CONFIDENCE FOODCHAIN IMPROVEMENTS



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CONFIDENCE overview of improvements in radioecological human food chain models and future needs

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Abstract – Radioecological models used to make predictions of the radionuclide activity concentrations in human foodstuffs must be sufficiently robust and fit for purpose with uncertainties reduced where practicable. The CONFIDENCE project had a work package with the objective to improve the capabilities of radioecological models and this paper presents the key findings of this work. Recommendations for future radioecological studies/model developments are made based on the findings of the work conducted and consultations with end-users.

Keywords: food chain / radioactive particles / caesium / ¹³¹I / ⁹⁰Sr

1 Introduction

There are considerable uncertainties associated with the radioecological simulation models used to predict the transfer of radionuclides along food chains. Initially after an accidental release, the factors determining the contamination of foodstuffs will largely be defined by vegetation interception and the time of year. During the transition phase, factors controlling the uptake of radionuclides to vegetation from soil will become more important and these will dominate during the long-term rehabilitation phase. However, predictions made using radioecological models may be used in the early part of the transition phase to make longer-term decisions, such as those associated with remediation strategies. Therefore, models must be sufficiently robust and fit for purpose with uncertainties reduced where practicable. A classic example of where predictions were made using models/information not fit for purpose is the post-Chernobyl case in upland United Kingdom. In 1986, it was stated that restrictions on sheep management because of high radiocaesium levels following the Chernobyl accident would last for a matter of weeks (Wynne 1992). However, restrictions remained in place until 2012.

The work programme of CONFIDENCE WP3 had three over-arching and interlinked tasks:

- improving models:
- characterise and analyse the underlying probability distribution functions (PDFs) associated with transfer parameters to better enable uncertainty/sensitivity analyses,
- conduct targeted field ¹³¹I tracer studies on the plantanimal-milk pathway,
- characterise the behaviour of radionuclides in Mediterranean production systems (including seasonality and key regional produce),

The objective of the CONFIDENCE project's Work Package 3 (WP3) was to improve the capabilities of radioecological models used to predict activity concentrations in terrestrial foodstuffs and to better characterise, and where possible, reduce uncertainties. The work programme addressed key challenges identified in the Radioecology ALLIANCE Strategic Research Agenda (Hinton *et al.*, 2013) and specifically those of the Human Food Chain Roadmap¹.

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https://radioecology-exchange.org/sites/default/files/T1_WG_for% 20Radioecology%20Roadmap_Human%20Food%20Chain_ver sion02022015.pdf

- consider how recent knowledge would change/improve terrestrial food and dose module predictions,
- learn from post-Fukushima experiences,
- evaluate the application of extrapolation approaches (phylogeny, allometry, stable elements [see Beresford et al., 2016]) to improve predictive ability for poorly studied radionuclides;
- can process-based models reduce uncertainties?:
- determine why existing process-based approaches for Cs gave poor predictions after the Fukushima releases,
- investigate the applicability of process-based Cs model to European soil types (focusing on soil types not included in model parameterisation/validation studies),
- investigate process-based model options for Sr,
- assess the added value of using processed based models,
- investigate how (spatial and temporal) process-based models can be incorporated into decision support systems (DSS);
- including radioactive particles in radioecological models:
- incorporate radioactive (or "hot") particles into models to improve predictions.

Results from the work programme can be found in a number of deliverable reports (Almahayni *et al.*, 2019a; Beresford *et al.*, 2019; Brown *et al.*, 2018; Guillén, 2019; Lind *et al.*, 2019) available from https://concert-h2020.eu/en/Publications. In this paper, we give an overview of key findings and make recommendations based upon the work programme and consultations with end-users.

2 Key findings

The key findings of WP3 are highlighted and discussed below.

2.1 Incorporation of the FDMT model into a flexible modelling platform (ECOLEGO)

The FDMT model is the food chain transfer module of the JRODOS and ARGOS decision support systems (Müller et al., 2004; Brown et al., 2018). ECOLEGO is a modelling platform for creating dynamic models and performing deterministic or probabilistic simulations (Avila et al., 2005; http://ecolego. facilia.se/ecolego/show/HomePage). The software incorporates powerful numerical solvers for complex and dynamic systems (i.e. solver for ordinary differential equations including "stiff" problems) and provides support for probabilistic simulations using Monte Carlo or Latin Hypercube sampling. Incorporating FDMT into ECOLEGO (hereafter referred to as the "FDMT-ECOLEGO model") allowed us to conduct sensitivity analyses (Brown et al.. 2018), investigate regionalisation (Brown et al., 2018; Beresford et al., 2019) and replace the default model components with process-based models (discussed below) for soil-plant transfer (Almahayni

et al., 2019a; Beresford et al., 2019) and the presence of radioactive particles in soil (Lind et al., 2019).

2.2 Development and assessment of soil-plant transfer process-based models

Commonly used models to predict radionuclide activity concentrations in human foodstuffs tend to use empirical soil-to-plant transfer factors (also known as soil-plant concentration ratios) to describe the transfer of radionuclides from soil to crops (*e.g.* Brown and Simmonds, 1995; Brown *et al.*, 2018). Such models cannot easily cope with variation in root uptake caused by variation in soil properties (*e.g.* Bogdevitch *et al.*, 2002; Panov *et al.*, 2009). Process-based models offer an alternative, which take into account soil (and potentially plant) characteristics.

2.2.1 Development of process-based soil-plant models for Sr

The Chernobyl accident highlighted that some areas may be more "sensitive" or "vulnerable" (e.g. have comparatively high transfers to foodstuffs or contribute relatively high fluxes of radionuclides to the public via contaminated foodstuffs) to radiological contamination than other areas (e.g. see Howard et al., 2002). This lead to the development of process-based soil-plant models that were parameterised using commonly characterised soil parameters (see Almahayni et al., 2019b). However, the development of such models was restricted to radiocaesium (see below). We have developed two processbased approaches for predicting soil-plant transfer of Sr. One of these models was based upon adaptation (and simplification) of an established chemical speciation model (Tipping et al., 2011), the other requires simply a 90 Sr concentration in soil and the Ca concentrations in soil and crop(s) of interest (Almahayni et al., 2019a). The models gave predictions of Sr concentrations in a range of crops grown on different soil types (Barnett et al., 2019b) which were in considerably better agreement with measured data (R²>0.67) than prediction using traditional empirical plant-soil concentration ratios ($R^2 = 0.11$) (Almahayni et al., 2019a). Currently these models give equilibrium predictions; consideration is needed to their incorporation, after further testing and validation, into models making dynamic predictions. To support the application of the developed models a dataset containing Ca concentrations in a range of crops has been established (Chaplow et al., submitted). A demonstration of the application of one of the Sr process-based models to a deposition scenario (De Vries et al., 2019) can be found in Brown et al. (2020).

2.2.2 Assessment of the strengths and weaknesses of the "Absalom" process-based soil-plant model for Cs

In Almahayni *et al.* (2019b), we reviewed soil-pant transfer modelling approaches for radiocaesium and concluded that the process-based model initially described by Absalom *et al.* (1999) was practical, robust and fit for purpose. However, predictions using this model had been shown to be relatively poor for some crops and/or non-European soils (Almahayni *et al.*, 2019a).

To further test the "Absalom" model (as described in Absalom et al., 2001) we determined radiocaesium transfer to grass, radish (edible root) and spinach from 20 arable and pasture soils collected from Norway, Belgium, the UK and Spain. The soils covered a range of pH values, organic matter contents, clay contents, CEC (cation exchange capacity) and RIP (radiocaesium interception potential) values. The transfer of radiocaesium to plants in our studies varied by up to three orders of magnitude across soil–plant combinations.

The Absalom model predicted the radiocaesium transfer to grass and radish edible root relatively well (predictions were mostly within an order of magnitude of the measurements). However, predictions for spinach were relatively poor. We recommend expanding the Absalom model database by considering more soils (with different mineralogies) and plant types in its parameterisation.

The Absalom model was implemented within the FDMT-ECOLEGO model and Brown *et al.*, (2020) discusses its application to a scenario case study.

2.2.3 Recommendation on when to use process-based soil-plant models

As already noted, in Brown et al. (2020) we demonstrate the application of soil-plant process-based models for Cs and Sr to the scenario as described by De Vries et al. (2019) (an accidental release from a nuclear power plant). The models were run for five diverse, though not extreme, soils; the default FDMT model was run for comparison. At the end of the prediction period (c. 27 years after deposition) there was approaching three-orders of magnitude difference between the minimum and maximum predicted ¹³⁷Cs activity concentration in milk: for ⁹⁰Sr the difference was nearly one-order of magnitude. However, over the first six months after deposition predictions for most soil types were generally similar and also similar to predictions using FDMT default parameters; the only exception were predictions for an organic soil (68 % organic matter) and ¹³⁷Cs for which predictions were approaching an order of magnitude higher than FDMT after 90 days.

We recommend that in the short-term, process-based soil-plant models for Cs and Sr will generally give little added benefit, *i.e.* models such as FDMT are sufficient for predictions during this phase (because soil-plant transfer contributes little to radionuclide activity concentrations of crops in the short-term). However, longer-term predictions made using FDMT, or similar models, during the early phase after a deposition event should be communicated with care. Process-based models should be used to make longer-term predictions, and identify potentially vulnerable areas, once spatial predictions of deposition are available.

2.3 Development of a model for radioactive particle behaviour in the soil plant system

Following severe nuclear events, radioactive particles maybe released into the atmosphere and deposited in the environment (e.g. Kashparov et al., 2018). A bespoke compartmental model was conceptualised based upon an understanding of particle characteristics and behaviour, based on comprehensive particle archives and associated databases

(Lind *et al.*, 2019). Parameters, such as those describing particle weathering rates and leaching rates from soils containing particles were derived from laboratory and field experiments. The model parameterisation of U fuel particle weathering rates, which strongly depends on soil pH and solid-state speciation of the carrying matrix (*i.e.*, oxidized or non-oxidized UO₂ fuel particles, or U transformed to extra inert forms such as UZr_xO_y) was based on extensive datasets from the Chernobyl exclusion zone. The developed particle-soil model was then implemented into the FDMT-ECOLEGO model replacing the default soil radionuclide transfer models (Lind *et al.*, 2019).

Running the revised model (including comparison with data from close to the Chernobyl exclusion zone) suggested that in the short term following an accidental release, accounting for the potential presence of radioactive particles in the soil is unlikely to be critical (ingestion dose rates, from the soil-plant pathway may be overestimated if particles are not taken into account during this phase). In the longer term (decades), not accounting for particles in the deposit may underestimate ¹³⁷Cs and ⁹⁰Sr transfer to food products (Lind et al., 2019).

2.4 Assessment of phylogenetic models for crops and Cs and Sr

For approaching 20 years, it has been suggested that "phylogenetic relationships" offer a scientifically supported extrapolation approach to determining radionuclide activity concentrations in plants (e.g. see Willey, 2010). However, to our knowledge, the ability of the approach to predict radionuclide activity concentrations in crops has never been tested. We tested published phylogenetic relationships for Cs (Beresford and Willey, 2019) and Sr (Willey and Fawcett, 2006) using data from a plant growth study considering a range of soils and crops (Barnett et al., 2019b). Predictions were generally acceptable (within an order of magnitude of measured values) with the exception of those for Sr transfer to fruits/tubers. This is likely because the original phylogenetic relationships were established using measurements for shoots and not fruits/tubers.

2.5 Radionuclide biological half-lives for farm animal products

Many predictive models use biological half-lives (or rate constants derived from them) to describe the rate of loss of radionuclides from animal tissues and products (e.g. Müller and Pröhl, 1993; Brown and Simmonds, 1995). However, whilst there have been international compilations of transfer parameters for modelling purposes (e.g. IAEA, 2010), these have not considered biological half-life values. To address this, we have conducted a review of biological half-life values for farm animal products (meat, milk, eggs, etc.) and compiled a dataset of quality-controlled entries. The final dataset contains over 600 entries for 12 animal types (cattle, sheep, goats, deer, geese, hens, horses, pigs, rabbits, camels, ducks and red grouse) for 33 elements relevant to radiological protection. Entries include values for milk, muscle (meat), eggs, whole body, carcass and various tissues (e.g. liver and kidney).

The dataset has been published (Barnett et al., 2019a) and will be used to make recommendations in a future publication.

2.6 Regionalisation

Regionalisation has been considered in two ways:

- variation of largely non-radiological parameters within the FDMT-ECOLEGO model to better match specific regions of Europe (for the purposes of demonstration these were Norway and Spain);
- the collection of transfer parameter data for Mediterranean systems.

Discussion of the effect of replacing model default parameters with regional values, for instance for animal diets, animal slaughter times, crop harvest time, productivity, can be found in Brown *et al.* (2018) and Beresford *et al.* (2019). The magnitude and temporal development of the radionuclide activity concentrations in foodstuffs are seasonally dependent and hence using regionally appropriate parameters, such as harvest dates, can impact considerably on the predicted transfer of radionuclides to the human diet.

Compilations of radionuclide transfer parameters for the human food chain (e.g. IAEA, 2010) are dominated by data for temperate ecosystems. With respect to Europe, data are sparse for Mediterranean ecosystems. Guillén (2019) discusses the sampling of wheat, triticale, grapes (including wine), olives (including olive oil), lamb, beef, pork and dairy products from sheep, goats and cows from throughout Spain. The resultant data were used to derive transfer parameters for Mediterranean production systems.

2.7 I-131 tracer studies on the plant-animal-milk pathway

In the early phase of an emergency situation, ¹³¹I is one of the most important radionuclides for which information on contamination of human foodstuffs is essential. There is also the potential for economic and societal consequences from the loss of crops that are vulnerable to contamination, particularly those with a short harvest to market window, such as leafy vegetables, soft fruits or new potatoes.

2.7.1 Field plant studies

Field tracer experiments using ¹³¹I have been carried out at two sites in Norway: a coastal site with high sea salt and stable iodine deposition, and an inland site with low salt and iodine deposition. Iodine-131 was sprayed onto plots on which different standing crops (grass, barley, strawberry and potato) were growing.

Results showed that ¹³¹I concentrations in grass and barley at both sites were dominated by interception and changes in biomass, with little wash-off from plant to soil. There was also no discernible soil to grass transfer and no effect of stable I on vegetation activity concentrations. These results support previous studies on the importance of biomass on ¹³¹I interception, but also demonstrate that changes in plant concentrations after a deposition can be adequately modelled

by biomass changes. Results for potato and strawberry plants showed a small, but measurable transfer of 131 I from leaves to tubers and from leaves/flowers to fruit three weeks after spraying (leaves/flower to fruit/tuber concentration ratio of <0.02 and <0.10, by fresh mass (FM), respectively).

2.7.2 The influence of protein source on the transfer of I-131 to milk

Results from a study in which cows were administered ¹³¹I showed that rapeseed (which contains goitrogens) in the diet resulted in lower ¹³¹I activity concentrations in milk as a consequence of reduced transfer of ¹³¹I from blood to milk; there was increased excretion of ¹³¹I *via* urine.

3 Recommendations

We present recommendations based upon the outcomes of our work programme as discussed above and reported in various deliverables (Almahayni et al., 2019a; Beresford et al., 2019; Brown et al., 2018; Guillén, 2019; Lind et al., 2019). We also present recommendations from a joint workshop (September 2019) organised by CONFIDENCE WP3 in association with the Radioecology ALLIANCE Human Food Chain Working Group. The recommendations and findings from CONFIDENCE WP3 have helped to revise the Strategic Research Agenda (SRA) for radioecology (Salomaa, 2019).

3.1 Recommendations for future research arising from CONFIDENCE studies

The following recommendations are based upon the work we conducted within CONFIDENCE WP3:

- soil-plant process-based models are worth pursuing for Cs and Sr;
- the "Absalom" process-based model for Cs soil-plant transfer needs to be tested, and potentially adapted, for a wider range of crops grown on a variety of soil types with differing mineralogies;
- the soil-plant process-based models developed by CON-FIDENCE for Sr need further validation and testing; consideration needs to be given on how to incorporate the models into dynamic food chain models such as FDMT in JRODOS and ARGOS;
- how to use the potential ability of soil-plant process-based models to model the effect of soil based countermeasures (namely K-fertilisation and liming) needs to be better considered and included into DSS;
- scientists need to clearly make the case for using processbased models in post-accident management and be clear when they would be useful; training (appropriate to specific audiences) in the use of process-based models needs to be developed and provided;
- there is a need to include uncertainties in models and their outputs. This work has been started in CONFIDENCE WP3 for the FDMT in ECOLEGO model, but further work is needed to expand the statistical data collation to parameters not originally covered and to consolidate the information for those parameters that have been considered;

- further work is needed to perform a global sensitivity analysis and investigate the correlation between parameters.
 Discussion is required on how ignoring interdependency of variables in a model can contribute to uncertainty. This may provide a deeper understanding of the model behaviour by interpreting the dependency and interaction pattern;
- it is recommended that FDMT parameters be updated.
 Greater transparency should be provided where parameters have to be extrapolated because data are lacking; where data are lacking experimental work should be encouraged;
- there is a need to consider deposition, interception and retention of radioactive particles, all processes which are likely to be important in the early stages (weeks and months) post-accident; such evaluations should also consider the potential for animals (and humans) to ingest radioactive particles;
- phylogenetic models, which allow predictions to be made for a wide range of crops for a given site without the need for soil type specific studies, need to be validated and where required, parameterised using the component of plants consumed;
- the application of transfer parameters derived from stable elements in radioecological models need to be further reviewed (see Beresford *et al.*, 2019);
- transfer parameters are required for some regionally important agricultural products in Europe (e.g. nuts, rice, sunflower products) (though to some extent if taken forward phylogenetic and potentially soil-plant processbased models [for Sr at least] may negate the need for some data collection).

3.2 Future priorities for human food chain radioecological studies from end-user consultation

In September 2019, CONFIDENCE WP3 held a joint workshop with the Radioecology ALLIANCE (Beresford et al., 2019). The approximately 40 participants included representatives from regulatory bodies, industry, governmental agencies and the IAEA. The aim of the workshop was to identify future priorities for radioecological research with respect to the human food chain; discussions are summarised below (see Beresford et al., 2019 for further information).

3.2.1 Radionuclides

There was wide agreement that data on some radionuclides was poor and that some emphasis should be given to providing data and/or recommending modelling approaches. Radionuclides highlighted included:

- those released by medical facilities for which data are poor or often totally lacking (e.g. radioisotopes of Cr, F, Fe, Ga, Ho, In, La, P, Re, Sm, Tc, etc.);
- radionuclides associated with the decommissioning of nuclear licenced sites (including, ^{108,108m}Ag, ²⁴³Am, ¹⁰Be, ⁴¹Ca, ^{152,154,155}Eu, ^{55,59}Fe, ²⁰³Hg, ⁹³Mo, ²²Na, ^{93m}Nb, ¹⁴⁷Nd, ^{93m}Nb, ¹⁹³Pt, ⁴⁶Sc, ¹⁵¹Sm and ¹⁸²Ta);
- radionuclides relevant to fusion reactors (including activation products such as radionuclides of Ag, Fe, Mn, Nb, Ni, Tb in addition to focussing on ³H); long-lived radionuclides associated with geological disposal facility assessments.

It was noted that requirements for data for many of these radionuclides was not only restricted to the human food chain, but also to the need for parameters for biota assessments. In some instances, it is likely that doses to the public through food consumption (and also doses to biota) will be low from some of these radionuclides (short-lived radioisotopes discharged from medical facilities likely being an example). However, assessments have to be conducted by industry/regulators to assess the potential impact of these radionuclides. Therefore, we need to advise on how best to conduct these assessments such that they are fit for purpose and proportionate. A scoping study on how best to address this need is required as a first step.

3.2.2 Regionalisation

There is a need to take into account potential regional variation in radionuclide transfer and also regional variation in diet and farming practices, including seasonality (e.g. in northern Europe farm stock are fed stored forage in winter, in southern Europe stored forage may be fed in summer) (Brown et al., 2018; Beresford et al., 2019). Radionuclide transfer data in compendia such as IAEA (2010, 2014) are biased towards temperate systems. Data are also lacking for what were termed "exotic foodstuffs" which, in some instances, may be regionally important (e.g. snails, dates, wine). There is also a need to consider our ability to predict radionuclide behaviour under changing climate scenarios. CONFIDENCE WP3 and other recent work have made a step to providing radioecological data for Mediterranean ecosystems (Guillén et al., 2018, 2019; Guillén, 2019).

3.2.3 Novel foodstuff and changing agricultural practices

Our diets and agricultural practices evolve continually with different (potentially new to Europe or a given country) foods gaining popularity (chia and quinoa would be relatively recent examples, with interest in insect based foods for farm livestock and humans currently increasing [e.g. van Huis et al., 2013]). The workshop recommended that we need to ensure our models (and underlying data) keep up with changes in diet and foodstuffs. To some degree, "phylogeny-based" extrapolation approaches and/or some process-based models may help us to derive radionuclide transfer parameters for novel foodstuffs. With respect to agricultural production, it was noted that satellite data could be used to identify agricultural production (what crop is grown where and when) and to estimate crop yields (e.g. https://www.ceh.ac.uk/crops2015).

3.2.4 Innovative ways of providing transfer parameters

In reality, the number and diversity of radionuclides and foodstuffs (and for biota assessments, wildlife species) means that, we are never going to have data for everything. In some ways, clear acknowledgement of this, and the consideration of open and robust extrapolation approaches, has progressed further for biota/wildlife models (*e.g.* Brown *et al.*, 2013; Beresford *et al.*, 2016) than for human food chain models. There was general support for the use of phylogenetic models (as discussed above) with the recommendations that the models needed to be for the plant parts consumed and that more rigorous testing was required. It was also suggested that

the phylogenetic approach could be a useful "add-on" to process-based models, which have been parameterised for radionuclide transfer to grass (*e.g.* see discussion in Almahayni *et al.*, 2019a).

Ionomics and/or ecological stoichiometry were also suggested as scientifically based extrapolation approaches, whereby similarities in the behaviour of some elements/radionuclides could be used to make predictions of radionuclide activity concentrations in foodstuffs (or biota) (e.g. Sr predictions based on Ca data would be an example). Whilst this has been suggested previously (see Beresford et al., 2016) to date little progress has been made.

For farm animals, it was recommended that there should be a move away from the transfer coefficient (defined as the ratio of the radionuclide activity concentration in an animal derived foodstuff to the daily intake of the radionuclide) towards using the dietary concentration ratio (*i.e.* the ratio of the radionuclide activity concentration in an animal derived foodstuff to that in its diet [on a dry matter basis]) instead. Concentration ratios for one animal can be used with some confidence for other animals (farm livestock and potentially wildlife) for which data are lacking (see discussion in Beresford *et al.*, 2016).

Well-founded extrapolation approaches will also help us to address the lack of data for many radionuclides and the need to upkeep parameter databases/models to account for novel foodstuffs.

3.2.5 Foliar uptake

There was general recognition that radionuclide interception by plants and subsequent retention and translocation has received relatively little attention. The lack of relevant data was highlighted after the Fukushima accident with unexpected transfer of radiocaesium to fruit being reported (*e.g.* Sato *et al.*, 2015).

3.2.6 Remediation

There was discussion of the work of Penrose *et al.* (2015, 2016, 2017) on the selection of plant varieties with low radionuclide uptake. It was noted that in the event of any future accident, low accumulating varieties could be identified relatively quickly by collaborating with the many worldwide plant-breeding programmes. There was also the suggestion that CRISPR (clustered regularly interspaced short palindromic repeat; Wang *et al.*, 2019) technology could be used produce crops with low uptakes. However, there are socio-political challenges associated with CRISPR technology as it may be considered as genetic modification of organisms.

There were recommendations that modelling approaches to improve the assessment of soil based countermeasures were needed, which links to the recommendation above that we need to better exploit the potential ability of process-based models to consider the effect of soil based countermeasures.

3.2.7 Radioecological models

In addition to the specific recommendations above, the needs to communicate radioecological models to end-users and to ensure model validation were stressed. Users (regulators, governmental agencies and ministries) need to

have confidence in the outputs of the models at their disposal. The example was given of the lack of confidence of Japanese authorities to use predictions from the Japanese government's System for Prediction of Environmental Emergency Dose Information (SPEEDI) in the management of the post-Fukushima situation (see Funabashi and Kitazawa, 2012).

Model validation would benefit from participation in programmes such as those organised by the IAEA (*e.g.* MODARIA II follow-on²).

Other comments on radioecological models were:

- the need to include uncertainties in models and their outputs;
- lack of consideration of the presence of other contaminants:
- parameterise models with parameters which are readily available or relatively easy to determine;
- predictive models should be linked to monitoring data, such that the monitoring data can be used to refine assessments:
- the need to consider the societal consequence of models being wrong and/or over-conservative.

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² https://www-ns.iaea.org/projects/modaria/

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