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Report on the potential impact of particle contamination on site-specific transfer factors (Kd, CR/BCF) and dose estimate

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1 Executive summary

Due to the existing gaps in the knowledge about environmental behaviour of particle radioactive contamination affecting some ecosystems over the world, efforts have been devoted to quantify the processes of particle transformation in the environment and radionuclide leaching into various media to better evaluate ecosystem transfer in ecosystems affected by radioactive particle contamination. With this end, systematic transformation experiments exposing well characterized particle samples to environmentally relevant media have been performed thus providing weathering rates to be linked to site-specific particle characteristics.

This quantification has been performed by applying well defined abiotic and biotic leaching protocols to isolated radioactive particles of different origin previously characterized by means of advanced techniques (e.g., autoradiography, ESEM-EDX, TOF-SIMS, synchrotron radiation based x-ray techniques). The information gained in these characterizations, such as elemental composition and distribution as well as crystallographic structure and oxidation states, was linked to particle weathering rates, extraction kinetics and potential bioavailability of remobilised particle associated radionuclides.

The obtained results are the basis for new approaches to evaluate the potential impact of particle contamination on site-specific transfer factors (Kd, CR/BCF) and dose estimate, because traditional assessments based on average bulk mass or surface activity concentrations of radionuclides in environmental compartments (Bq/kg, Bq/m^2 or Bq/L) would not be valid.

In particular, through the research performed the biological relevance of radioactive particles affecting some particular ecosystems has been highlighted. Particles can be inhaled or ingested, and can be retained within organisms, acting as point sources, as has been demonstrated in this project in several field experiments. This fact is very important because until now bioavailability and uptake of particle-bound radionuclides compared with those existing as ions or simple molecules has so far largely been ignored when impact and risk are assessed. As a result, there is a high degree of scientific uncertainty about the long-term ecological consequences and risk to human health from radioactive particles present in the environment. Advances in the mitigation of these uncertainties have been reached, but it is necessary to devote efforts to improve dosimetry models for proper internal dose evaluation associated to high-specific activity particles.

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3 Introduction

History has shown that a series of different nuclear sources associated with the nuclear weapon and fuel cycles have contributed to the release of radioactive particles to the environment (Salbu et al., 2015). Following nuclear weapon tests, safety tests, conventional destruction of weapons, and reactor explosions and fires, a major fraction of released refractory radionuclides such as uranium (U) and plutonium (Pu) were present as entities ranging from sub-microns to fragments. Furthermore, radioactive particles and colloids have been released from reprocessing facilities and civil reactors, from radioactive waste dumped at sea and from NORM sites. Thus, whenever refractory radionuclides are released following nuclear events, radioactive particles should be expected.

Radioactive particles are defined as a localized aggregation of radioactive atoms that give rise to an inhomogeneous distribution of radionuclides significantly different from that of the matrix background. In water, particles are defined as entities having diameters larger than 0.45 μm, i.e., entities that will settle in still water due to gravity. Following a nuclear event, radionuclides released from a source can be present in different physico-chemical forms ranging from low molecular mass (LMM) species believed to be bioavailable, to particles that can be retained in soils and sediments.

To assess environmental impact and risks associated with ecosystems contaminated with radioactive particles, links must be stablished between the source term and deposition, ecosystem transfer, biological uptake and effects in exposed organisms. The traditional assessments based on average bulk mass or surface activity concentrations of radionuclides in environmental compartments (Bq/kg, Bq/m^2 or Bq/L) should be considered in these cases not valid for the reasons that will detailed in the coming paragraphs, and new approaches should be used to evaluate the potential impact of particle contamination on site-specific transfer factors (Kd, CR/BCF) and dose estimate. The experience and knowledge gained in the running of the RATE project in this matter is summarized in this report.

4 Initial assessments

The ecosystems affected by radioactive contamination in particulate form, especially when this contamination is formed by U/Pu refractory particles, present two main peculiarities that makes the use of conventional magnitudes and parameters for its evaluation impossible:

a) One of the fingerprints of any contamination produced in particulate form is its uneven distribution, associated to their discrete character. This inhomogeneous distribution of the contamination is observed at different scale levels, even at the very short scale used conventionally for the monitoring and sampling for example of soils at one defined point. Different aliquots from the same sampling point can present different activity concentrations depending of the number and magnitude of the radioactive particles in each aliquot. Hence, localized heterogeneities such as particles will be unevenly distributed, making representative sampling difficult.

b) In addition, the refractory character of these particles provokes in most cases only its partial dissolution when conventional procedures of dissolution are applied, as an essential step prior to the measurement of the radioactive contamination. Thus, the inventories can be underestimated, and consequently the ecosystem transfer can be incorrectly described. Furthermore, the magnitude of the partial dissolution is very much dependent on the method applied, causing additional, unacceptably large uncertainties in the inventories.

These two main peculiarities have been observed in all the terrestrial ecosystems affected by particle radioactive contamination where the RATE partners have concentrated most of their efforts along the project: Palomares (Spain), Thule (Greenland) and Maralinga (Australia). As an example we show some results obtained in one of the terrestrial contaminated areas of Palomares, Spain, related to these issues.

a) Several aliquots heavily contaminated with radioactive U/Pu particles were treated conventionally either with aqua regia, or with pure nitric acid plus drops of percloric acid, resulting only in partial dissolution of the Pu and U. Importantly, the percentage dissolved was quite erratic, oscillating between 20% and 80% and with no clear correlation with any controlled peculiarity of the aliquot under study. Total dissolution of the Palomares particles were obtained only after applying an alkaline fusion method.

b) When the upper 5 cm of soil were collected from the same sampling point (surface 1 m^2), and different one gram aliquots were randomly analysed, the results obtained showed very high heterogeneities, seen in Table 1. This heterogeneity, intrinsic to the characteristics of the analysed contamination, even question the expression of the contamination using the magnitude of activity per unit area (Bq/m^2) that is simply calculated from the determined activity concentration in the aliquot under analysis by assuming an uniform distribution of this contamination.

Sampling point	Range in $^{239+240}$Pu activity (Bq/m^2)	Number of aliquots analysed
A2	6500 – 65000	4
A3	2000 – 5500	4
A5	800 – 10000	6
A7	1000 – 7500	4
A10	100 – 400	4
A12	50 – 2200	4
A15	80 – 300	5

Table 1.- Range of activities (expressed in Bq/m^2) determined by analysing several aliquots of different sampling points belonging to one of the terrestrial areas still affected by the Palomares accident

The intrinsic heterogeneity observed also question the assignation of a single value of activity concentration (Bq/kg) to each sampling point, and consequently question the use of parameters such as Kd, TF and CR conventionally used to evaluate transfer between different compartments of the analysed ecosystem, that are based in the precise determination of the previously indicated activity concentration.

If for example, the concentration ratio parameter, CR, conventionally used to predict activity concentration in wildlife assuming equilibrium between the whole organism (wo) under consideration and the appropriate medium (i.e soil in the case of terrestrial ecosystems)

CRwo = (Activity radionuclide X in whole body (Bq/kg FM))/ (Activity radionuclide X in soil (Bq/kg DM))

is calculated, one important problem appears as the denominator in the previous expression should be defined without high uncertainty for a specific site in an ecosystem affected by particle contamination.

The use of the mentioned transfer parameters even in the hypothetic case where the activity concentration in the medium (soil, water) can be defined with precision will in addition be associated with some other problems or inconsistencies. For example, these parameters are defined for equilibrium conditions (are independent of time) and are independent of the magnitude of the contamination affecting to the ecosystem under analysis. Both conditions are not fulfilled in particulate contaminated sites as will be shown later.

In section 6, inconsistencies in the use of conventional transfer parameters associated with wildlife in the ecosystems of Maralinga (rabbits), Palomares (snails and rabbits) and Thule (muskox) will be highlighted and associated challenges will be described in order to consider properly the transformation processes and interactions when particles are involved.

In summary, the mobilization of radionuclides as well as their uptake by wildlife to assess ionizing radiation exposure, has been generally quantified incorrectly until now by using parameters such as distribution coefficents (Kd) and concentration ratios (CR) taken from databases constructed with basis in studies generally done in areas only affected by homogeneous radionuclide distributions mostly in ionic form (i.e. worldwide weapon fallout), without recognizing that:

a) radioactive particles contains a significant fraction of the bulk sample activity, leading to sample heterogeneities problems because these in general are inhomogeneously distributed, and

b) the ecosystem transfer will generally be delayed until weathering and remobilization of associated particle radionuclides occur.

The proper Kd and CR parameters should avoid the mentioned problems and should be defined as rate functions, avoiding the simplistic approach of parameter constancy.

In the analysed ecosystems affected by radioactive particles, the contamination is in general in less bioavailable form than in standard reference sites. This dampens the effectiveness of radionuclide uptake such that the use of standard reference concentration ratios (CRs) would over-predict animal radionuclide body burden. Soils affected by nuclear weapons fallout differ from soils affected by particle deposition in that they present a higher percentage of the radionuclide under analysis in water soluble and readily exchangeable form. They also differ in their rate of change in availability in the soil because the mobility of radionuclides from nuclear weapons testing decrease with time due to fixation by soil components, while in the particle contaminated areas, the availability increase with time due to weathering.

On the other hand, the whole-body activity concentration can experience significant variations depending of the possibility of uptake by the living organism, through inhalation or ingestion, of a significant amount of the radionuclide under analysis associated to a particle. In fact, the radioactive particles represent point sources of potential long-term environmental and ecological significance, due to risk of inhalation, dermal absorption, skin exposure and ingestion of particles. Some animals as filter-

feeders (e.g. molluscs) and soil-dwelling animals (e.g.gastropods) can retain particles in their organism and eventually be ingested by man. All these facts should be considered for proper environmental risk assessment in ecosystems affected by particle contamination.

5 Linking particle characteristics to ecosystem transfer

Nowadays, the release and presence of radioactive particles in the environment has been accepted as a phenomenon by the scientific community, but after its deposition for example onto soils and sediments, information about the ecosystem behaviour of particles is still quite limited. The influence of various characteristics of these particles on their ecosystem transfer and their uptake/retention by biological systems should be analysed and evaluated for a proper environmental impact assessment in areas affected by contamination in particulate form. In the great majority of cases analysed in the frame of the COMET project, the U/Pu particles disseminated in different ecosystems associated to nuclear weapon tests and/or conventional nuclear detonations (Semipalatinsk, Maralinga, Palomares, Thule) have initially a quite inert behaviour due to its refractory character. In the first assessments performed just after release, extremely high distribution kd values and quite low soil to plant transfer factors, TF, and whole-body concentration ratios, CR, were obtained.

The performance of modelling exercises in these areas by adopting only as inputs bulk activity determinations and taking by default the values of the different transfer parameters defined in other releases and environmental conditions, will lead to erroneous conclusions about the transfer of radionuclides along the different compartments of the ecosystem under analysis and to incorrect dose estimations. Transfer factor parameters defined for actinides deposited in ionic or bioavailable form will be completely different from the real ones affecting the ecosystem under analysis due to radioactive particle deposition.

As soil and sediments initially will act as sinks for radioactive particles in our analysed ecosystems, these soils and sediments may also act as a potential diffuse source in the future. Over a long-term time interval, information about particle weathering and remobilization is needed in order to assess long-term impact from radioactive particle contamination, being dependent this behaviour of particles characteristics. In consequence, in the scenarios considered in COMET, such as Palomares, Thule and Maralinga, apparent soil-water distribution coefficients (Kd) will have extremely high values if high activity particles are present in soils, while the apparent kd will change over time due to weathering (Salbu et al., 2017).

Since early 1960s, leaching experiments have been performed to simulate possible transfers between different compartments in ecosystems affected by radioactive particle releases in order to gain information about transformation processes (Crocker et al., 1966; Salbu et al., 2017). However, these leaching experiments were quite scattered and were carried out applying different procedures, making it difficult to obtain useful/comparable information from them. If they are properly established/justified and validated, leaching experiments can provide important data on particle characteristics and the links to solubility, particle weathering rates and remobilization potential for particle associated radionuclides (Lind, 2006). Within RATE and in collaboration with the IAEA-Co-ordinated Research Program (CRP) on Environmental behaviour of radioactive particles, an abiotic protocol has been established with the aim of identifying key parameters affecting properties of particles when exposed to a variety of different water qualities and abiotic leaching agents.

These leaching experiments have been performed on single radioactive particles, but also on bulk samples from where the particle was isolated because it will help in the interpretation of results obtained on particle transformation processes including re-adsorption processes and also link new data with literature data. In the abiotic protocol, isolated and well characterized radioactive particles or small bulk samples known to be contaminated with particles are transferred to centrifuge tubes and extracted with 20 ml of the leaching medium for 2 hours (representative of a residence time in the stomach), 24 hours (transit time through the intestine) and 168 hours (long term potential mobility of radionuclides). The abiotic reagents used in the experiments were rainwater and HCl 0.16 M (in this last case mimicking the acidity and strength of gastric fluids).

Similar experiments have been also performed on small bulk soil samples using cow rumen as in vivo biotic reagent during 24 hours following the work of Cook and co-workers (1995).

More than fifty different leaching experiments have been carried out, generating a large amount of data. In this report, only some examples and general conclusions will be highlighted.

Figure 1 shows the results from the leaching of three different particles generated as local fallout in places where several weapon tests were carried out: two particles have their origin in the area called “Ground Zero” in Semipalatinsk (Kazakhstan) and the third one corresponds to particle material released in the weapon tests done in the 50-60s in Nevada (USA). The leaching agent in the three cases was 0.16 M HCl.

The main and important conclusion obtained from these leaching experiments, performed with single isolated particles, is that even with the relatively strong reactive agent used, the percentage of Pu dissolved or liberated from the particles is extremely low, not exceeding 2% in any case. The glassy/inert composition of these particles, with the actinide contamination present as embedded concentrated small inclusions, precludes solubilization. These particles from an environmental point of view have a quite inert behaviour, not experiencing appreciable transformation processes since their release and deposition due to weathering.

Similar experiments have also been performed with isolated particles from contaminated soils collected in Thule (Greenland) and Palomares (Spain), but in this case using rainwater. The cumulative solubilized Pu from these particles after one week of leaching did not reach 1% of the Pu associated to the particles in any of the experiments. This confirmed the reactive behaviour of the contamination released in the two conventional nuclear detonation accidents involved (Jimenez-Ramos et al.2008), and that since the initial deposition of the contamination in the sixties, no big changes have been produced inducing increments in transformation processes.

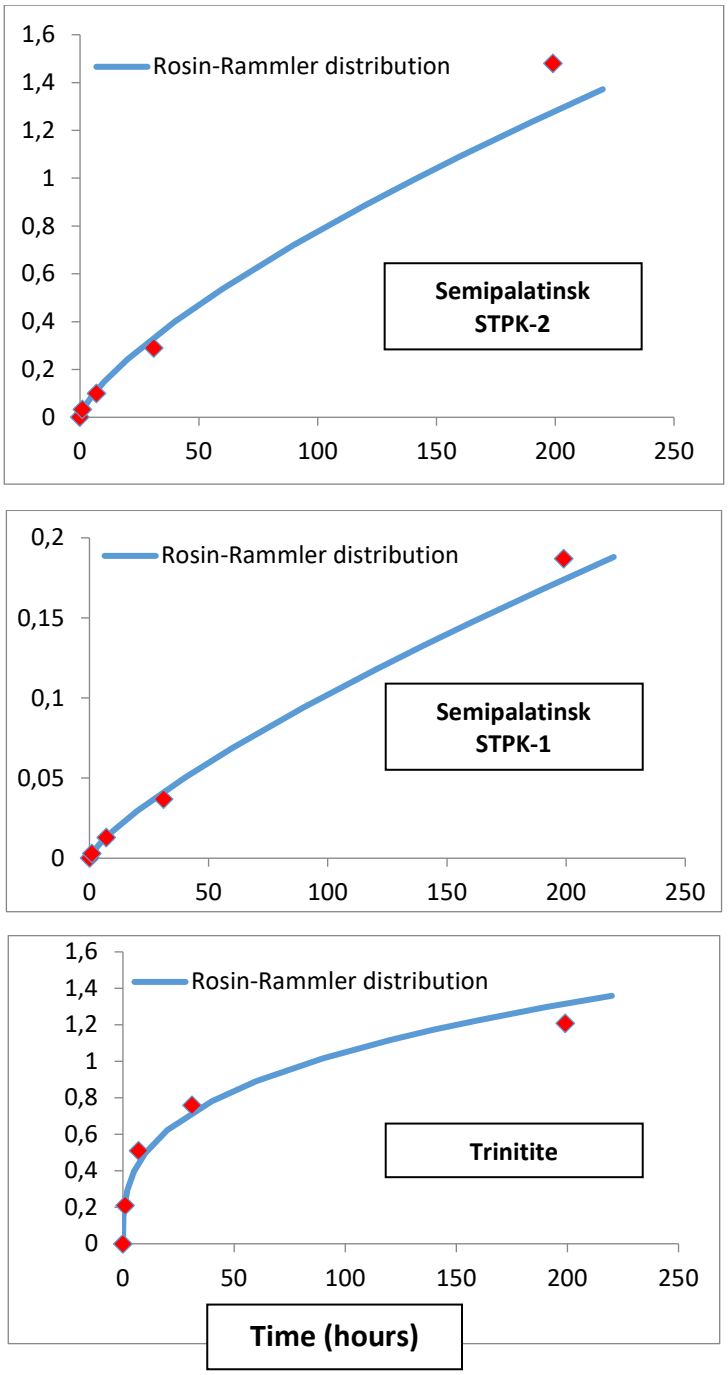


Figure 1.- % of cumulative Pu extracted from Semipalatinsk and Trinity radioactive particles with 0.16 M extractant solutions

As it is shown in Figure 2, when Thule isolated particles were leached with natural water of varying conductivity, roughly equal proportions of Pu and U lost from individual particles was observed, giving evidence of solid mass transfer from particle to solution rather than chemical transformation from solid to ionic state.

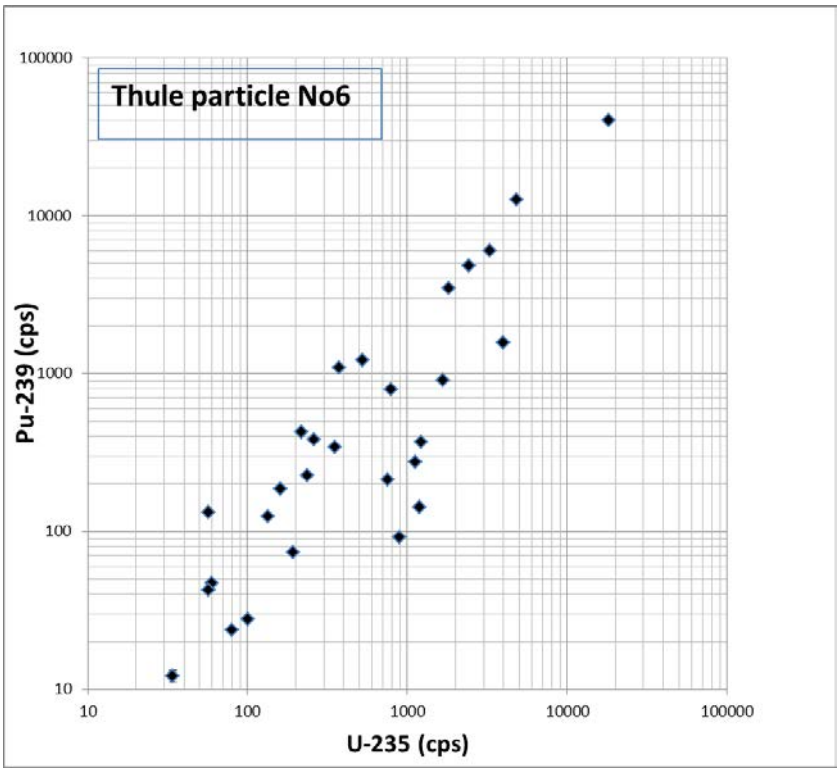


Figure 2.- Amounts of Pu-239 vs U-235 liberated in washings/leaching experiments performed using natural water as reagent on a Thule radioactive particle

The results obtained in the leaching of Palomares and Thule contaminated soil samples ratify the conclusions obtained from the analysis of the isolated radioactive particles. Negligible amounts of Pu were dissolved in these leaching experiments, being more related the small percentage dissolved with dissolution of material that with the conversion of the Pu to ionic form.

In addition, in these soil bulk samples, competitive effects trying to capture the dissolved Pu should not be neglected. The typical cumulative desorption curves observed in the leaching over time of isolated particles (as shown in Figure 1) will follow the same structure but with lower percentages of dissolution for their associated soils due competitive re-adsorption effects.

On the other hand, contaminated soil samples collected in Palomares and Thule have also been subject to in vivo biotic experiments with rumen cow at 39ºC. Here, after 24h of leaching the amount of Pu found associated to the leachates were quite small in proportion to the present in the bulk soil. This fact confirms the refractory behaviour of the Palomares contamination even 50 years after the accident and after the consequent dispersion of contamination. This is contrary to what was observed for the leaching of soils affected by weapon fallout, where the contamination is in a more bioavailable form, and high percentages of dissolution are obtained in biotic experiments with cow rumen (Espinosa et al., 2005).

In conclusion, through the abiotic and biotic leaching experiments carried out in the RATE project on isolated particles and corresponding bulk soils from places affected by local fallout of nuclear weapon tests (Maralinga, Semipalatinsk) and for conventional nuclear detonations (Thule and Palomares), it has been observed in all cases very high kd and very low CR values, even more than 50 years after the dissemination of the radioactive contamination in particulate form in the affected ecosystems.

This on-site specific information is essential for a proper radiological and environmental assessment in these areas, because it differs considerably in behaviour to that taken as default in most of the models developed for this purpose.

6 Biological uptake in ecosystems affected by radioactive particle contamination

In addition to previously described leaching experiments, during the RATE project:

a) the phenomenon of retention or uptake of particle associated radionuclides in various biota from contaminated areas have been studied, centering the efforts in terrestrial areas affected by Pu/U releases to the environment in particulate form with origin in accidental or provoked conventional detonations of nuclear weapons: Maralinga (Australia), Palomares (Spain) and Thule (Greenland). Additionally, the retention capacities of radioactive particle by mussels have been analysed and evaluated through the performance of laboratory experiments.

b) an evaluation of the factors affecting the magnitude and uncertainty in the prediction of Pu/U uptake by selected biota in some areas contaminated by Pu/U particles have been performed.

6.1 Plutonium uptake by wildlife at the Maralinga legacy site, Australia

Maralinga Test site (MTS) is located on the southern edge of the Great Victoria Dessert, South Australia, In this area, semi-arid conditions exist with sparse and erratic rainfall. MTS was used by UK for the detonation of seven nuclear devices, each conducted at a separate firing site. In particular, at the Taranaki firing site, 22 kg of Pu was dispersed in 12 non-fission test carried out in 1961-63. This dispersion was not produced by nuclear explosion, but involved burning and high explosive detonations.

One of the RATE partners, ANSTO, has created a database of soil-to animal transfers (as measured by CRs) for mammals, reptiles and arthropods in Taranaki (Johanssen et al., 2014; Johanssen et al. 2016a). This database was created 50 years after conventional detonations were carried out, with basis in the following sampling and field work strategy:

First of all, composite soil samples were gathered in different 200 m diameter plots of the affected terrestrial ecosystem, each plot with a different level of contamination. In each plot, a random sampling approach was applied, being observed high variation among samples within the same plot, a fact that is consistent with a patchy, inhomogeneous deposition of particles. This emphasizes the need at particle contaminated sites for sufficient soil sampling to represent the forage area of the biota. The practice of taking a single soil sample to represent soil-to-biota uptake would potentially lead to three orders of magnitude uncertainty at these sites.

The CRs at Taranaki forming part of the ANSTO database were calculated as the activity concentration in the organism related to that of the composite soil samples (n=5) of the forage-range. The use of multiple soil samples enables the distribution of the CR values over the forage range to be estimated and expressed in terms of geometric means.

For mammals, the CR values were calculated following standard approaches: the whole-organism was calculated from measurements on a range of tissues where Pu had accumulated via circulation system (muscles, bones, liver, kidneys, spleen, heart, lungs and blood). The GI tract was excluded for CR determination. For reptiles in the CR determinations the GI tract was excluded, but the skin was considered. For arthropods, calculations include the entire organism.

The obtained results indicate the uptake of plutonium in mammals is persistent over time. The Pu associated with respirable soil fraction has remained over constant between the 1980s until nowadays and similarly the soil-to-mammal transfer has remained about constant from 1975 to the present. These data are consistent with delayed, or gradual, weathering of particles over time and suggest that the reservoir of particles in the soil at this site will endure for many years into the future as an ongoing source term to the biosphere.

The Pu transfer rate in the whole area is lower than in areas affected only by weapon fallout. These lower values are likely due to the presence of a low-soluble, particulate form of the Pu in Maralinga soils. In addition, the CR were not consistent among rabbits captured from various plots, but rather indicated a distinct spatial pattern which appears to be related to the physical form of the Pu contamination. In the plot with high Pu concentration in soil, high values were found within rabbits, but the CRs were the lowest. At plots with lower activity concentrations in soil the CR values increased approximately linearly by three orders of magnitude. The most likely explanation is the increased density of larger non-respirable low-solubility particles at areas with high levels of Pu contamination, i.e. closer to the test detonation locations. Such particles contribute to the soil concentration in the denominator of the CR, but make a limited contribution to the numerator as they are not readily respired or absorbed. This effect dampens the effectiveness of Pu uptake such that the use of standard reference CR would over-predict animal Pu body burdens where such particles are present.

The described ANSTO database also includes information about internal Pu distribution in rabbits inhabiting Taranaki weapons test site, Maralinga, Australia. From this database, the following Pu distribution was observed in tissues: skeleton 83%, muscles 10%, liver 6%, kidneys 0.6% and blood 0.2%. This distribution aligns with previous studies related to environmental exposure (worldwide fallout), but contrasts with the partitioning models that have traditionally been used for human worker protection purposes. These models are based in laboratory experiments with Pu acute exposures carried out via injection and with short evaluation periods.

The Pu activity concentrations in the lung tissues were elevated relative to the body average, indicating the presence of non-absorbed Pu mostly incorporated by inhalation of Pu-soil respirable fraction. On the other hand, the amount of Pu in the gastrointestinal (GI) tract was also highly elevated relative to that absorbed within the body, potentially increasing the transfer of Pu to wildlife (predators) and human consumers that may ingest gastrointestinal tract organs.

The Pu results in the GI tract are consistent with the low absorption rates for Pu in mammals published by the International Commission on Radiological Protection (ICRP, 2012), which indicate that most ingested Pu is not absorbed across gut mucosal cell, but rather remains in the non-absorbed material becoming progressively more concentrated as it moves through the intestines. In addition, rabbits

occasionally feed through coprophagic reingestion of soft faeces, which also contributes to concentration build-up as the material is recycled through the digestive system.

We can conclude that in the Taranaki weapon test site, the uptake of Pu by wildlife appears to be regulated and highly influenced by Pu form, as some Pu-containing particles are of not readily respirable, absorbable size. This peculiarity is also observed in the internal distribution of Pu in rabbits with higher Pu levels than expected in lungs and in the gastrointestinal tract due the presence of persistent non-absorbed Pu in these organs.

6.2 Plutonium uptake by wildlife at Palomares, Spain

Through a joint collaboration between the University of Seville, CIEMAT and NMBU groups, the degree of contamination and possible uptake of radioactive particles by snails, which is one of the main living species which can be found in the semi-desertic environment affected by the Palomares accident, has been evaluated and analysed.

A sampling campaign was performed in the spring 2015, where a good set of representative snails were collected in the affected area. After collection, all the snails were cleaned individually with deionised water in order to remove possible external contamination due to the adherence of some soil grains to their shells. Afterwards, they were fed during some days in the laboratory with inert material in order to collect their faeces and provoke the removal of all the material present in its digestive system at the moment of collection. After the cleaning of their digestive system, the snails were sacrificed in order to separate the body mass from the shell. The whole-body tissue of each separated individual was introduced in a small vial and the whole-body ^{241}Am content in each snail was determined by high-resolution gamma-ray spectrometry using an ultralow-background system equipped with a well germanium detector and an anti-Compton guard device. The ^{241}Am activities for the different snails analysed, are compiled in Table 2 (Lind et al., 2016).

Identification Code	Whole-body ^{241}Am, mBq	Identification Code	Whole-body ^{241}Am, mBq
Snail 2	6 ±1	Snail 13	18 ±1
Snail 3	7 ±1	Snail 14	132 ± 3
Snail 4	15 ±1	Snail 15	80 ± 2
Snail 5	14 ±2	Snail 16	2 ± 1
Snail 6	46 ± 3	Snail 17	68 ± 2
Snail 7	174 ±3	Snail 18	63 ± 2
Snail 8	89 ± 3	Snail 19	48 ± 2
Snail 10	10 ± 1	Snail 20	87 ± 3
Snail 11	2 ± 1	Snail 21	19 ± 2
Snail 12	2 ± 1	Snail 22	32 ± 2

Table 2.- ^{241}Am activity content in tissues of snails collected in the Palomares contaminated area.

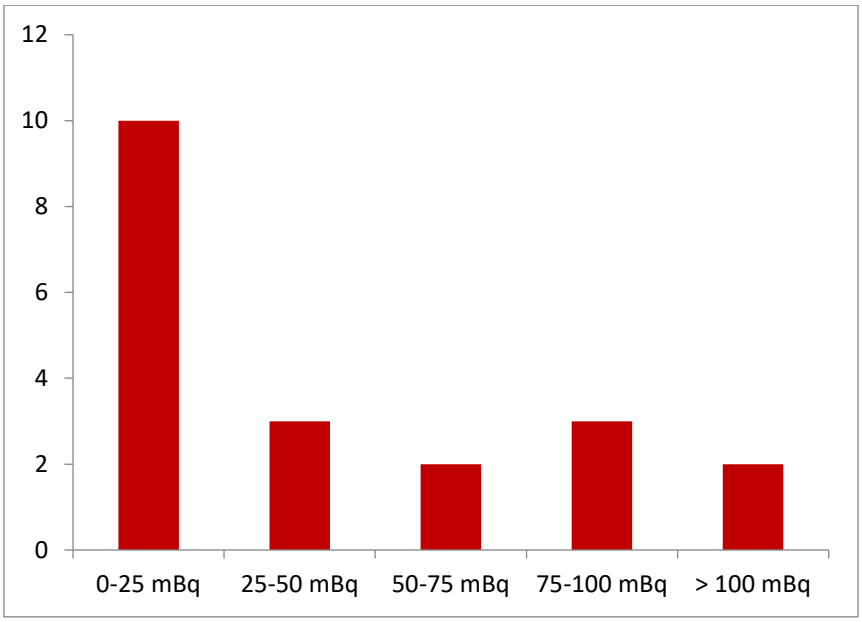


Figure 3.- Histogram showing the distribution of the snails analysed as a function of its ^{241}Am whole-body activity

All the snails collected show the signal of the accident, but in a quite variable level of contamination. Although all the snails were collected from the same contaminated area (collection surface was not higher than 200 m x 200 m), differences in the levels of ^{241}Am of up to two orders of magnitude (from two to hundreds mBq) have been observed. This high variability in the uptake by animals captured in a defined terrestrial area is generally not found when the radionuclide contamination is homogeneously distributed, with the bioavailable fraction in the soils being relatively uniform, and can be associated with the presence of most of the radionuclide contamination in the soils of zone 2 in particulate form, inhomogeneously distributed. The ^{241}Am in superficial soils collected randomly in the sub-zone where the snails were collected can differ up to two orders of magnitude.

The variability observed in the whole-body ^{241}Am levels of the snails, and the variability existing for the same radionuclide in the associated soils, make questionable the utility of use and determination of the ^{241}Am concentration ratio (CR) parameter for these gastropods in the area under analysis due to its large associated uncertainty. As the ^{241}Am-CR parameter is defined as the ratio between the ^{241}Am activity concentration in the snail (in fresh weight) and the ^{241}Am activity concentration in soil (in dry weight), its value in zone 2 of Palomares will have an associated uncertainty of several orders of magnitude. This is in contrast to the narrow range of ^{241}Am-CR values compiled in the IAEA database for gastropods inhabiting in areas only affected by an homogeneous weapon test fallout contamination (IAEA, 2014). The use of CR values defined from experimental areas affected by weapon test fallout on the other hand, will over-predict the uptake of ^{241}Am in the majority of the snails analysed in our case due to the low bioavailability of the radionuclide contamination in Palomares (in the present experimental study, the majority of snails contained less than 10 mBq of ^{241}Am). At the same time, it will not take into consideration that a limited but random number of snails can have much higher ^{241}Am values due to the ingestion/retention of discrete radioactive particles.

An additional experiment with some of the snails collected in Palomares was performed after the determination of their ^{241}Am level of contamination. The snails 14, 15 and 18 (see table 2) were subjected to dissection by a specialist in order to search for the internal distribution of the ^{241}Am in the different parts of their bodies.

Each snail body was divided in four different parts.

a) **ADI:** Initial part of the digestive apparatus (mouth, esophagus and stomach).

b) **ADF:** Final part of the digestive apparatus (intestines, anus).

c) **HP:** Hepatopancreas; digestive gland, that in gastropods provides the functions which in mammals are provided separately by the liver and the pancreas.

d) **R:** Rest of the animal, formed mostly by muscles

and the obtained results are compiled in Table 3

	^{241}Am, mBq	% of whole-body activity
Snail 14		
ADI	**14 ± 1**	**10**
ADF	**68 ± 4**	**47**
HP	**41 ± 2**	**28**
R	**21 ± 4**	**15**
Snail 15		
ADI	**8 ± 1**	**14**
ADF	**8 ± 1**	**14**
HP	**36 ± 3**	**64**
R	**4 ± 1**	**7**
Snail 18		
ADI	**12 ± 1**	**17**
ADF	**18 ± 2**	**27**
HP	**36 ± 4**	**52**
R	**3 ± 1**	**5**

Table 3.- ^{241}Am internal distribution in snail collected in Palomares

The majority of the ^{241}Am in the analysed snails was found in their gastrointestinal system (GI), even knowing that the snails were sacrificed after having assured that all the remains from material fed in Palomares ecosystem were excreted. In the three cases analysed, 85-95% of the ^{241}Am was found in the GI, with a minimal proportion in the tissues. These results indicate that the plutonium (americium)

incorporated by ingestion by the snails is in a not bioavailable form, being not absorbed and, consequently, not distributed in the whole body.

In conclusion, the experimental work performed in Palomares reinforces all the conclusions obtained in Maralinaga. In fact, the Palomares results indicate that the the conventional concept of whole-body concentration factor should be avoided, and that generic evaluations about transfer factors should be substitued by the specific and individual studies. The low solubility of the Pu/U particles disseminated in the area preclude finding significant levels of contamination in wildlife although the possible retention of particles could change totally the dosimetric implications as it will be demonstrated below.

6.3 Retention of particles by wildlife in contaminated sites

In the two sites previously assessed, Palomares and Maralinga, the possibility of retention of particles inside the whole body was also evaluated due mainly to the associated dosimetric implications that will be evaluated in the final section of this report.

The ^{241}Am levels found in the analysed Palomares snails, that were shown in Table 2, can be evaluated as moderate with little evidence existing about the possibility of the presence of some micrometer highly radioactive particles retained in their digestive system. Only the snail 7 showed ^{241}Am levels indicative of the possible presence of a radioactive particle in its body, although all the efforts devoted by the RATE partners in this snail for radioactive particle identification gave no positive results. Consequently, if due to their feed habits, some hot-particles are ingested by these animals, it seems that in most cases these particles should flow through their digestive system and then would be excreted in a high proportion.

In order to obtain some evidences supporting this last hypothesis, and to gain additional information about the interaction of these animals with hot-particles disseminated in its living area, two different approaches were followed.

In the first one, the faeces collected before sacrificing the snails was analysed for the determination of its ^{241}Am content by gamma-ray spectrometry. The results reflect the fact that due to their feeding habits, the snails are ingesting Pu contaminated soils by the Palomares accident, but with the particularity that most of the contamination should flow along their digestive system, being excreted. This conclusion is in agreement with previous results and conclusions obtained by Ciemat in the area (Aragón et al., 2006).

The second approach followed to confirm that most of the ^{241}Am ingested by the snails is finally excreted, was the performance of a laboratory experiment, where initially non-^{241}Am contaminated individual snails were, under controlled conditions, fed with a mixture of Palomares soil contaminated with particles and uncontaminated fodder, and the proportion of the ingested ^{241}Am found in the faeces determined.

A detailed description of the laboratory experiment is given below:

a) Two individuals of the specie *Otala punctata* were collected for the controlled feeding experiment. One of them (snail) was fed a mixture of Palomares contaminated soil and fodder, the other (snail) only fodder.

b) The two snails were introduced in independent containers with ventilation and periodic humidification with pulverized water. The experiment was done at controlled temperature of 20-21ºC, and with a photoperiod of 12 hours of light per day.

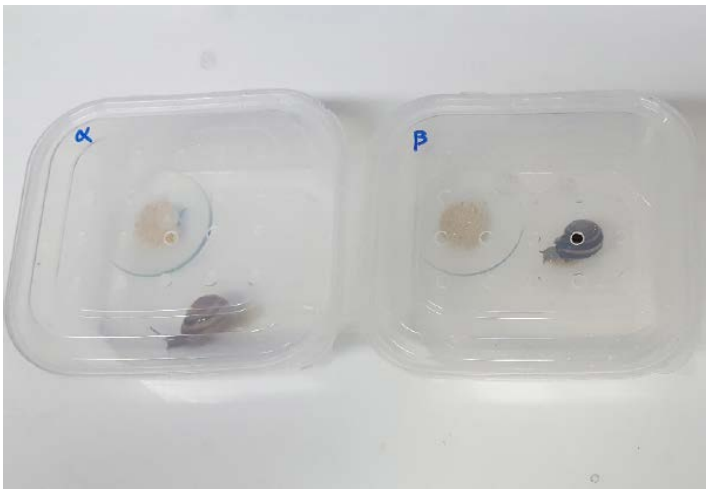


Figure 4.- Experimental set-up used in the experiment of controlled snail feeding with Palomares contaminated soil.

c) The experiment lasted for 14 days. During the first two days of the experiment, both snails (and) were fed only with fodder. After that, the snail was fed with a mixture of Palomares contaminated soil plus fodder for 10 days, while the snail continued to be fed only fodder. In the last two days both snails were, as at the beginning of the experiment, fed only with fodder in order to assure that snail had excreted all the Palomares soil ingested.

d) After the 14 days experiment, both snails were sacrificed following the methodology described in previous pages of this report (immersion in water) and the separated soft tissues were independently measured by gamma-ray spectrometry looking for ^{241}Am determination. Obviously, the ^{241}Am content in the Palomares soil ingested by snail was determined at the beginning of the experiment.

e) During the 14 days of the experiment the faeces generated were collected, forming at the end and for each snail a composite sample also measured by gamma-ray spectrometry.

The results of the laboratory experiment were quite conclusive. In the case of snail , used as control sample, no ^{241}Am was detected in the fodder provided to the snail, in the whole-soft body after being sacrificed and in the faeces collected along the 14 days. In the case of the snail , the following results were obtained.

- ^{241}Am activity in the Palomares soil fed by snail during the experiment: **213 mBq**
- ^{241}Am activity in the faeces of snail , collected along the experiment: **177 mBq**
- ^{241}Am activity detected in the body of snail after being sacrificed: **not detectable**

Taking into consideration that the measurements performed in the soil and the snail can be affected by an uncertainty of around 10%, it is clear to observe, a) the coherence of the results obtained from an activity balance point of view, and b) the association of the great majority of the radioactive contamination ingested by the snail with the faeces excreted is in total agreement with the conclusions obtained previously by a different approach.

Note that the snails used for the laboratory experiment were collected in a place located several hundred kilometers far from Palomares, and for that reason did not have any significant ^{241}Am background in their bodies. Hence, it is not surprising that after only a single feeding event of 14 days duration with contaminated Palomares soil, the body of snail does not show detectable levels of ^{241}Am. In the snails collected in Palomares, even in the less contaminated ones, the ^{241}Am was detectable and measurable, but these snails were exposed to the contamination along all their life, demonstrating its cumulative uptake.

Studies analysing the possible uptake of radioactive particles by living species in the area affected by the Thule accident (Eriksson et al., 2008) were also performed in the RATE project, by the DTU partnership. This partner concentrated its efforts on the evaluation of the possible uptake of contaminated particles by muskoxs (*Ovibos moschatus*), a large Arctic mammal of the Bovidae family that inhabit this contaminated ecosystem. The muskox first evolved in temperate regions of Asia and adapted to a cold tundra environment late in its evolutionary history. The clear interaction of these mammals with contaminated particles have been demonstrated by looking for detectable actinide levels in faeces generated by these muskox specimens and in searching for the presence of heterogeneities and hot-particles in them.

The levels of ^{241}Am were determined by gamma-ray spectrometry in several dozens of muskox faeces samples, the results of which are shown in Figure 5 (levels of ^{210}Pb determined in the same set of samples are also shown for comparison purposes).

All the analysed muskox faces samples included in this study show levels of ^{241}Am over the limit of detection, but in most cases the levels are quite moderate, under 100 Bq/Kg. In a quite simple trophic chain as the existing one in the artic environment, there is some transfer of actinide to the Muskox specimens. In a limited number of the aliquots analysed, the levels of ^{241}Am found were clearly higher than the average. In a couple of them the ^{241}Am activity concentrations wree clearly higher than 1000 Bq/Kg, indicating the presence of large radioactive particles.

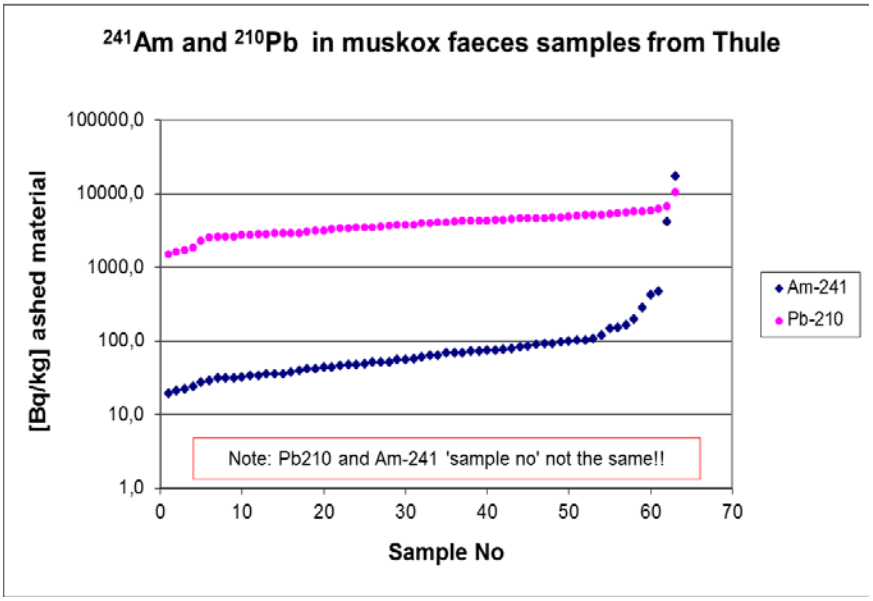


Figure 5.- ^{241}Am and ^{210}Pb activity concentrations (Bq/kg) in muskox faeces samples from Thule

All the analysed muskox faces samples included in this study indicated the interaction of the particulate contamination even with the higher levels of the trophic chain, but in spite of the obvious continuous oral intake, the analysis of body parts (liver, kidney, bones, lungs, spleen and meat) from singles muskoxen showed no detectable ^{241}Am with detection limits below 2 Bq/kg (Roos, private communication). These results are not suprising, and even go in the same direction that the conclusions obtained in the study with wildlife in Maralinga and Palomares. Radioactive particles are obviously ingested by mukoxens in Thule, but they exhibit a quite low solubility, with no detectable amounts of actinides being absorbed and with the particles being excreted with faeces. This conclusion is also in agreement with the results obtained in the performance of some biotic experiments along the RATE project, where Thule soil aliquots heavily contaminated were submitted to leaching with rumen cow. The percentage of leached Pu in these experiments was very low (Lind et al, private communication), reinforcing the comment of the insoluble behaviour of Thule radioactive particles along the gastrointenstinal tract of the studied animals. The particles ingested by the muskoxes are then in practice excreted in their totallity without experiencing any significant losses in their radioactive content due to their low solubility. However, research under development is trying to evaluate if larger radioactive particles could be retained in the intestines, acting as a localized point source.

The Thule muskox results are in agreement with the those previously published, analysing the impact in animals of to the Chernobyl accident. During the Chernobyl accident, agricultural animals were exposed to fuel particles in pastures through forage, sod and swallowed soil. The bahaviour of Chernobyl particles in the gastro-intestinal tract of cows after a single peroral intake showed that the bioavailability of cesium and strontium isotopes associated with particles was two orders of magnitude lower than species in water-soluble form (Yoschenko et al., 2009). The absorption coefficient of ^{137}Cs from fuel particles in cattle (0.8 – 1.4%) was considerably lower than from ionic species (50 – 75%).

The possible retention/accumulation of radioactive particles is not exclusive to terrestrial wildlife. This retention/uptake phenomenon can potentially affect filter-feeders in some contaminated aquatic ecosystems. However in spite of this obvious potential impact, as far as we know, and worldwide, these type of studies have only been performed by one RATE partner, NMBU. The work developed until now has been mostly laboratory work, although some results about the presence of radioactive heterogeneities in mussels collected on the deck of one submarine dumped in Artic waters has also been obtained.

The Russian nuclear submarine K-27 was launched in April 1962. Powered by two liquid metal (Pb-Bi) cooled reactors, it remained operational until 1968, when it suffered a reactor event damaging approximately 20% of the portside reactor's fuel. After attempts to repair the submarine, a decision was made to decommission it and the submarine was ultimately scuttled at 30 m in September 1981 in the shallow waters of Stepovogo Fjord, Novaya Zemlya. More than three decades later, the K-27 has become a topic of discussion as part of plans for remediation of the Arctic Seas, and some monitoring surveillance programs are periodically performed in the surroundings to control the absence of radioactive leakages. In one monitoring program carried out in 2012, some mussels were collected on the deck and in the vicinity of the dumped submarine (Gwynn et al., 2016). The soft tissues of the mussels were recently screened looking for possible radioactive heterogeneities by obtaining digital autoradiograms (Figure 6).

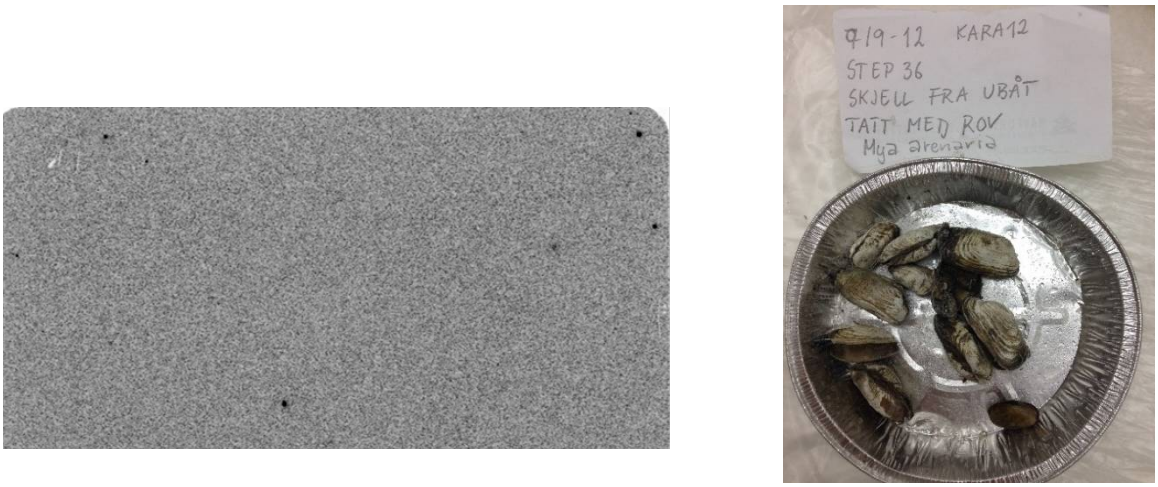


Figure 6.- Digital autoradiogram of mussel tissues collected on the deck of the dumped nuclear submarine K-27.

In the autoradiogram, signals indicating the presence of a few radioactive particles were observed. The origin and composition of these particles are not known, although analysis by ultra-low background gamma-ray spectrometry indicates that all common fission and activation gamma-emitters were below the limit of detection. Further analyses are needed, although a first conclusion confirming the possible retention of radioactive particles by filter-feeders can be highlighted.

In order to investigate such possible retention in filter-feeders, a detailed laboratory experiment was planned and executed in order to evaluate: a) if radioactive particles of variable size can be retained

in a selected filter-feeder species (blue mussels; *Mytilus edulis*) under laboratory conditions, and b) if retained radioactive particles may induce adverse effects if the contact dose is sufficiently high.

In these laboratory experiments, highly radioactive (~3.2 Gy/h contact dose), well characterized spent fuel particles released from the nuclear reprocessing plant of Dounreay, were introduced into mussels in suspension with plankton food or through implantation in the extrapallial cavity (Jaesckhe et al., 2015).

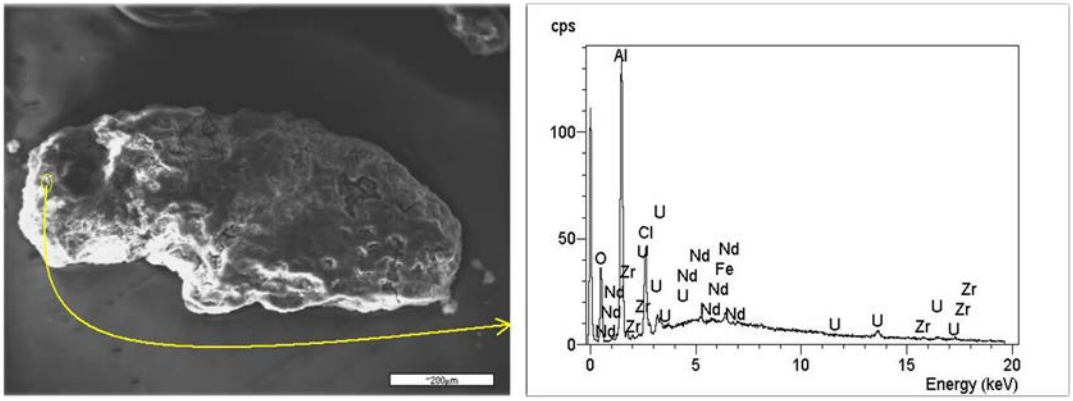


Figure 7.- Scanning electron microscope image and an elemental spot analysis of a Dounreay Materials Test Reactor (DMTR) particle (Jaesckhe et al., 2015).

For the introduction of radioactive Dounrey particles to the mussel via pipette with 0.5 ml of plankton food, the mussel valves were opened, in air, using a small tool, and the particle, with the plankton food, placed inside the mussel. Once inside the mussel it was not known whether the particle would move, how quickly or to where. In this way, particles were introduced to the mussels with food a total of 16 times. In a total of 10 times, particles were expelled (taken precautions to avoid losses or simple ejections due to gravity) while particles were retained 6 times until the end of the experiment (after 70h).

Of the 6 particles retained, a total of 5 were found "loose" within the mussel, i.e found in the siphon and gills, with the notable exception of one particle that was ingested and found in the stomach. It was not determined if the 5 particles found "loose" within the mussel have been caught by gills and bound within mucus, but it appears likely if the experiment has continued such particles would be collected by the mussel and ejected without a short timeframe.

No correlation with size was found for the 10 expelled radioactive particles. The rejection of these radioactive particles could probably be more related to the absence of organic content in them and with their high density values.

Due to the high rate of loss of particles introduced via fed from the mussel, often within 24 h from introduction, a second series of experiments were performed where the particle was implanted into the mussel tissues.

Introduction in the extrapallial cavity is far less likely to occur than introduction via food, but not impossible, as evidenced by the presence of pearls. Pearl formation could occur if a filtered particle

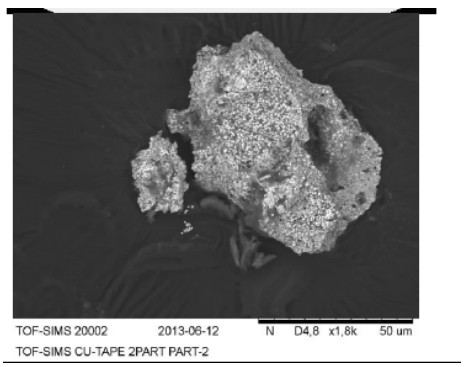
were to become embedded into the mantle or trapped in the extrapallial cavity. In such events the inclusion of a radioactive particle into a pearl or into the shell would make the incorporation permanent with all the consequent implications in dose assessments.

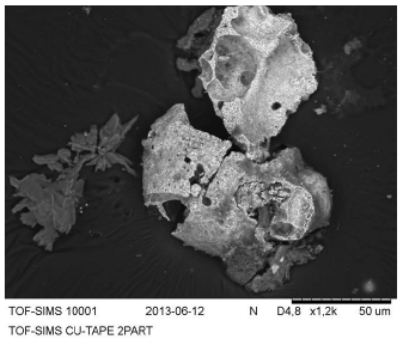
We conclude that the mussels actively collected the introduced particles and ejected them from the body, as mussels are capable of selecting filtered particles, rejecting and expelling undesirable or unpalatable particles from the body (Jaesckhe et al., 2015).

The rate of retention of radioactive particles that may be encountered in blue mussels when filtering seawater is low. Even within the relatively short time of a few days, an introduced particle may rapidly be lost or ejected from the body. Present observations suggest that mussels are capable of actively detecting and removing such particles, although further research is needed in order to obtain a proper frequency distribution pattern of particle uptake.

The implantation of particles generates localized doses, a situation different that that seen when radionuclides are homogeneously distributed. Consequently, current methods used for risk assessments are inadequate for radioactive particles exposures. This point will be particularly analysed in the following section of this report. In particular, the largest and most radioactive particle introduced in the previous experiment induced a significant increase in COMET tail-DNA % and caused a large white mark in the mantle tissue. The white marks found in the tissue were mainly attributed to ionizing radiation effects, and also to physical irritation.

Uptake and retention of particles by wildlife is not constrained to ingestion/peroral intake. Small size-respirable particles can enter the body by inhalation, being in most cases heavily attached to the internal surface of lungs and acting as concentrated point sources, with all the consequent implications in dosimetric studies. These point sources, in most cases are acting continuously over time due to their general refractory/insoluble behaviour. This uptake route in some of the ecosystems contaminated by radioactive particles will gain steadily weight due to the friable and fragile character of the particle contamination that is provoking the break down of the particles since their release /deposition in the affected ecosystems. This is the case for example observed in Thule where the friable behaviour of the radioactive particles is evident (see Figure 8) and only a slight mechanical pressure over a particle causes a break down into small sized particles (Roos, private communication). Small sized respirable radioactive particles have also persisted in Maralinga for decades.





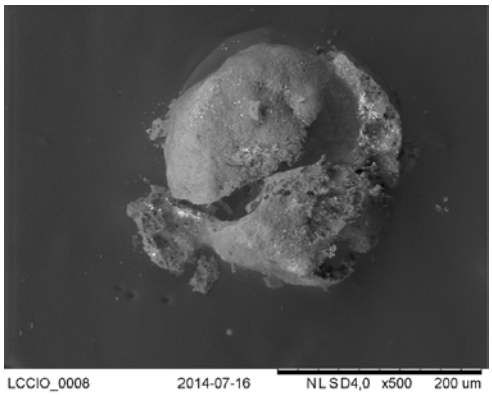


Figure 8.- TOF-SIMS images of different Thule particles indicating their fragility

7 Dose estimate on ecosystem affected by radioactive particle contamination

With all the information shown in the previous sections, it is clear that dose estimations for wildlife in ecosystems affected by radioactive particle contamination is not a straightforward issue.

If the uptake of the particulate contamination is via ingestion, site-specific information, as well as information about the organ distribution of this contamination, will be mandatory for proper evaluation of the magnitude of the solubilized contamination incorporated through this route. The adoption of concentration ratios compiled in established databases generally obtained in different scenarios and under different contamination characteristics, and the application of models calibrated under different conditions concerning the source and physico-chemical form of the released contamination, lead to wrong conclusions and, in most cases, to a clear overestimation of the dose due to the generally refractory behaviour of the contamination released in particulate form.

If the radioactive particle is inhaled and becomes lodged in the lung, it will cause a non-uniform distribution of dose, as has been demonstrated in several occasions with particle track images indicating potentially high dose rates to the immediately surrounding tissues. In addition, there is substantial uncertainty in the effects of such localized dose, although the literature suggests that non-uniform exposure from an inhaled radioactive particle is likely less carcinogenic than that from a uniform exposure for the same average dose.

Conventional methods for calculating radiation dose are then unsuitable when the inhaled activity is in the form of radioactive particles. Until quite recently, the calculation of dose rates from particulate radionuclides assumed that particles were broken down or dissolved with activity distributed homogeneously throughout each organism once they are inhaled. These doses are likely not representative of the actual doses received from a particulate source, as the energy deposited will not be uniformly distributed over the entire mass of the organ, but rather highly localized around the particle. As the dose is confined to a small tissue volume, averaging the dose over the entire organ or tissue volume seems erroneous.

For an appropriate dose estimation, ANSTO has developed a model for localized dose rate calculations, using MCNP6 to obtain energy deposition in the lungs for a particle of each size in each localization of the lung (Caffrey et al., 2016). In particular, they have examined dose rates from single particles ranging in size from 0.01 to 150 □m (alpha) and 1-150 □m (beta/gamma) placed within three averaging volume

alternatives: the volume of a rabbit lung (64 cm^3) and 1.0 and 10.0 cm^3 spherical volumes within the lung.

Dose rates from betta-emitting particles (represented by ^{90}Sr) were found one order of magnitude higher than those from gamma-emitting radionuclides (represented by ^{137}Cs), the self-shielding effect within the particle being negligible for gammas and minor with betas, with independence of the dimension of the particle.

In the case of alpha-emitting particles (represented by $^{239+240}Pu$) self-shielding in larger particles is substantial, with >90 % of emissions captured within particles of +20 □m diameter, but for smaller sizes of the respirable range of 0.01 to 5 □m, an average of 85 % of the energy escapes the particle and is deposited in the surrounding tissues. This is particularly important as it is particles <5 □m in diameter that penetrate deep into the lung and can remain there for extended periods, imparting energy and contributing to the long-term dose rates. These particles may represent a significant contributor to the internal doses of wildlife.

Model calculations assuming a uniform distribution of a pure alpha emitter as ^{239}Pu inside a spherical particle in the form of plutonium oxide (density 11 g/cm^3) as a function of its size are compiled in Figure 6. This figure indicates the percentage of alpha emission energy that is deposited within a Pu-containing particle vs that deposited in surrounding tissue and allows us to conclude that is when the particles are of respirable sizes or when the Pu exist on exterior surfaces, that a radioactive particle that has been internalized may produce relatively intense dose rates to adjacent tissues, as well as providing an ongoing source available for absorption into the body.

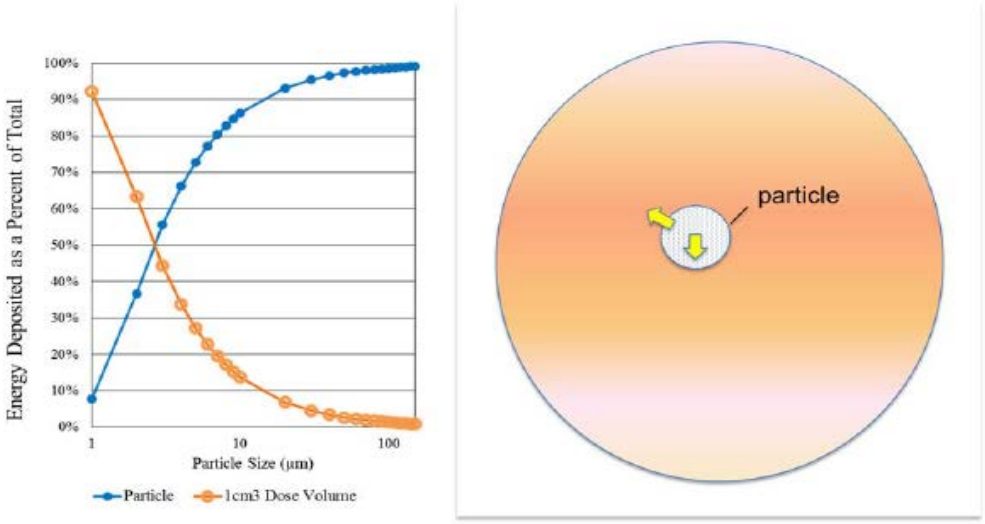


***Figure 9**. MCNP6 model data indicating percentage of alpha emission energy that is deposited within a Pu-containing particle (blue symbols), vs that deposited in surrounding tissue (orange symbols)(adapted from Caffrey et al., 2016)*

The effect of the radionuclide being concentrated in a single particle versus the equivalent amount that is homogeneously distributed as is assumed for organisms in the ERICA-tool, has been also analysed by ANSTO comparing particle versus homogeneous sources for an example organ (rabbit lung) using the MCNP6 computer code (Caffrey et al., 2017). The results indicate decreased dose rates

for particles, with small decreases for particles containing gamma emitters and substantial decreases for particles containing beta or alpha particles. (Caffery et al., 2016; Johanssen et al., 2016b).

The results obtained in the previous study on localized dose rates determinations due to punctual source emphasises importance of the characterization studies performed on radioactive particles, a challenge along the RATE project (Lind 2006, Salbu et al., 2017). The knowledge of the proportion of radioactive emitters existing in each particle and their internal distribution can be essential for a proper dosimetric evaluation of its impact. Particles with similar size and containing mainly alpha emitters, such as the U/Pu particles, will have a different radiological impact depending if the radioactive content is in the central part of the particle covered by a layers of inactive material or if they are mostly located in the outer layers of the particle.

Typical particles generated in places contaminated by local fallout associated to nuclear weapon tests present a glassy structure with the exterior surfaces dominated by elements such as silicon, calcium and other light elements as elucidated using for example SEM-EDX or PIXE. Within the particle structures, the location of the radionuclides relative to the surface is important for dose considerations and radionuclide release during weathering.

In one glassy fission fragment from a detonation test at Maralinga, the ^{137}Cs was clustered on the exterior, while ^{90}Sr occurred mainly in the interior of the same fragment. In contrast, a Pu-dominated particle from the safety trials at Maralinga indicated an inhomogeneous core-shell structure, with most Pu in the interior, (Figure 10)(Ikeda-Ohno et al., 2016). Understanding the structure of particles is important since it is their external surface that interacts with the environment, or in the case of inhalation or ingestion, with potential lung or gastrointestinal tissues and fluids. The particle surface was dominated by Ca, Fe, and U originating from the indigenous Maralinga soils, while the Pu was present as an oxyhydroxide species similar to particles isolated from Palomares (Lind et al., 2007) and Thule (Lind et al., 2005).

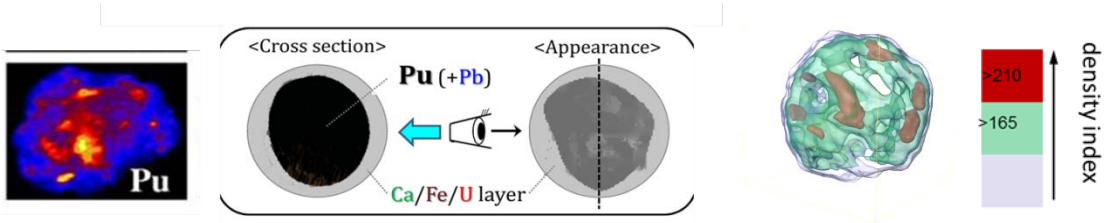


***Figure 10**. Synchrotron XFM data (left) indicating most Pu is situated in the interior, while Ca, Fe, and U is situated on the exterior shell of a particle from the non-nuclear testing at Taranaki, Maralinga. The right image is of a different particle from the same site, showing higher density regions expected to contain Pu and U (M. Johansen pers.comm, OC. Lind, pers.comm).*

The retention of radioactive particles in mammals by ingestion has been evaluated in previous sections of this report as a phenomenon with quite low probability of occurence. All the evidence indicates, that in case of ingestion most probably the particle will flow through the digestive system, being excreted in the faces. In its transect along the gastrointestinal system all the experiments performed indicated also the relatively low solubility of the radioactive content of these particles and consequently the low percentage that can be distributed over the animal body under consideration in solubilized form, in the affected ecosystems analysed by RATE. The application of conventional models

not considering these peculiarities conduit to an overestimation of dose rate that in some cases can be considerable.

The possibility of retention in mammals can only play a role for particles with big sizes, with possible retention in the intestine, where the particle may act as point source. However, in the ecosystems that we have analysed in our work (Palomares, Thule, Semipalatinsk, Maralinga), if this phenomenon occurs, the particle will mostly be formed by alpha emitters (Pu and U) and the self-shielding of the alpha emissions due to the dimension of the particle will be considerable, with all the consequent implications in the dose estimations. Hence, in ecosystems affected by refractory U/Pu releases as the evaluated in this work, the main radiological impact will be associated nowadays to the inhalation route of particles with "respirable" size.

8 Conclusions

To assess environmental impact and risks associated with radioactive particle contamination of ecosystems, links must be established between the source term and deposition, ecosystem transfer, biological uptake and effects in exposed organisms.

It is well established that a major fraction of refractory radionuclides such as uranium (U) and plutonium (Pu) released to the environment from the nuclear weapon and fuel cycles is present as particles ranging from submicrons to fragments.

Following deposition, radioactive particles can be retained in soils and sediments. Thus, the ecosystem transfer of particle associated radionuclides will be delayed compared to mobile species. Due to particle weathering processes, particle associated radionuclides are remobilized and contaminated soils and sediments can act as diffuse sources of radioactivity in the future. A challenge is therefore to link particle characteristics to ecosystem behaviour, and to identify how environmental factors such as pH, redox, TOC and the influence of microbial activities can influence on particle weathering processes. Thus, information is needed not only on transformation processes and interactions, but also on the kinetics of the processes and to utilize this information to replace thermodynamic constants (Kd, CR, TF, TC) with time functions, as has been demonstrated in this work.

The work has also demonstrated that radioactive particles are of biological relevance. Particles can be inhaled or ingested, and can be retained within organisms such as snail shells, acting as point sources. The bioavailability and uptake of particle-bound radionuclides compared with those existing as ions or simple molecules has so far largely been ignored when impact and risk are assessed. As a result, there is a high degree of scientific uncertainty about the long-term ecological consequences and risk to human health from radioactive particles present in the environment. As particles and their associated dose will be inhomogeneously distributed within organisms, a key challenge is to improve dosimetry models used for characterizing uneven distribution of dose from high-specific activity particles, being inadequate today.

Although RATE has contributed with important knowledge on particle weathering and leaching being essential for ecosystem transfer, as well as on biological relevance of particles, further research is clearly needed to address the challenges identified. As particle releases were seen following all kinds of historic nuclear events, particle releases should also be expected for events occurring in the future. Therefore, the accidental sites should be utilized to improve the knowledge needed to link particle

characteristics to sources, to transfer and to biological effects on which sound environmental assessments can be made.

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