those of uranium having undergone a chain reaction of fission.

It was the first time we discovered a natural fission reactor.

The discovery of <sup>235</sup>U depletion and the chain fission reaction in the Oklo ore will be the subject of two communications to the Academy of Sciences.

#### Historical interpretations of isotope measurements and applications

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The French PWR power reactors were using uranium enriched to about 3.5% in U235. Prior to enrichment, a routine check at Pierrelatte revealed abnormalities in U235/U238 isotope ratios. Such anomalies made UF6 inappropriate for enrichment, which was essential. An investigation requested by the General Manager of the CEA has made it possible to clarify the origin of the ore treated and the causes of these anomalies: chemical, nuclear or other? Interpretation of isotope measurements of various fission products (particularly Nd then Sm, Eu... ) demonstrated the occurrence of sustained fissions in the extracted ore: once the small contribution of natural elements to this ore was deducted, the resulting isotopic vectors corresponded well to the isotopic vectors of the fission products. In the autumn of 1972, the use depletion codes to calculate the concentrations of 600 PF made it possible to better characterize the "operation" of these reactors: some reaction zones were indeed fast-breeder reactors, since the existence of Pu239 and "rapid fast neutron" fissions on U238 had occurred. The possibility of a critical state has been demonstrated with a simple reactor model. This possibility results from the combination of two particularities: a minimal presence of water with an enrichment in U235 of the order of 3.5% due to the age of formation of the uranium deposit (2Gy). Forty years after NAUDET's first Oklo ore criticality study, more complete studies of inception condition were carried out at the University of Strasbourg with realistic models, explaining the startup condition of these cores.

In 2010, Professor EL ALBANI highlighted the presence of fossils dating back to the same period and located in the vicinity of the reactors. This event was be the object of a bomb in the scientific world since the appearance of such an elaborate life form would have appeared only 0.6Gy before our era.

The joint occurrence of the two separately improbable phenomena (reactors and fossils) can only draw attention; this was the origin of the conference/debate organized in autumn 2018 by SFEN/PACA: "Chance hazard or causal relationship?".

In order to stimulate discussion with radiobiologists, we proposed, as a first step, to calculate the deposition of energy in matter over versus the ages. For this purpose, we use isotopic measurements made nowadays. The case of the Oklo reactors is the first application. The second case, the *"La Crouzille"* uranium mine, is complementary to the first in terms of the nuclear conditions (intensity and age of formation). The third case is the situation of a lagoon environment, looking for possible explanation of the start of Life on Earth. For all these cases we determine neutron spectra and concentrations of about 1800 isotopes (fissile with their progeny filiation products, fission and activation products). These concentration values can be used to discuss the storage of natural or non-natural radioactive waste.

# Oklo natural analogue of radioactive waste disposal, summary of European Commission projects'results (1991-1999)

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At the dawn of the 1990s, the question of the feasibility of the geological disposal of radioactive waste became significant. Among the tools for studying this feasibility, natural analogues had a special place, making it possible to obtain both quantitative and qualitative demonstration elements. Among natural analogue, the Oklo site, in Gabon (equatorial Africa), represents a unique geological environment where nuclear reaction products have been naturally introduced and can still be detected in fossil reaction zones.

From 1991 to 1999, two international research projects conducted under the auspices of the European Commission, studied radionuclide mass transfer processes to the surface, focusing on quantitative assessment of radionuclide migration/retention within the Oklo hydrogeological basin. These projects compiled useful information and tools for the safety assessment of radwaste disposal. The most relevant part included new data on the long-term evolution of spent fuel, suggesting the possible occurrence of a coffinitisation process and measurement of short term leaching rates of Oklo uraninite; modelling tools and data to quantitatively describe the interaction of uranium and rare earth elements with a complex clayey material; the demonstration of radionuclide trapping in generic mineral phases such as Mn and Fe oxides, chlorite, illite and specific secondary minerals such as phosphates and Zr-silicates; and a consistent understanding of redox buffering in a clayey environment, from deep to surface conditions, generic enough to be adapted or applied to other sites.

Influence of Initial poisons and clays on the criticality of Oklo natural nuclear reactors.

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In the aim to assess more the Oklo phenomenon through the numerical modelling and simulation, within the geological context based on field observations and measurements, a Python-based code is developed to automate the criticality research for a given configuration among a specific parameter, considered as the main variable of investigation. The home-made python program interacts with dedicated code for nuclear reactor criticality calculation, namely MCNP. This allowed us to investigate the asymptotical criticality occurrence, which corresponds to infinite multiplication factor  $k_{-\infty}$  as a function of Uraninite fraction volume and total saturated porosity:  $k_{-\infty}$  (V\_U02,  $\Phi$ )  $\cong$  for different situation defined with relevant parameters, namely: Initial Poisons (Gd, Sm and Nd) and Clay fraction in the Gangue part of the U-rich ore. Indeed, in the first step of the present work, a generic U-rich ore was simulated over a given interval of Uraninite volume fraction and the corresponding porosities needed (Critical Porosity  $\Phi$ ) to reach criticality were obtained. It shows that an optimal point can be defined as the minimal one on the isocritical curve  $k_{-\infty}$  (V\_U02,  $\Phi$ )  $\cong$ . This point, called "Inception point", is the most likely configuration to occur with low Uranium and less water defined by totally saturated porosity.



Figure 1. Typical results of criticality research for a generic U-rich ore obtained with Python-based code with MCNP

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# Session 2 : Special Olko Anniversary

# From Oklo samples to natural core simulations

**Chair** François GAUTHIER-LAFAYE (Strasbourg, France)

The Gabonionta: Great Oxidation Event, reactors and Life...

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The emergence of complex life more than five hundred million years ago marked the beginning of change in the Earth's biosphere. This evolutionary change is associated with numerous events including increasing predation, burrowing, and animal diversity during the so called "Cambrian Explosion" at the Ediacaran-Cambrian boundary. However, several studies have reported that scattered fossils of large individual multicellular macro-organisms that use cells as building blocks existed during most of the Proterozoic Eon, and some of these early lineages (such as red or green algae) still exist. The recent discovery of centimetre-sized fossils of more than 1500 specimens from the 2.1 Ga Paleoproterozoic black shales in Gabon reveals growth of macro-organisms in a coordinated manner. The biogenicity of these remains was investigated using a multi-approach studies. On the surface, the fossils resemble irregularly shaped cookies with split edges and a lumpy interior. High-resolution Xray tomography revealed their varying elongate, lobate, string, circular and a sheet-like structure with a pervading radial fabric and a neat pattern of central fold shapes and sizes. These structures are too complex to be simple products of inorganic processes. Geochemical data confirmed that the carbon contents in the fossilized tissues were assembled by biological processes. Moreover, the iron-sulfide (pyrite) mineral replacing most of the tissue were formed by bacteria "breathing" sulfate, rather than oxygen, during decomposition of the organisms in sediments. Some of these species showed evidence of organism motility in shallow marine waters with free oxygen and provide support for presence of multicellular life to a minimum of 2.1 billion years ago, almost at the beginning of the Proterozoic Eon. Large size generally signifies an energy-demanding way of life. Breathing oxygen, as we do, is a much more efficient way of obtaining energy than other physiological processes. The Proterozoic Eon saw two major events of oxygen build-up in the atmosphere (and, thereby, in the oceans); the first near the beginning of the Eon, 2.45–2.2 billion years ago, and the second at the end, 0.8–0.54 billion years ago. The evolution of the Gabonese biota, representing an early step toward large-sized multicellularity, may have become possible by the first boost in oxygen, whereas the "Ediacara biota" could have been fuelled by the second. Why it took around 1.4 billion years for the multicellular organisms to take over is currently one of the great unsolved mysteries in the history of the biosphere.

Natural Nuclear Reactors: prediction, search, discovery, operation and implications

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Natural nuclear reactors operated in Oklo (Gabon) about 2.0 billion years ago [1]. This phenomenon was predicted and led to a systematic search for uranium deposits that went critical. Fifty years ago, just a few years after this search was abandonded, the Oklo phenome-non was discovered by chance. Many elements extracted from the Oklo reactor material still carry clear isotopic signatures of <sup>235</sup>U fission, <sup>239</sup>Pu production and neutron capture reactions. Isotopic compositions of these elements provided reconstruction of neutron fluence, amount of consumed <sup>235</sup>U, and an effective duration of nuclear fission chain reaction that was estimated to last for hundreds of thousands of years [2]. It was not clear, however, whether the reactor was operating continuously or in pulses. One proposed mechanism was based on burning up highly neutron absorbing impurities (RRE and/or boron) [3]. As the strong absorbers were burned up at one edge of the reactor zone and uranium at the other one, the active zone could have shifted along the U-vein making different parts of the natural reactor been active at different times [3]. Another potential self-regulation mechanism could have involved water acted as a neutron moderator. As the temperature of the reactor increases, all unbounded water was converted into steam, reducing neutron thermalisation and shuting down the chain reaction. Only when the reactor cooled down and the water concentration increases again, could the chain reaction resume. A tiny sample from reactor zone RZ-13 kindly provided by Maurice Pagel, Philippe Holliger and François Gauthier Lafaye carried the answer to this and several other questions.



Novel analytical techniques LENGA and NAUTILUS used for analyses of this 4×3 mm slab revealed:

the highest concentration (~8´10<sup>17</sup> atom/g) of fission Xe ever observed in natural materials [4]
cycling operation of RZ-13 of Oklo and self-regulating mechanism [5],

\* lowest  ${}^{235}U/{}^{238}U = 0.3655$  and capture of fission Cs and Ba 5 yr. after the shutdown [6] ,

\* Al-phosphates and metallic aggregates preserve certain fission products over a geologic time [4, 7].

(Red circles show craters made by LENGA = Laser Extraction Noble Gas Analyses. Yellow square shows area studied by NAUTILUS = NAval Ultra-Trace Isotope Laboratory's Universal Spectrometer, the US Naval Research Laboratory).

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#### Evidence of fast neutron operating in Oklo

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The fission processes for thermal neutron induced <sup>235</sup>U and <sup>239</sup>Pu fission and for fast neutron induced <sup>238</sup>U fission produce fragments with a wide range of mass (72 < A < 162; A = mass number), and neutrons. As the results of fission events, many elements in the Oklo reactor zones (hereafter, RZs) and the related samples show the variations in the isotopic compositions caused by a combination of reactions of nuclear fission, neutron capture and radioactive decay. Isotopic measurements by mass spectrometry provide useful information of geochemical behavior of fissiogenic radioisotopes and nuclear characteristics of the reactors. Since the discovery of the first RZ in 1972, many isotopic studies have been performed to understand the mechanism of the operation as fission reactors and to trace the migration behaviors of fissiogenic isotopes produced in the Oklo RZs [1-2]. In this talk, I will show some typical examples of the isotopic data, and explain the interpretation how and why the fission reactions occurred in the Oklo RZs. In particular, one of the Oklo RZs, RZ 13, is several specific features in the view point of nucleonic characteristics. As representative parameters to characterize the operating conditions of RZs, neutron fluence as the time integration of a neutron flux generated in RZ, duration of RZ operation, restitution factor of <sup>235</sup>U from decay of <sup>239</sup>Pu produced by neutron capture of <sup>238</sup>U, and the proportion of fission events due to <sup>235</sup>U, <sup>238</sup>U and <sup>239</sup>Pu, are listed in Table 1.

By comparison of the data between RZ13 and other RZs, fission contribution of <sup>238</sup>U for RZ13 is found to be significantly higher than those of other RZs. Considering that <sup>238</sup>U reacts with fast neutron rather than thermal neutron, RZ13 might have located at less moisture area, and had reacted with fast neutrons. Furthermore, the lower restitution factor of RZ13 may also support the fast neutron operating in RZ13 because of the preference of fission reactions of <sup>238</sup>U instead of neutron capture reaction caused by the generation of fast neutrons.

RZ No.	3	5	9	10	13
sample No.	SC52-1472	KN267-2194	mean (n=8)	SF84-1485	SD37
neutron fluence(X10 <sup>20</sup> n/cm <sup>2</sup> )	2.28	4.37	3.62	6.22	7.80
operating duration(X10 <sup>5</sup> years)	3.00	0.95	2.24	1.56	0.242
Restitution factor of <sup>235</sup> U	0.41	0.47	0.48	0.38	0.11
fission contribution					
<sup>235</sup> U (%)	95.0	96.0	87.7	92.4	74.9
<sup>238</sup> U (%)	2.0	3.0	7.8	3.8	17.9
<sup>239</sup> Pu (%)	3.0	1.0	4.5	3.8	7.2

Table 1. Nuclear parameters to characterize the operation conditions of the Oklo RZs [3-5]

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#### The Oklo natural fission reactors and dynamical models of dark energy

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Paul Dirac is credited with being the first physicist to speculate in print that fundamental constants like the Newtonian gravitational constant G may change over cosmological intervals. Incongruous though this notion may be, it has gained traction among physicists. Modern theoretical frameworks, which attempt to unify all the fundamental forces, accommodate the possibility of varying fundamental constants, as do dynamical models of dark energy. There are intriguing hints, from absorption spectra of interstellar matter, that the fine structure constant  $\alpha$ , which determines the strength of electromagnetic interactions, may vary spatially across the cosmos, the changes found being at the level of about 10 parts per million (ppm). This astrophysical result is in stark contrast to a seminal study (conducted by Thibault Damour and Freeman Dyson) of Oklo neutron capture data which concludes that, in the time since the Oklo natural fission reactors were active (about 1.95 billion years ago),  $\alpha$  has changed by less than 0.1 ppm. There is a tendency to take this Oklo bound *cum grano salis* because of the perception that the nuclear physics invoked in its derivation is fraught with substantial unquantifiable uncertainties. I discuss excitation, Coulomb, and deformation corrections, using deformed Fermi density distributions fitted to the output of Hartree-Fock + BCS calculations (with both the SLy4 and SkM $^{st}$  Skyrme functionals), the energetics of the surface diffuseness of nuclei, and thermal properties of their deformation. Although the net correction is uncertain to a factor of 2 or so, it constitutes no more than 25% of the Damour-Dyson estimate. Making allowance for additional uncertainties in the modelling of the Oklo reactors, I conclude that, subject to a weak and testable restriction on the change in light quark masses, the relative change in lpha over the last 1.9 billion years is less than 0.01 ppm (95% C.L.). This bound reinforces the idea that, of the many dark energy models which predict that fundamental constants do change, only those which suppress the variation of  $\alpha$  in the presence of matter are phenomenologically acceptable.

Parallel between natural Oklo cores and industrial reactors operating

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The first man-made nuclear reactor was developed by Fermi at the University of Chicago and was first started in December 1942. This was the confirmation that one is able to use sustained fission reactions to produce energy. Following this success, the many types of nuclear reactors studied have given rise to several families of reactors corresponding to different orientations and technical choices. They are linked mainly to the choice of fuel (natural uranium, enriched uranium, plutonium), coolant (water, carbon dioxide, helium, sodium), fast or slow of neutrons and moderator for slow neutron reactors (graphite, light water, heavy water).

Out of all these choices the Pressurized Water Reactor (PWR) family is the closest to the Oklo natural reactors. Many intriguing similarities are observed and discussed in the present Supplementary Information C. Our presentday understanding of the PWR operating conditions has been a great help for understanding the Oklo reactors. On the other hand, the fast neutron reactors can also be put in parallel to Oklo cores since they did breed significant amount of plutonium-239 and since some zone are known to be operated as fast neutrons. The presentation will set a parallel between what Nature offered us with Oklo cores and the optimized cores we are able to build and operate.



Figure 1 | Illustration of PWR circuits and Oklo core. The primary circuit is a forced heat conduction loop. The 155 bar pressure is regulated by the pressurizer and prevents water from boiling in primary circuit. The secondary circuit is a water/vapour circuit with a pressure at full power of 71 bar. Steam is produced in the steam generator runs the turbine/alternator before being again transformed into liquid water in the condenser. Approximately one-third of the thermal power is transformed into electrical power and the other two-thirds is exhausted to the cold source (river or refrigerating tower). The Oklo core (figure from [1]) was 2000 m below surface with rather sililar pressure and temperature as the PWR reactors !

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# Session 7

# Radon, Thoron & Decay Products Measurements

# Chair

Peter BOSSEW (Berlin, Germany)

# Invited Talk 7.1

Up-to-date dose conversion factors for radon isotopes and their historical overview

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New radon conversion factors (DCF) for radon/thoron progeny have been presented in the ICRP Publication 137 (2017). There used to be a large difference in the DCF between those derived from epidemiological (ICRP 65) and from dosimetric approaches (ICRP 66). This revision results in a higher DCF than before. Hereafter a variety of radon issues may arise. In the present talk, the following topics will be presented:

- 1. Characteristics of radon and progeny
- 2. How to assess the effective dose due to inhalation of radon progeny
- 3. Lung dosimetry and influential parameters for dose assessment
- 4. Review of dose conversion factors in published data
- 5. Thoron issues

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# **Invited Talk 7.2**

On the use of numerical models to predict/mitigate indoor radon levels in highly contaminated areas

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The publication of the EURATOM directive BSS 2013/59 [1] has increased considerably the interest on radon studies in all EU countries. Certainly each member state has to establish a national action plan addressing longterm risks from radon exposures in dwellings, buildings with public access and workplaces for any source of radon ingress, whether from soil, building materials or water. The complexity generated by the number of parameters and processes affecting radon generation in the source, transport in source media, entry into dwellings, and its accumulation in the different rooms of the dwelling, makes the development of numerical models a very challenging exercise that might take also into account that the detailed information of the buildingsoil interface in an existing dwelling is normally not available. A new project funded by the Spanish Nuclear Safety Board (CSN) is starting in 2020. Its main goal is to establish and validate a numerical tool to predict and mitigate indoor radon levels in new and existing buildings in general, but paying special attention to the case of areas contaminated due to NORM industrial activities. Such a numerical tool might be of interest for Radiation Protection authorities to manage highly contaminated areas. In particular, 2 different numerical modelling strategies will be adapted to real sites and compared. The RAGENA [2] code, which was developed in the late 90s, will be updated with the last findings from experimental studies. This code allows modelling all radon sources and processes affecting radon accumulation indoors from a dynamic point of view in a very simple way, but lacks from spatial resolution. On the other hand, a CFD (computational fluid dynamics) model recently developed in Spain [3] numerically solves radon transport equation by finite elements with a good spatial resolution. The project focuses also on the experimental characterization of real sites. In this talk we will introduce the project, discuss the main features of both modelling approaches and describe in more detail the current status of the RAGENA code updating.

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# Talk 7.1

# Proposal of an affordable method to estimate indoor thoron concentration close to the walls using active radon monitors

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Several continuous radon monitors (CRM) are also sensitive to thoron. This phenomenon, called thoron interference, could lead to overestimate the actual indoor radon concentrations and it is considered an issue for some CRMs, especially for those not able to perform alpha spectrometry of radon daughters. For this reason, radon measurement protocols generally recommend to deploy such CRMs not close to the walls in presence of building materials for which thoron exhalation is expected to be high.

However, thoron interference of CRMs could be used as a mean to detect thoron itself in an affordable way. In this work, we propose a simple indirect method to detect thoron concentration close to the walls using two types of inexpensive CRMs available on the market. For each of these CMRs, firstly we have estimated their thoron interference in an indoor environment having tuff as building materials, i.e. with a not negligible indoor thoron concentration close to the walls. Afterwards, we have estimated their thoron sensitivity using a professional monitor as active thoron reference instrument.

### Talk 7.2

# Some Applications of Novel Polycarbonate/ACF Radon Detectors for Personal and Environmental Monitoring

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Polycarbonate track detector (PCTD) have found wide applications in particular when used in radon monitoring cups [1-3]. In order to overcome some deficiencies such as long-term exposures required, recently Tommasino and coworkers [4] introduced a novel Activated Carbon Fabric (ACF)/CR-39 detector in which the ACF adsorbs radon on its active sites and exposes the CR-39; when CR-39 is chemically etched leads to an ACF/CR-39 response significantly enhanced<sup>[4]</sup>. Using the ACF combined with PCTD, Sohrabi and Ebrahiminezhad have recently introduced electrochemically-etched (ECE) PCTD/ACF multi-function radon individual and environmental monitors<sup>[5-7]</sup>. A comparative PCTD/ACF and PCTD/bare method was applied which also introduced an amplification factor (AF) which can be correlated to radon/progeny equilibrium factor, yet to be further studied and calibrated. The PCTD/ACF registers alpha particles from radon adsorbed on its carbon active sites at a higher rate than that of PCTD/bare which registers alphas from radon and progeny. The ratio of PCTD/ACF tracks to that of PCTD/bare leads to a track density ratio or amplification factor (AF)  $\ge 1^{[5-7]}$ . In this line of development, the methods have been successfully studied for individual and environmental radon monitoring in air as well as radon and radium-226 monitoring in water. In particular, a novel mega-size radon monitoring method using a mega-size radon PCTD detector (33 x 75 cm<sup>2</sup>) [8], processed in a mega-size single-cell ECE image processing system<sup>[9]</sup>, for large area radon monitoring. Another novel development is Long Strip Polycarbonate Radon Monitor with or without ACF methodology processed in a novel Long ECE Image Processing Chamber developed in this research for continuous monitoring of radon; e.g. over a long wall. In this paper, the highlights of such developments are presented and discussed.

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# Session 8

# Nuclear Power Plant Accidents & Risk Communication

# Chair

Mireille DEL NERO (Strasbourg, France)

# Invited talk 8

Exposure of people living around nuclear power plants and other nuclear facilities

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The presence of nuclear plants has been often controversial because of possible health effects on the surrounding populations. In Italy, a total of 9 nuclear plants have been operating, including 4 nuclear power plants which are inactive for many years. The Italian Ministry of Health commissioned a study on the health status of inhabitants of all the 9 municipalities hosting a nuclear plant in their territory using the available population and mortality data.

A mortality study was conducted at municipal level. Several causes of death (single or groups) were analyzed, but particular attention was given to the 24 cancer forms associated with ionizing radiation. Due to the small population sizes, analyses were performed on data aggregated for time periods (1980–1989, 1990–1999, 2000–2008), for municipalities, and for specific cancer sites. The standardized mortality ratio was calculated for each municipality with reference to the corresponding regional population. In addition, the expected number of cancer deaths due to radiation was estimated under three different scenarios of exposure to ionizing radiation due to radioactive emissions from the nuclear plants.

No increase of cancer mortality for the whole group of cancers related with ionizing radiation exposure was observed (SMR 92, 95% C.I. 89–95). However, the analyses for specific causes of death revealed an increase of mortality for thyroid cancer (SMR 150, 95% C.I. 105–210), while a reduced mortality was observed for other cancer sites potentially related with exposure to ionizing radiation. The total expected number of deaths in these populations in the observed period is greater than 1 under the assumption of continuous exposure of all individuals at an effective dose rate of 1 mSv/year.

Mortality in the areas where nuclear power plants were located in Italy are generally similar to that of the reference population but an excess of thyroid cancer emerged. These results will be compared with other studies carried out in Italy and other countries.
#### Detection limit variation against coarse aerosol during airborne contamination measurement with CAM

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In nuclear facilities, the mandatory airborne contamination surveillance is operated by CAMs (Continuous Air Monitors). A CAM samples the ambient air on a filter and measures the deposited activity. It is designed to trig an alarm whenever the measured activity concentration exceeds a defined threshold. However, in some particular sites, such as dismantling sites, a high rate of false alarm is experienced, mainly for artificial alpha [1].

The effect of the characteristics of aerosol deposited on the filter on the nuclear measurement has been studied [2]–[4]. However, it has been shown that false alarms are directly related to the sudden presence of a large amount of coarse particles, i.e. a variation of the aerosol mass size-distribution [5].

Experiments on the ICARE tests bench [6]–[8] have been carried out to characterize the effect of the aerosol mass size-distribution variation on the CAM's performance [9]. A false alarm is mainly due to a wrong detection limit evaluation. Thus, we compared the detection limits in the artificial alpha region; the first is the actual CAM algorithm, the second is calculated as a function of the radon daughters and aerosol characteristics. This new estimation, covering a wider range of aerosol characteristics, shows a significant improvement over the previous one (Figure 1) and highlights the need to take into account the aerosol characteristics for a correct airborne contamination measurement [10]. Moreover, the zone of false alarms occurrence can be now precisely defined between the current (red) and new (black) detection limits in CAM's working condition.

#### Figure



**Figure 1** Comparison of the experimental relationships between the detection limit calculated by the current CAM algorithm (in red) and a new detection limit for which the estimation is based on the aerosol properties (in black). The dotted red line indicates the equality between the estimated and true detection limits. The latter is calculated from the alpha energy-spectrum.

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Grain size effect on radon exhalation rate and uranium activity of Moroccan sediment

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Human is continuously exposed to ionizing radiation emitted from natural radioactivity. This later includes

the cosmic radiation and the terrestrial radiation from the various radioelements (<sup>238</sup>U, <sup>235</sup>U, and <sup>232</sup>Th series) present everywhere in the earth's crust in varying amounts. This variation depends mainly on geological and geographical conditions and appears at different levels in soils from different geological regions. Radon gas is one of the decay products of these series. Once generated, it easily gets released from its birthplace to the pores space (emanation). Driven by various transport mechanisms, such as advection, convection or diffusion, it moves toward the surface and escapes to the atmosphere.

The present study investigates the granulation effect on the radon exhalation rates from a phosphate sample. The phosphate sample was sieved into different grain size dimensions ranging from < 0.063 to >2 mm. The exhalation rate of each fraction was measured using the accumulation method. The radon exhalation rate was found to decrease with an increase in grain size above 0.125 mm. the influence of grain size on uranium content was also studied using the autoradiography method. The uranium concentration increased for the smallest grains (<0.25 mm), reached a maximum for the grains (0.25-0.5 mm), and decreased for the larger grains (>0.5 mm). The measured values of radon exhalation rates are under the limits reported worldwide.

Keywords: Radon exhalation; Grain size; Uranium content; Phosphate

## A drone-borne gamma spectrometry system for environmental radioactivity monitoring

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Different sites in France and all around the world are contaminated with anthropic radionuclides or technologically enhanced naturally occurring radioactive materials (TENORM). The spatial distribution of the radioactivity over these large sites is often totally or partially unknown. One of the main difficulties is to find a good compromise between the precision of the radioactivity measurements and the size of the mapped sites. Deep contamination can also poses data correction problems due to soil attenuation. The IPHC laboratory is currently developing a drone-borne gamma spectrometry system dedicated to the environmental radioactivity monitoring. This system used advanced data analysis algorithms, based on Monte Carlo simulation and machine learning algorithms, to achieve a precise estimate of the different radionuclides activity. The experimental validation of the system will be presented for an artifical anthropic punctual contamination using sealed sources. Future applications for radiation protection or geological studies will also be discussed.

Development of a new material for neutron detection

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Neutron detection is still a challenge. In fundamental research the detection of fast neutrons is mainly based on organic liquid scintillators which offer a good neutron- discrimination. This technology which is now more than 50 years old presents main drawbacks as these materials are toxic, corrosive, flammable, explosive, carcinogenic and dangerous for the environment. They will no longer be usable even at research facilities. Concerning slow neutrons, the shortage of <sup>3</sup>He all over the world leads industrials to find alternative solutions for the <sup>3</sup>He based detectors mainly used. An innovative technology in the detection field is under development in the frame of an interdisciplinary collaboration. It is based on ionic organic compounds which offer the possibility to detect both slow and fast neutrons with the same material. The advantages of these new solid scintillators and their possible applications will be presented.

## Poster 8.1

Calculation tool for iodine 131 biodistribution depending on the aerosol size distribution

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In 2019, the Nuclear Regulation Authority (NRA) in Japan designated the National Institutes for Quantum and Radiological Sciences and Technology (QST) as a Core Center for coordinating and guiding four Advanced Radiation Emergency Medical Support Centers in Japan. If a radiation accident occurs in Japan, QST will be the last port of call offering treatment to exposed people.

Radiation damage to tissues depends on radiation exposure levels. Therefore we have studied accurate estimations of radiation exposure levels so far. Recently, we developed a tool that can calculate the respiratory tract deposition of radionuclides on the basis of polydisperse particle size distribution [1]. As a next step, there is a need for a new calculation tool for the biodistribution of radionuclides because some parts of radionuclides deposited in the respiratory tract are absorbed into the body. We have been trying to develop tools that can calculate the biodistribution of radionuclides in the body lately. In this study, a calculation tool for iodine 131 biodistribution depending on aerosol size distribution developed as our activity is reported.

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## Poster 8.2

# Preliminary study on radon level, ambient aerosol, and external gamma radiation dose rate at lung cancer area in Northern, Thailand

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Lung cancer is the most common insidious disease worldwide and is the major cause of death from cancer [1]. Thailand is one of those countries in which lung cancer has been the leading cause of mortality and healthcare burden compared to other cancer types, especially in the northern region [2]. Chiang Mai Province, the capital of Northern Thailand, lung cancer also is one of the most common causes of cancer mortality and incidence in males and the third for females [3,4]. Moreover, Chiang Mai, the high-risk districts have a problem with high air pollution from particulate matter with a diameter smaller than 10 µm (PM<sub>10</sub>) in northern Thailand [5]. However, the causes of lung cancer in northern Thailand have not been completely understood, but it is strongly believed that they are multi-factorial. The environment is one factor that has played a significant role in lung cancer. Universally, radon is the second leading cause of lung cancer developing after tobacco smoking and the number one cause of lung cancer among nonsmokers, according to World Health Organization (WHO) estimates. Therefore, the initial study of this topic has been obviously carried out in some patients with lung tumors and control case dwellings at Chiang Mai and Lumpang (Thailand).

This study presents the results of indoor radon concentrations using an active detector (AlphaGUARD), ambient aerosol particle size distribution (PAMS Model 3310), and external gamma radiation dose rate using a car-borne survey (3<sup>2</sup>x3<sup>2</sup> Na(Tl) gamma spectrometry). The results show that the indoor radon levels varied from 11 to 18 Bq/m<sup>3</sup>, with an average of 15±2 Bq/m<sup>3</sup>. The maximum, minimum, and geometric mean of the absorbed dose rates in the air by the car-borne survey were estimated to be 47, 171, and 65±12 nGy h<sup>-1</sup>, respectively. In the particle size distribution of ambient aerosols, the modal diameter was found in the accumulation mode (<100 nm) with count median diameter (CMD) and geometric standard deviations (**q**) values of 55–93 nm, and 0.31–0.35, respectively. This is a preliminary result on a small regional scale; therefore, a further detailed study should be undertaken.

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#### Acknowledgments

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## Poster 8.3

## Calibration experiments for radon in drinking water measurements using portable-type electrostaticcollection radon monitors

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We started a new research project on dose estimation for residents of the Pacific coastal area of Fukushima Prefecture from FY2021 [1]. During this project, we will estimate annual effective doses due to inhalation, ingestion, and external exposure from both artificial and natural sources. The doses assessed in this project will be compared to enable residents in the area to judge the influence of the Fukushima Dai-ichi Nuclear Power Plant accident on the overall dose. We plan to collect drinking water samples from 30 houses to estimate an ingestion dose from radiocesium and radon intake. Portable-type electrostatic-collection radon monitors (RAD7, Durridge Co. USA) will be used for in situ measurements of radon in drinking water. For this report, calibration experiments were conducted for quality assurance of radon in drinking water measurements using the portable-type electrostatic-collection radon monitors. Based on our previous study [2], an approximately 10 L sample of groundwater was collected at the Kobe Pharmaceutical University campus, and then the sample was transferred to Hirosaki University. Two RAD7 (SN: 5642 and 5116) were used to evaluate the calibration factors (CFs) and their uncertainties. The CF values were evaluated based on comparative measurements with the liquid scintillation counting method which had been calibrated with a <sup>226</sup>Ra standard source. The water sample was transferred from the sampling vessel to three 250 mL glass vials for each measurement using RAD7. Radon concentrations in water samples were measured using the Wat250 protocol of RAD7. Two calibration experiments were conducted for the following reason. In the 1st experiment, the CF values with standard uncertainties, (k = 1) for SN5642 and SN5116 were evaluated as 0.91 ± 0.30 and 1.30 ± 0.34, respectively. From this, we found that both RAD7 monitors had relatively large uncertainties due to leakage of radon gas after bubbling from the gaps between the lids of the desiccant container and the glass vial. Therefore, these gaps were closed as much as possible using parafilm and clay, respectively. The 2<sup>nd</sup> calibration experiment was conducted for confirmation of the gap closing effect for the measurement system. As a result, we obtained the CF values for SN5642 and SN5116 in the 2nd experiment as 0.76 ± 0.06 and 1.21 ± 0.08, respectively. That is, the relative uncertainties for both RAD7 monitors were decreased respectively from 33% to 8% and from 26% to 6% after closing leakage points of the system. This work is supported by the Research Project on the Health Effects of Radiation organized by the Ministry of the Environment, Japan.

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## Session 9

## Radiobiological Effects, Epidemiology & Health Impacts

**Chair** Lluis FONT GUITERAS (Barcelona, Spain)

Towards the characterization of single-cell molecular response to ionizing radiation

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The focus of the studies of ionizing radiation damage has historically been centered on the direct impact on DNA due to the important mutagenic effects, especially at higher doses. This outlook tends to minimize the potential for cellular damage on other macro-molecules responsible for cellular functions such as RNA which share a highly similar structure with DNA and can thus be considered at least as vulnerable to ionizing radiation. The analysis of transcriptomes offers insight on the cellular response at a given time when exposed to the stress of ionizing radiation and could thus potentially explain the impact of low doses irradiation on cellular behavior. Third generation sequencing devices have made the study of the RNA metabolism more accessible on a larger scale, allowing for direct sequencing of long-read molecules and the identification of base modifications. More recently, single-cell sequencing has become widely available making the analysis of individual cellular response to external stress possible. The combination of these technologies has opened the way for the study of biological and molecular mechanisms at the scale of the individual cell which have yet to be defined in the interactions with ionizing radiation.

The use of charged particle microbeam @ AIFIRA facility [1] emitting 3 MeV protons offers us the possibility to selectively irradiate subcellular or cellular compartments at controlled doses and observe the impact on an organism's development [2]. We are working with the reference organism Caenorhabditis elegans, a nematode with an identical development among individuals, offering a solid frame of reference for comparative analysis. A protocol of immobilization has been established to reproducibly irradiate selected cells on this organism, allowing us to progress towards single-cell analysis. Our current objective is to complete this protocol with sequencing and bioinformatic tools to produce thorough transcriptomic analysis of ionizing radiation induced damage on individual cells.

The main results obtained using those technologies will be presented :

1 - Irradiations of young worms (L1 stage) on their Z1-Z4 cells which develop to form the gonads and reproductive system. These worms grew with varying levels of vulva's structural anomalies depending on the deposed dose.

2 - Flow cytometry analysis by COPAS on worms exposed to whole-organism irradiation at the L1 stage have produced quantitative results on the dose-dependent induced development delays to reach the adult stage.
3 - Transcriptome analysis by direct sequencing of mRNAs extracted 4 hours after irradiation to identify the cell response to the dose-dependent induced stress in terms of expression, epitranscriptome and base modifications.

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# Maternal and paternal exposure to low dose radiation and adverse birth outcomes: preliminary findings from a systematic review and meta-analysis

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Exposure to ionizing radiation is recognized to increase the risk of cancer and cardiovascular disease, while the impacts on adverse birth outcomes are less understood. We undertook a systematic review and a meta-analysis to summarize the epidemiological literature of maternal and paternal exposure to radiation (<5 Gy) and birth outcomes including preterm birth, miscarriage, and low birthweight. This systematic review adhered to the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statement (Liberati et al., 2009). The Population, Exposure, Comparison, Outcome (PECO) framework (Morgan et al., 2007) was followed to help plan the systematic review components, and our literature search included papers published between January 1<sup>st</sup>, 1990, and June 30<sup>th</sup>, 2021. Our search included studies of those exposed to radiation from nuclear disasters, occupationally, medical sources radiation, and individuals who lived near nuclear power plants. The literature search was conducted using four databases (PubMed, Environmental Index, GeoBASE, and the Cumulative Index to Nursing and Allied Health Literature). A quality assessment of the studies was completed, and a meta-analysis was performed to generate summary measures of association using random effects models. Forest plots were generated and we evaluated the potential for publication bias with funnel plots. A total of 26 studies were identified and formed the basis of our meta-analysis. Of these, 10 studies evaluated birthweight as an outcome. The summary meta-regression odds ratio associated with having a low low birthweight child among those exposed to radiation was 1.43 (95% CI: 1.00 - 2.03) relative to those who were unexposed. The corresponding funnel plots were asymmetric suggesting the potential for publication bias. Meta-analyses of other adverse birth outcomes including miscarriage, and spontaneous abortion are currently underway, and will be presented at the conference. The findings from our review suggest that exposure to ionizing radiation may increase the risk of adverse birth outcomes, though these associations should be interpreted cautiously due to a small number of studies, and potential publication bias.

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# Low dose rate radiation exposure effects among medical workers from the knowledge of epidemiologic studies

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In medicine ionizing radiation is used for therapeutic and diagnostic purposes, it inevitably leads to inadvertent exposure of medical workers to a certain level, especially in the earlier years when equipment was less sophisticated and radiation protection was not stringent so the dose received by the medical staff has been greater.

Many people are or have been exposed to ionizing radiation in the course of their work, and epidemiological studies of occupationally exposed populations provide an important opportunity to supplement estimates of the health risks of radiation exposure. In addition, radiation exposure in the workplace often involves exposure conditions directly related to the main problem of radiation protection: prolonged exposure to low-level of radiation. Medical workers who are occupationally exposed to low levels of radiation also provide an opportunity to investigate possible physical health consequences other than cancer, which are currently the subject of much discussion [1].

Epidemiological studies of populations occupationally exposed to ionizing radiation provide important information on radiation-related health risks [2]. The studies that were made on medical workers cover exposure circumstances of the importance for radiation protection, and we should continue to review the risk estimated on which ICRP the International Commission on Radiological Protection are based [3]. In this paper we will provide an overview of recent epidemiological results and ongoing research in the era of non-cancer diseases related to ionizing radiation exposure.

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Ionising radiation exposure of Medical staff: risks and prevention

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Nowadays, the use of ionising radiations in medical exams is unavoidable: whether for radiology diagnostic or radiotherapy. The use of such radiations is in most cases necessary or benefit for patients but the risks to health for medical staff are not always neglected.

In this study, we review the results of some studies in term of radiation doses received by hospital staff in different medical exams: radiotherapy, conventional radiology, interventional cardiology...etc. [1,2]

We dress a mean evaluation of the rate exposition to ionising radiations in each radiological exam with an estimation of the maximal dose received by the medical operator per year.

Finally, we propose, when it is possible, some solutions to minimize radiations risks.

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## Radiolysis of concentrated myoglobin by accelerated ions

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In order to better describe the biological effects of ionizing radiation, understanding the mechanisms of radiolysis at the molecular scale is a key step. Proteins are by far the most abundant biomolecules in the cell, yet very few studies describe their radiolysis by accelerated ions. The aim of our team is therefore to develop a systematic study of these effects on protein biomolecules, from amino acids to whole proteins, and with various ions, energies and dose-rates.



Figure 2 Evolution of the secondary structure of myoglobin under irradiation by 2 MeV protons, as a function of cumulative ion fluence. Error bars were determined with a duplicate of irradiation experiments.

In this work, myoglobin, a small heme-protein, was irradiated by accelerated ions of a few MeV energies, in highly concentrated native gels, 20 % w/w, similar to protein content in the cell. The impact of ions on its secondary structure was followed by mean of infrared spectroscopy, showing reproducible and organized change in its conformation, from alpha helices to mostly beta-structures (cf. Figure 1). UV-Visible spectra were also recorded under irradiation, and the combination of the data allowed identifying the formation of a significant quantity of carbon monoxide under irradiation.

The results obtained with low-energy protons will be presented and compared to helium and carbon ions, and the possible source for carbon monoxide will be discussed.

84

#### Effects of the dose-rate on the radiolysis of water and small protein biomolecules

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Ionizing radiations are known to have important effects on living organisms, as their energy deposition causes a lot of damages. Studying fundamental chemical mechanisms of the effects of ionizing radiations on biomolecules is crucial to have a better understanding of their radiobiological effects. These ionizing radiations can come from radioactive isotopes, or be used in a therapeutical context in radiotherapy to treat cancers by damaging tumoral tissues. Particle therapy, using accelerated ions, is very interesting for its better targetting of tumors compared to classical X-Rays. FLASH radiotherapy, which has been attracting a lot of interest recently, could have a preserving effect towards healthy tissues, using very high dose rates (>40Gy/s) [1]. At this moment, the molecular mechanisms of the FLASH effect are still far from being completely understood.

Our team is developing a systematic study of the chemical effects of water radiolysis species on protein biomolecules, especially under ion irradiation. For this purpose, we compare yields of water radiolysis species (esp. H0<sup>•</sup>,  $e_{aq}^{-}$ , H<sub>2</sub>O<sub>2</sub>) to that of amino acids, peptides and proteins, in order to study the mechanisms of degradation of the biomolecules. Experiments are performed with ions and lowLET ionizing radiations (X-rays,  $\gamma$ , electrons) for comparison, to identify eventual ion specific mechanisms [2]. The effect of dose-rate on the radiolysis mechanisms are also scrutinized.

Hydroxyle radical (HO<sup>•</sup>) is the most potent chemical species towards biomolecules degradation. It has been quantified using scavenging probes at various dose-rates, from 0.1 to about 2000 Gy/s, under irradiation by 1MeV electrons, showing a significant dose-rate effect. Radiolysis of amino acids and of a small peptide, aspartame, was studied in the very same conditions and correlated to the dose-rate effect on HO<sup>•</sup>, as shown in *Figure 1* with the example of the 2,5-dopa, one of the radiolysis products of phenylalanine.





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## **Onsite Poster Session**

Radiolysis of a small peptide in solution by accelerated ions, electrons and X-rays

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Evaluating at molecular scale the effects on biomolecules of ionizing radiations is crucial to better understand their radiobiological impact. The majority of studies on the radiolysis of biomolecules have been carried out with irradiation by photons or electrons. The mechanisms of degradation of biomolecules under irradiation by accelerated ions are still poorly understood, with few studies at the molecular level. Our aim is to describe these mechanisms for protein biomolecules (amino acids, peptides and proteins), which are by far the most abundant in the cell. The results obtained are of interest to understand the effect of ions produced by radioactive elements, as well as in a context of particle therapy using accelerated ions to kill a tumor.





At the molecular scale, water radiolysis species, oxygen and biomolecules radiolysis products may be all engaged. Our team has shown that phenylalanine radiolysis by ions led to the formation of specific products, not observed with photons or electrons [1].

The aim of this work was to study how these mechanisms may be modified when the amino acid is included in a peptide. Aspartame, a small dipeptide containing phenylalanine and aspartic acid, was irradiated by ions (carbon ions, helions and protons), and 6 MeV X-Rays, 1 MeV electrons for comparison. The radiolysis products were identified and quantified, and their radiolytic yields were compared to that of phenylalanine alone, showing similar mechanisms (Figure 1). Effects of the dose-rate on the radiolysis of aspartame was also studied.

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Assessment of radioactivity levels and radiological hazard indices in phosphate samples from Algeria

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Phosphate ore is the starting raw material for production of all phosphate products including phosphate fertilizers. It can be of sedimentary, volcanic or biological origin. Like any other geological material found in nature, it contains various amounts of naturally occurring primordial radionuclides, such as <sup>238</sup>U series, <sup>232</sup>Th series and <sup>40</sup>K.

The assessment of natural radioactivity levels and radiological hazard indices in phosphate samples has been done using gamma spectrometry. The Metrological aspect will thus be undeniably strengthened, even for environmental matrices with a majority organic or mineral component. This is also essential for the monitoring of environmental radioactivity. In this work, the specific activity concentrations of <sup>238</sup>U, <sup>232</sup>Th and <sup>40</sup>K of phosphate ores and fertilizers samples were determined. Based on the activity concentrations, absorbed gamma dose rate in air (D), Radium-equivalent activity (Ra<sub>eq</sub>), hazard indices (H<sub>ex</sub>, H<sub>in</sub>) and annual effective dose (AED) were calculated and compared with the recommended safety limits.

Comparison of experimental and Geant4-DNA simulated data of yields of water radiolysis species

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This study aims at better evaluating the indirect effects of water radiolysis on the ionizing radiation damage mechanisms. Under irradiation of a cell, ionizing radiations will generate radicals, which will yield damages to biomolecules, including proteins and DNA. Our body is mostly made up of water (~65 – 70%), proteins (~20%), lipids (~10%), with DNA accounting for only 0.1%. When an ionizing radiation enters a cell, it can either interact directly with a biomolecule (direct effect) or interact with water, resulting in the formation of reactive species by radiolysis of water. These reactive species will then react more or less quickly with the surrounding biomolecules, inducing damage by indirect effect. Given the proportion of water, indirect effects will play a very important role in the phenomena occurring under irradiation in the cell. The precise quantification of water radiolysis species is therefore essential for understanding the mechanisms of damage formation to biomolecules. Regardless of the ionizing radiation (accelerated ion, electron, X-ray or gamma), reactive species generated by radiolysis of water are about the same: hydroxyl radical H0', hydrogen atom H', hydrogen peroxide H<sub>2</sub>O<sub>2</sub>, and hydrated electron  $e_{aq}$ .

In this poster, we will present the production yield kinetics of hydroxyl radical HO<sup>+</sup>, hydrogen peroxide H<sub>2</sub>O<sub>2</sub>, and hydrated electron e<sub>aq</sub><sup>-</sup>, on time scales from nanosecond to microsecond. Experiments with 6 MeV X-rays (ICANS – Strasbourg) were realized simultaneously and under the same conditions for all species with specifics probes and kinetics were obtained with variable molar concentration of scavengers. A same experiment are underway with 2 MeV protons (ACACIA – Strasbourg). Experimental data of reactive species have been compared to Monte Carlo simulation results produced with the Geant4-DNA software [1], an extension of Geant4 that was developed to simulate physical, chemical and biological effects of ionizing radiation on DNA.

#### References

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Study of the mobility of uranium (VI) and organic matter of a soil from an enhanced natural radioactivity area. S. FERRERES <sup>1\*</sup>, L. FEVRIER <sup>2,3</sup>, V. CHAPON <sup>2,4</sup>, O. COURSON <sup>1,2</sup>, S. MEYER-GEORG <sup>1,2</sup>, M. DEL NERO <sup>1,2</sup>

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A better understanding of the links between Uranium (VI) speciation, its lability, and biodisponibility is needed to understand its fate in enhanced natural radioactivity environments. The objectives of this thesis work are to identify the effects of organic matter (OM) – a complex supra-molecular blend of thousands of organic molecules, with diverse chemical properties and reactivity –, and bacterial communities, on U (VI) adsorption on minerals and plant uptake, by using laboratory-controlled experiments.

This study focused on U, and other metals, lability in a TE-NORM enriched horizon of a soil from the wetland surrounding the former uranium-mining site of Rophin, in function of pH and desorbed OM composition. In CO2-free conditions, desorption studies were carried out using several pH, ranging from 3 to 6. U and trace metals concentration have been measured by ICP-MS. Synchronous and 3D fluorescence spectroscopy were also performed to determine the macroscopic properties of the desorbed OM.

First results show that U concentration, but also the quantity and type of OM desorbed is pH-dependent: proteinlike and fulvic-like components are desorbed in the whole pH range, but the mobility of fulvic-like and humiclike components are favored at higher pH.

Other experiments, using ESI-FTMS and synchronous fluorescence, are also performed on OM extracted from this horizon, to achieve both a good yield and a good molecular representativeness. These results will permit to begin laboratory-controlled experiments and help understanding the lability results.



#### Figures/Tables (If necessary)

Fig.1 : Effect of pH on desorbed organic matter (OM) from the studied horizon of Rophin wetland's soil, analyzed by synchronous fluorescence.

## Mechanistic study of Myoglobin and Apo-Myoglobin radiolysis by accelerated ions

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Hadron therapy is an alternative radiotherapy technique for cancer treatment. It offers a better irradiation ballistic than conventional techniques using X-rays, with a maximum dose deposited close to the end of the ion's track. In addition to the damages caused by direct ionizing events to the biomolecules existing in cells (DNA, proteins, lipids...), a significant part of the damages results from the radiolysis of water and the generation of reactive oxygen species and hydrated electron. The result of direct and indirect effects is the development of biological and physiological alterations of the cell. Elucidating and modelling mechanisms of radiolysis by accelerated ions at the molecular level are thus of paramount importance.

The aim of our study was thus (i) to characterize the damages induced by energetic  $C^{6+}$  and  $H^+$  ions on myoglobin, a cytoplasmic hemoprotein which is present in skeletal muscle fibers and plays a vital role in oxygen storage and transport and (ii) to compare the indirect effects of accelerated ions with those of high energy photons.

Experiments were performed on a 4 MV Van de Graaff accelerator (ACACIA, Icube, Strasbourg, France) and on the Japanese synchrotron HIMAC (QST, Chiba, Japan). Solutions of irradiated proteins were analyzed by HPLC-ESI-MS using top-down and bottom-up approaches. The evolution with dose of the secondary structure of the proteins was monitored by circular dichroism.

With photons, important alterations of the myoglobin polypeptide chain were detected, including oxidation of methionine residues and multiple hydroxylations (mainly on tryptophan, histidine, proline residues). On the other hand, with ions, the damages were found to converge to the heme moiety, leading to a covalent crosslinking of the heme to the protein. By comparison, apo-myoglobin, that is myoglobin without its heme, shows much higher levels of hydroxylation in identical irradiation conditions, even with ions. Important modifications of the protein's secondary structure were observed especially with -ray irradiations, with a conversion of -helices to -structures and random coils.



**Figure 1.** ESI-MS analysis of myoglobin (Mb) after irradiation with 2 MeV H<sup>+</sup>, showing the covalent crosslinking of heme to the protein (Mb-Heme).

Study of the speciation and mobility of uranium (VI) in natural waters : Effect of organic matter

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Understanding the mobility of Uranium (VI) in natural surface waters in NORM and TE-NORM zones requires a thorough knowledge of uranyl species in presence of natural organic matter (NOM).

Studies, via U mobility and metal-organic speciation analyses, have been carried out in the stream waters of the catchment area of the former uranium-mining site of Rophin (S. Meyer-Georg, 2021). Elemental, microscopic (TEM-EDS), and spectrometric analyses of the dissolved and colloidal fractions of the waters were performed, including analyses of organic matter (OM) by synchronous fluorescence spectrometry and electro-nebulization source ionization high resolution mass spectrometry (ESI-FTMS).

U (VI) shows seasonal and spatial dynamics in the Rophin stream. In particular, the wetland was found to be a secondary source of U to the stream water flowing through it. An important scientific breakthrough was made on the transport mechanisms of U(VI) in the water flowing through the wetland: U(VI) migrates there in the form of suspended pseudo-colloids. It is associated with a colloidal fraction of size 0.45µm-10 kDa made up of iron oxides of 10-20 nm covered by an organic gangue with a strong humic component, and with a colloidal fraction of size 3-10 kDa, which is organic. U(VI) is therefore co-adsorbed with humic acids on colloidal iron oxides, and complexed by organic colloids (with humic and fulvic components). These colloids are the vectors of U (VI) transport in the Rophin stream.



Fig. 1: Filtration results of E4-E5 water samples at the Rophin wetland (May 2019): effect of filtration threshold (a) on U and Fe concentration (insert), (b) on organic matter components analyzed by synchronous fluorescence (PL: protein-like, FL: fulvic-like, HL: Humic-like), and (c) TEM image of iron oxide colloid aggregates surrounded by an organic gangue (S. Meyer-Georg, 2021).

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# Spatial variability in large buildings of university campus: a case-study to identify the best measurement protocols

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In light of the upcoming transposition into national law of Council Directive 2013/59/Euratom [1] (EU\_BSS in the following), Italian National Institute for Insurance against Accidents at Work and the Basic and Applied Sciences for Engineering Department of Sapienza – University of Rome have recently promoted a radon survey in workplaces of university buildings. The survey has interested 11 buildings of the same Department campus, all with similar building characteristics and grouped within less than 1 km<sup>2</sup>.

The measurements have a duration of one solar year to evaluate the annual average of radon activity concentration in air, as required by the EU\_BSS. Rooms sampled for radon measurements (i.e. the overall sample) have been split into two subsamples: for the first group measurements have been performed for 2 consecutive 6-mo periods, for the second one, measurements have been going through 4 subsequent 3-mo periods. Censed rooms, among whom the overall sample has been picked, are more than three hundred workplaces whose intended use is always clearly identified: administration and professors' offices, research and educational laboratories, conference rooms and classrooms.

Spatial variability of indoor radon concentration is evaluated among the buildings in campus (*i*), inside each building among the different floor levels (*ii*), and finally, for each floor level, among each rooms (*iii*). In doing so, separate analyses are carried out for the intended uses identified during the census in order to make results independent from specific occupancy pattern of workplaces.

In view of the results of such analyses, different measurement protocols are evaluated in terms costeffectiveness.

## References

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### POSTER 8

# Evaluation method of natural gamma dose rates using an Nal(Tl) spectrometer under coexistence of radioceasium and natural radionuclides

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Release of radioactive materials into the land by nuclear accidents elevates ambient gamma dose rate levels in addition to terrestrial gamma radiation from natural radionuclides. In-situ measurements based on gamma-ray spectrometry are frequently applied to evaluate ambient gamma dose rates from artificial and natural radionuclides separately and, consequently, evaluate additional external radiation exposure. However, in a highdose-rate area, the ambient gamma dose rates from natural radionuclides are overestimated using an Nal(Tl) spectrometer because the energy resolution of the spectrometer is low and sum peaks of artificial radionuclides overlaps the photon peaks of natural radionuclides (e.g.,  $^{40}$ K) in a gamma-ray pulse-height distribution. In the present study, gamma-ray pulse-height distributions were measured indoors and outdoors using a 3 in × 3 in NaI(TI) spectrometer in Chiba Prefecture and Fukushima Prefecture, Japan, including high-dose-rate evacuation areas of Okuma Town and Namie Town (dose rate: < 0.1  $\mu$  Sv.h<sup>-1</sup> to 25  $\mu$  Sv.h<sup>-1</sup>) where <sup>134</sup>Cs and <sup>137</sup>Cs mainly contributed to the dose rates. In addition, gamma-ray spectra were derived from gamma-ray pulse-heigh distributions unfolded by a response matrix method, and the gamma-ray fluxes at energies of <sup>40</sup>K, <sup>214</sup>Bi (in the <sup>238</sup>U decay chain), and <sup>208</sup>Tl (in the <sup>232</sup>Th decay chain) were compared with respect to ambient gamma dose rates. The results showed that the gamma-ray fluxes at the energies of 40K and <sup>214</sup>Bi varied depending on the ambient gamma dose rates more than 2  $\mu$  Sv.h<sup>-1</sup> and 5  $\mu$  Sv.h<sup>-1</sup>, respectively, while those at the energy of <sup>208</sup>Tl were nearly constant over the range of 0.1  $\mu$ Sv.h<sup>-1</sup> to 25  $\mu$  Sv.h<sup>-1</sup>. This finding means that the sum peaks of radioceasium overlapped photon peaks of <sup>40</sup>K and <sup>214</sup>Bi, but they did not affect those of 208Tl. In addition, the gamma-ray fluxes were well correlated with the natural gamma dose rates. This indicated that the natural gamma dose rates can be estimated from the gamma-ray fluxes at the energy of 208Tl even using the Nal(Tl) spectrometer with relatively low energy resolution compared to HPGe detectors in the ambient gamma dose rate up to 25  $\mu$  Sv.h<sup>-1</sup>. Based on the findings, the natural gamma dose rates were estimated as 0.02-0.06  $\mu$ Gy.h<sup>-1</sup> and 0.02-0.07  $\mu$  Gy.h<sup>-1</sup> in Okuma Town and Namie Town, respectively.

# Assessment of Potential Activation in Food Products Irradiated with High Energy X-Rays: Experimental and Modelling Approach

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High energy X-rays produced by accelerators are increasingly used for the food sterilization. More penetrating than electrons, easier to manage than radioactive sources, X-rays are very interesting for industrial processes. One of the main current concerns is the choice of the maximum energy of the electron beam that produces X-rays. The higher the energy, the more efficient the process (X-rays production and food sterilization). However, it is essential to take into account the risk of photonuclear activation which can lead to the production of radioactive nuclei inside the irradiated sample.

Most of the reference publications, including the IAEA report "Natural and Induced Radioactivity in Food" (IAEA-TECDOC-1287), conclude that food irradiated with X-rays up to 7.5 MeV to a dose of 30 kGy has a radioactivity well below natural radioactivity in non-irradiated food. These very low levels of induced radioactivity make it necessary to question the possibility of developing a tool for easily and efficiently controlling industrial food sterilization process. Experimental measurements by gamma spectrometry are a useful but insufficient solution because it does not detect the presence of pure beta or alpha emitting radioactive nuclei, which are much more difficult to measure. Monte Carlo simulation codes for radiation-matter interactions and analytical codes for activation calculation are thus considered to be more powerful and practical tools for quantifying the radioactivity potentially induced by irradiation. The objective of this study was to estimate the reliability of these numerical calculations, and to propose a methodology allowing a step-by-step experimental validation of a future industrial solution to control the induced radioactivity.

We first identified the three main critical steps for the accuracy of the calculations: photonuclear data (crosssections), photoneutron production and activation calculations. By comparing the results from Monte Carlo (Geant4 and MCNPX) and analytical (FISPACT-II) codes, we have highlighted the importance of experimentally validating the outputs obtained at each of these three stages. One of the most critical parameter is the X-ray energy distribution. As photonuclear thresholds are between 6.5 and 8 MeV for most of the nuclei, only photons in the tail of the X-ray distribution will be involved. It is so needed to model very precisely not only the shape but also the maximum energy of the X-ray spectrum.

Several experiments made it possible to test various protocols that could be used for the validation of a numerical activation calculation tool. For the X-ray energy distribution, we showed that the measurement of the depth dose distribution (as performed in radiotherapy controls) is not sufficient. This measurement must be supplemented by a measurement of the maximum energy of the X-rays, which could be carried out through the use of materials with different activation thresholds.

This work conclude that the progress made in the fields of Monte Carlo simulation and activation calculations over the last twenty years now makes it possible to develop a reliable and precise numerical tool for controlling radioactivity in food products irradiated with high energy X-rays. The complexity of the calculations and the importance of critical parameters, such as the X-ray energy spectrum, necessitate however the definition of an experimental protocol (including neutron measurements) making it possible to regularly ensure the validity of the calculations.

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