Terms of Reference for Radioecology WG

Title and acronym:

Atmospheric Radionuclides in Transfer Processes

Topical area

The Fukushima accident has highlighted several lacks considering air transport and inhalation dose assessment, deposition stage from atmospheric releases or long-lasting secondary emissions from previous deposits. Inhalation dose assessments have suffered from the relatively low-number determinations related to the gaseous iodine contribution. Attention should also be paid on the health effects of the particles, based on their size distribution, composition, crystalline structures, oxidation state influencing weathering rates and solubility in soil-water and sediment-water systems. Adachi et al. (2013) have found highly cesium-labeled airborne particles up to 2.7µm far (180 km) from the damaged NPP. Information on the source term and release scenarios is essential as input to atmospheric transport, and particle codes should be implemented in transport models. Regarding atmospheric processes, some peculiarities of winter time meteorological conditions such as snow and fog events have shown to be responsible for additional radionuclide deposition in some parts of the northern rim of the Kanto plain, Japan (Hososhima & Kaneyasu 2015). The European situation is not very different from that of Japan both regarding the current capability to attest for a gaseous contamination and specific wintertime meteorological conditions. Such conditions would provide the same interactions of snowflakes or fog droplets with radionuclide-labeled aerosols in case of an accident release. Finally, the long lasting post-accident stage would be also characterized by secondary emission through resuspension, biomass burning and biogenic aerosol production that remained to be fully understood despite some preliminary works performed after the Chernobyl accident.

Leadership

Olivier MASSON (IRSN, France) and Jochen TSCHIERSCH (HMGU, Germany)

Partners with a brief description of their assigned role

Interested Organisation	Assigned role
IRSN (France)	WG coordination; Field experiments on fog deposition, secondary emissions of biogenic aerosols; Improvement, test and validation of gaseous sampling/measurement aimed at lowering detection limits ; Reference size distributions for low level anthropogenic radionuclides; Atmospheric ^{210}Po baseline.
HMGU (Germany)	WG coordination; Field experiments on snow deposition; Model implementation of deposit and release of winter time contamination; Retrospective size determination; Parameterization of deposition/resuspension processes for post accidental forecast; Tracing of hot particles after the Fukushima accident.
CERAD (Norway)	Characterization of radioactive particles released from a series of different sources. Implementing particle codes and characteristics (particle properties) in air transport models. Linking particle characteristics to weathering rates/solubility using extraction techniques. Testing cascade impactor in contaminated areas. Modelling of air transport and deposition of particles.
CIEMAT (Spain)	Field experiments on resuspension using time series of radionuclide contents in soils/air; Study of ^{239}Pu/^{240}Pu, ^{137}Cs, ^{226}Ra ^{210}Pb, ^{210}Po and U fluctuations in air particles; Study of Pu sources in air through their isotopic ratios, and retrospective analysis;
NCSRD Demokritos (Greece)	Monitoring of ambient levels and size distribution of radioactive tracers in the atmosphere. Studies of submicron radioactivity as tracers for atmospheric processes; Field testing and development of methods for size resolved aerosol radioactivity; Seasonal variability and long range transport of natural and anthropogenic radionuclides in the Eastern Mediterranean; Studies at high altitude and ground atmosphere; Wet scavenging and deposition.
SCK-CEN (Belgium)	Based on field experiments on snow deposition and the model simulations, sensibility and uncertainty analysis of the parameters and models involved in the research. Definition of strategies for conceptualization, calibration and validation of models for different scenarios with focus on key parameters identified after the sensitivity and uncertainty analysis.

Starting date and estimated duration of the WG to accomplish its plan

Starting date: September 30, 2015

Duration of the WG: 5 years

Intended activities

The intended activities contribute to 4 main tasks:

Task 1: Deposition

This task is devoted to the definition of relevant range of values that makes it possible to characterize the importance of fog deposition, snow deposition, dry vs. wet only deposition in order to prioritize and assess deposition mechanisms according to meteorological conditions. It includes knowledge of activity levels in the air (total particle content as well as according to various particle size ranges) and in water (snowflakes, raindrop, fog droplets). In Japan, it has been recognized that locations that showed significant differences between deposition computations and field observations had encountered snow or fog. This finding highlights 1) the contribution of such events to ground contamination and 2) that models do not specifically take them into account. Very recent observations confirm the high radionuclide deposition potential of fog events on vegetation on a yearly basis for frequent foggy locations) compared with annual rain deposit.

Particle size range is also of main importance with regard to the deposition velocity of radionuclide-labeled aerosols. It has also a great importance on inhalation intake and subsequent dose assessments.

Approach: Mostly based on field experiments and when possible on laboratory experiments to fix the analytical values for microphysical parameters in controlled conditions. Parameters shall be useful for implementation in existing decision support systems.

Steps to accomplish:

- at IRSN: Study on rainout process based on fog/cloud deposition of gamma emitter radionuclides and Tritium on plants. Size distribution of radionuclides in normal situations as reference values.
- at NCSRD Demokritos: Study on wet only vs. dry only deposition of radionuclides. Size distribution of radionuclides.
- at HMGU: Study on snow deposition of radionuclides and its implementation in forecast models. Retrospective size distribution from nebulization of filter solutions.
- at SCK•CEN : definition of strategies for conceptualization, calibration and validation of models for snow deposition of radionuclides based on sensitivity and uncertainty analysis.

Outlook:

Future step will concern tritium deposition by fog and cloud. Surface deposition on various kinds of vegetation (roughness, leaf area index) will be investigated based on a large variety of plant. The determination of deposition velocities will be assessed through different innovative method (to be tested) including Particle Image Velocity (PIV) and deposition flux by eddy correlation and compared with classical approach (mass measurements).

Refinements will also concern the characterization of snowflakes according to micro-physical parameters including different shape descriptors of the hydrometeors. Proposals will be developed to improve parameterization in existing decision support systems for a better forecasting of deposition in emergency situations.

Expected outcomes

All these knowledge will be aimed at filling lack of knowledge with respect to meteorological conditions (e.g. winter scenarios with snow and fog) we could experience in Europe, providing typical range of operational values for relevant parameters (rainout, washout and snowout coefficients, deposition flux and velocities, collection efficiencies) during routine or incident/accident situations.

Task 2: Gaseous species

This task concerns improvement of various steps from sampling to measurement in order to lower the detection limit of gaseous species that represent most of the total (gaseous + particulate content) as for iodine. The iodine gaseous fraction remains usually predominant but still suffers from high detection limits (about 100 – 1000 times that for the particulate fraction). After the Fukushima accident the number of gaseous samplings was only about 20% of that for aerosol in Europe. Even if the consequences of the Fukushima accident was of no concern for public health in Europe It can be asked on the reliability of dose assessment when 80 % of the concentration is missing. According to dose coefficients (ICRP 71), it can be demonstrated that including gaseous iodine (I_2 or CH_3I) to particulate iodine in inhalation dose assessment will multiply the inhalation dose by a factor of 10 to 20 depending on the gaseous species considered.

Apart from severe accident releases, some recent incidents (November 2011, February 2012, March 2015) in Europe were responsible for large scale spreading of ^{131}I traces on the European scale. It is known that iodine is volatile and remains predominantly in its gaseous form in the atmosphere. However, the ratio gas/particle remains difficult to assess and then any attempt in inhalation dose assessment will be inevitably associated with large uncertainties. Moreover existing gaseous sampler are rarely equipped with heater that could lower the competition between iodine and water vapour. This may lead to underestimation. Further improvements, such as gaseous speciation and trapping of other gaseous radionuclides will worth to be investigated and could benefit from preliminary improvements performed on ^{131}I.

Approach: Mostly based on laboratory experiments

Steps to accomplish:

1) Selection of the most efficient adsorbent regarding its capability to trap efficiently gaseous iodine in humid conditions. Definition of the size and shape of the trap taking into account both the required flow and the size of the detector. Compared with aerosol samplers, some of them having flow rate of several hundred m^3/h, most commercially available gaseous sampler have flow rate between 3 and 10 m^3/h). Characterization of the optimum detection efficiency by Monte Carlo simulation. Prototype building: sampling tests, measurement tests and optimization

   Semi-industrial/industrial building.

2) Iodine sorption / desorption kinetics on and from aerosol particle. Possible desorption after deposition.

Outlook:

1) Iodine chemical speciation (I_2, ICH_3, Ox, particulate) with high flow rate (and AMS measurements for ^{129I}),

2) Other gaseous compounds (Ar, Kr, Xe, Rn) pre-concentration steps.

Expected outcomes:

1) *Technical: Lower detection limits of gaseous iodine down to 1-10 µBq/m3. Improve chemical speciation for a better dose inhalation assessment. Promote harmonization of sampling equipment. Develop multi-gas sampler*

2) processes: knowledge on the gas/particle ratio based on routine ^{129}I releases and transposition to ^{131}I

3) Monitoring strategy: promote routine gaseous monitoring in Europe at a minimum number of sites. In a more effective way, gaseous sampling could be achieved but measurement could occur only if traces are detectable on the particulate form (i.e., on aerosol filters).

Task 3: resuspension and re-emission mechanisms

This task aims to have a better understanding and estimate of the long-term behavior and fate of radionuclides in the atmosphere long after their initial deposition on terrestrial ecosystem. The Japanese society has expressed her strong will for land retrieval and land occupancy. This wish could be disrupted or delayed due to the observation of peaks of airborne activity levels from time to time in Japan. Thresholds for land retrieval based on airborne concentrations may be exceeded on a more or less temporarily period depending on the resuspension or re-emission process involved. Secondary emissions encompassing wind resuspension, biomass burning or biogenic aerosol production (wax exfoliation, pollens, …) are clearly involved in those peaks and in the long lasting persistence of airborne radionuclides in the ground-level air.

The main mechanisms involved are:

- resuspension by wind erosion and human activities (e.g. remediation actions); biomass burnings encompassing combustion of agricultural waste and burning of branches for land clearing, and use of contaminated wood for heating purpose;
- biogenic aerosols (virus, pollens, spores, wax exfoliation)

This task will also cover volcanic eruptions (for thorium, uranium and polonium releases) since there exist only some scarce determinations of their average airborne levels in normal conditions. The 2010 and 2014 volcanic eruptions in Iceland caused important airplane traffic disturbances. ^{210}Po represents a specific volcano tracer and could at least be used to attest the presence of plume residues for insurance activities related to air traffic, and would worth for dose assessment computation for people embedded in plume.

Again, the knowledge of the size distribution will be helpful to characterize the resuspension of radionuclide-labeled aerosol.

Approach:

Mostly based on field experiments and when possible on laboratory experiments (wind tunnel or fire room) to fix the analytical values for microphysical parameters in controlled conditions.

Steps to accomplish:

- Lower the large uncertainties of resuspension coefficient by sorting main parameters (wind speed, vegetation cover, soil type, soil moisture or dryness…),
- Characterization of secondary biogenic emissions depending first on season (pollens, mould, yeast, fungi…) whose size ranges between hundreds nanometers and few micrometers),
- Use of cascade impactors in contaminated areas and in clean areas (to compare with the regional background distribution).

Expected outcomes:

Typical range of values for resuspension coefficient and emission factors according to the magnitude of mechanical parameters (wind speed, shear stress, roughness…) and biogenic production.

^{210}Po baseline in the atmosphere in order to characterize future eruption events.

Time series of air-particles compared to radionuclides (natural and artificial) fluctuations.

Task 4: Plutonium persistence at trace level and time series reconstruction; Characterisation of radioactive particles released from different sources

<u>Approach</u>:

Following high and low temperature nuclear events a large fraction of refractory radionuclides such as uranium and plutonium is present as particles, ranging from submicrons to fragments. The particles can contain a series of radionuclides (fission and activation products) as well as stable metals. These particles serve as input to atmospheric transport models. These particles can carry a substantial fraction of radioactivity, and following deposition, these particles can act as point sources of radiological concern. Deposition of particles in the environment may delay ecosystem transfer. Thus, information on particle weathering rates is essential for assessing long term transfer of particle associated radionuclides.

<u>Steps to accomplish</u>:

- Linking particle characteristics to source and releases scenarios: use of advanced techniques for particle characterisation, and determine particle weathering rates using extraction techniques
- Determination of atom/isotope ratios for source identification (ICP-MS, AMS)
- Possibility of using software for summing spectra of the same sample collected in different periods of time to detect peak (no detectable on individual samples) and to calculate average values.
- Comparison of Soil/Air Plutonium-239 and Plutonium-240 concentrations.

<u>Expected outcomes</u>:

Retrospective time series reconstruction of airborne Pu isotopes,

Sources identification based on atom/isotopic ratio analysis,

Work plan

Partner	Concerned tasks	Means, fundings	Resources
IRSN (France)	Coordination Task 1 Task 2 Task 3 Task 4	 T1: IRSN PhD T2: IRSN, NEEDS-Enviro. T3: IRSN, T4 : IRSN,	T1: 12 + 36 pers.months (PhD) T2: 72 pers.months T3: 12 pers.months T4: 6 pers.months
HMGU (Germany)	Coordination Task 1 Task 2 Task 3 Task 4	 T1: TransAqua, VAO II, HARMONE T2: HMGU T3: TransAqua T4: TransAqua	 T1: 72 pers.months T2: 6 pers.months T3: 12 pers.months T4: 24 pers.months
CERAD (Norway)	Task 3 Task 4	T3: CERAD T4: RATE, CERAD	T3: 6 pers months T¤: 48 pars. months

CIEMAT (Spain)	Task 3 Task 4	T3: UE/National? T4: UE/National?	T3: 24 pers.months T4: 24 pers.months
NCSRD Demokritos (Greece)	Task 1 Task 3	Task 1 : national Task 3 : national	T 1: 36 pers.months T 3: 12 pers.months
SCK-CEN	Task 1	Task 1 : national	T 1: 36 pers.months

Cited projects:

NEEDS- Environnement: French program supported by (CNRS + ANDRA +EDF + IRSN)

TransAqua: German joint project supported by Federal Ministry of Science and Education

VAO II: German program supported by Bavarian State Ministry of Environment

HARMONE: European project funded via OPERRA

Major elements of the communication plan (workshops, publications, guidance documents…)

Partner	
IRSN (France)	COMET meetings, workshops, conferences, publication in international journals
HMGU (Germany)	National workshops, intern. conferences, publication in international journals,
CERAD (Norway)	workshops, conferences, publication in international journals,
CIEMAT (Spain)	workshops, conferences, publication in international journals,
NCSRD Demokritos (Greece)	workshops, conferences, publication in international journals,
SCK-CEN	workshops, conferences, publication in international journals,

Links with other activities identified at the national and the international levels

Partner	
IRSN (France)	Ro5 (Ring of Five network) In Japan: Fukushima University, Meteorological Research Institute, National Institute of Advanced Industrial Science and Technology, Japan Atomic Energy Authority, Ibaraki University
HMGU (Germany)	IAEA-CRP "Environmental Behaviour and Potential Biological Impact of Radioactive Particles"; Ro5 In Japan: Japan Atomic Energy Authority, Fukushima University
CERAD (Norway)	IAEA-CRP "Environmental Behaviour and Potential Biological Impact of Radioactive Particles"
CIEMAT (Spain)	Ro5, ALMERA (IAEA), National Net of environmental radioactivity control, Environmental Radiological Surveillance in Nuclear Installation, National labs Intercomparison Evaluation, DOE-USA
NCSRD Demokritos (Greece)	Ro5
SCK-CEN	Crisis centre of the Federal Public Service (FPS) Internal Affairs, Belgium

Expected problems, gaps/lack of knowledge, etc. that might prevent the accomplishment of the research

As a general comment, airborne activity concentrations are at trace levels (order of µBq/m^3 or less, except for naturally occurring radionuclides). All studies presented here require high-sensitivity detection equipment to overlap detection limits. This is especially the case for Pu isotopes which are sample-consuming and require large number of filter samples.