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Paper

Projected external doses from an accidental release of ESS spallation-target products: time-dependence and radionuclide contribution

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Abstract

The Swedish Radiation Safety Authority has presented a report on severe accident scenarios at the European Spallation Source (ESS) for dimensioning the emergency preparedness zones around the facility. The source-term in the scenario consisted of more than 80 tungsten-target spallation products with physical half-life ($T_{½}$) exceeding 1 hour. The purpose of this study is to establish which of these radionuclides will become of highest importance in terms of the radiological consequences to residents in areas affected by an accident release. In analogy with accidents at nuclear power plants, where the fission product ^{137}Cs is a key nuclide for estimating projected external doses to affected residents, a corresponding key nuclide for ESS is required for the emergency preparedness. Using existing accident source terms in combination with reported values on ecological half-times of the gamma emitter ^{137}Cs, the external dose rates and cumulative doses per unit initial ground deposition of the suggested key-nuclide (^{182}Ta) could be estimated. In terms of 50 y dose from a dry deposition of the released source-term, ^{172}Lu ($T_{½}$ = 6.7 d, supported by ^{172}Hf with $T_{½}$ = 1.87 y) contributes up to 50% of the 50 y dose, depending on the ecological half-times for the element. The isomer ^{178n}Hf ($T_{½}$ = 31 y) is the second largest contributor to gamma-ray dose, followed by ^{182}Ta ($T_{½}$ = 115 d), that contribute with about 15% and 10% of the 50 y dose, respectively. The results thus suggest that ^{172}Hf/^{172}Lu may be more suitable for long-term follow-up of projected doses from accidental ESS releases than ^{182}Ta.

Introduction

The European Spallation Source (ESS) is currently under construction outside of Lund, in southernmost Sweden [1, 2]. Once open for research (anticipated in 2027), it will act as a giant microscope using neutrons as probes to study different materials in a broad variety of scientific fields ranging from molecular biology to nanotechnology. The facility will consist of a 500 m long linear accelerator, which will deliver a pulsed proton-beam with an energy of up to 2 GeV and an average power of up to 5 MW. Neutrons will be produced through spallation reactions occurring when the proton beam hits a helium-cooled, 11-tonne rotating target wheel containing hundreds of tungsten bricks. The neutrons will be moderated to thermal and cold energies before being directed to the neutron scattering experiment stations. 6000 tonnes of steel will surround the tungsten target for shielding of the very high intensity radiation being produced in the spallation target.

An inevitable consequence of the spallation process itself, of neutron activation and other processes in the target is the production of over 1000 different radionuclides (e.g. [3]). In 2018, a dimensional accidental scenario of the ESS facility was described by the Swedish Radiation Safety Authority [4]. The report summarized

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a scenario that would be the basis for dimensioning the emergency preparedness zones and site category classification according to IAEA recommendations [5]. The radionuclides that will be generated by the ESS will differ from those produced by nuclear reactors and other types of accelerator facilities. Simulations have shown that the ESS target will contain several radionuclides that are unique products of the high-energy spallation reactions at the ESS [6–8], and that some of these are expected to give the highest dose contributions in case of an accident [4], such as the alpha emitters ^{148}Gd (through internal contamination) and the gamma emitters ^{182}Ta (though external contamination). The source terms defined in these studies all contain at least 100 gamma-emitting radionuclides with a physical half-life >1 hour, and at least 4 that decay to gamma-ray emitters.

In this study, effective-dose calculations were based on the accident source-term estimated by SSM [4] and, in particular, the radionuclides termed as "Volatile" and "Less volatile". According to SSM's calculations, the most important radionuclides during the first 7 d after a worst-case scenario (referred to as an H5 event) accident are estimated to be ^{148}Gd, ^{187}W, ^{172}Hf, ^{182}Ta, and ^{178n}Hf, with ^{148}Gd having the highest dosemetric impact. SSM [4] also states that for this dimensioning accident scenario, the main part of the effective dose during the first 7 d will be dominated by the inhalation dose [4], followed by the so-called 'cloudshine' (external dose from passing airborne release plume) and 'groundshine' (external dose from radionuclides deposited on the ground after the passage of the radioactive plume). In this study the focus was placed on the external dose contribution over the long-term (up to 50 y) from the groundshine.

The specific aims of this study have been two-fold: firstly, to investigate which of the potentially released nuclides contribute the most to groundshine over a time period longer than 1 y; and secondly, to search for a suitable key nuclide, to be used in a similar manner to ^{137}Cs in the long-term environmental assessment of fallout from nuclear fission releases (e.g. [9, 10]). This nuclide should (i) be a gamma-ray emitter readily detectable by in-situ gamma-ray spectrometry, (ii) have a high enough T½ to monitor the long-term trend in the external dose rate, and (iii) contribute substantially to the 50 y projected dose together with its progenies. In connection with *in-situ* gamma-ray spectrometry, such a key nuclide could be used to monitor the projected external dose based on the local deposition density (kBq m^{-2}). In the report from SSM [4], it was suggested that ^{182}Ta could be used as such a key nuclide with respect to groundshine. In this study, the extent to which ^{182}Ta be used for projecting external doses over times as long as 50 y was investigated.

Material and methods

The activities of 50 gamma-ray emitting nuclides, $a_{Source,i}$ (Bq), listed in the SSM report (2018) were normalized to the source term activity of ^{182}Ta, since it has been suggested as a suitable reference nuclide. External dose coefficients, expressed in nSv h^{-1} Bq^{-1} m^2, for various infinite source geometries taken from the International Commission of Radiological Protection (ICRP) [11] were applied to each radionuclide, assuming a uniform planar deposition with either a 0 or a 0.5 g cm^{-2} initial ground penetration depth. These depths roughly represent the initial ground penetrations from a dry and wet fallout deposition event respectively. In this study, only the effective dose to adults was considered, although a possible extension of this study also will consider effective dose to children, equivalent dose and lifetime attributable risks.

In the computation of the time-integrated doses up to 50 y, the reduction of the external dose, by gradual migration of the fallout into soil layers, has also been accounted for. This phenomenon, that can cause the external dose rate above ground to decrease faster than the physical half-life alone, has been widely reported in previous studies on atmospheric fallout (e.g. [12–14]). The effect is accounted for by an 'ecological half-time' which can, for the element cesium, vary between a few years to several decades (e.g. [13]). In this study, the bi-exponential decay function:

$$Eco_i(t) = c_{short,i} \cdot e^{\frac{-\ln(2)}{T_{eco,short,i}} \cdot t} + \left(1 - c_{short,i}\right) \cdot e^{\frac{-\ln(2)}{T_{eco,long,i}} \cdot t} \quad (1)$$

based on the existing knowledge of cesium migration in soil (Gale et al., 1964 [15]), was used to represent the effect of the ecological half-time. It was assumed that the time components $T_{eco,short,i}$ and $T_{eco,long,i}$, range between 0 to 1 y and 1 to 100 y, respectively. The values for $c_{short,i} = 0.5$, $T_{eco,short,i} = 1$ y and $T_{eco,long} = 10$ y were chosen as reference values, based on studies of land areas within Sweden affected by Chernobyl ^{137}Cs [12]. In the absence of a precise knowledge of the ecological half-times for many of the elements included in the source term, a similar $Eco(t)$ function for all elements was assumed.

The cumulative effective-dose 1 m above ground from the sum of the contributions from the ground-deposited radionuclides, i, normalized to detected initial ground deposition of ^{182}Ta, E(t) (mSv/(kBq m^{-2})), can hence be computed according to:

$$E(t) = \frac{\sum_i \left(\frac{a_{source,i}}{a_{source,Ta-182}}\right) \cdot e_{ICRP144,i} \int_0^t e^{-\left(\frac{\ln 2}{T_{\frac{1}{2},phys,i}}\right)t} \cdot Eco_i(t)dt}{A_{dep,Ta-182}\,(t=0)} \quad (2)$$

where $a_{source,i}$ is the specified released activity in the SSM source term for a given radionuclide i, and

$a_{source,Ta\text{-}182}$ refers to the specified releasable activity amount of ^{182}Ta. $e_{ICRP144,i}$ is the nuclide specific dose coefficient between ground deposition and effective dose 1 m above ground (mSv kBq m^{-2}) given by the ICRP [11]. $T_{½,phys,i}$ is the physical half-life of the radionuclide (y) and $A_{dep,Ta\text{-}182}$ is the fictitious initial activity deposition density (kBq m^{-2}) at a location within an area of the potential release from the source (ESS). Eq. 2 allows for the computation of both the momentaneous effective-dose with time, as well as the relative radionuclide-specific contributions to $E(t)$ and the cumulative radionuclide contribution to E over long term ($t = 50$ y). The results of these computations then give a measure of which of the radionuclides are most important in terms of the projected long-term exposure in the affected area.

To investigate the effect of the nuclide composition on the estimated E(50 y)-values, an alternative source term of the accidental release was used, taken from the FLUKA simulation performed by Baurkaukas and Stenström [6]. However, since the FLUKA simulations did not include isomers such as ^{178n}Hf (referring to the higher (n) isomeric state that is not supported by ^{178}W), an activity amount of this isomer was added in the alternative source-term in proportion to that of the ^{182}Ta activity in the SSM-source term. The source terms are presented in Table 1.

Results and discussion

Time-integrated effective dose and effective dose rate per unit initial ground deposition from an accident fallout

The time-integrated effective dose over 50-y per unit of initial deposition density of ^{182}Ta, $A_{dep,Ta\text{-}182}$ (kBq m^{-2}), is estimated to 0.13 and 0.082 mSv/(kBq m^{-2}) for dry and wet deposition, respectively, using the source term from SSM [4]. If instead using the target inventory from Baurkaukas and Stenström [6] the corresponding values become 0.23 and 0.15 mSv/kBq m^{-2}.

The results of the time-dependence of the effective dose rate are illustrated in Fig. 1(Left), which show that the effective dose-rate will, after an initial rapid decline over the first 10 y, decline according to the effective ecological half-time of the nuclide with the longest physical half-life, namely ^{178n}Hf. In both cases about 40% of the 50-y effective dose is delivered within 1 y after the fallout. An illustrative comparison with the corresponding time-pattern of the Chernobyl fallout in Russia can made, assuming an ecological half-time of ^{137}Cs of 10 y (Fig. 1(Right)). From Fig. 1(Right) it is seen that the long-term "tail" of the effective dose-rate is an order of magnitude larger for the nuclear power-plant fallout than for the spallation-source term by SSM [4]. From a radiometrical point of view, this means that the detectability of gamma-ray spectrometry *in-situ* from ESS fallout will be more challenging over long-term than what is the case when monitoring ^{137}Cs levels from a nuclear fallout event.

Radionuclide specific contribution to 50 y cumulative dose

Concerning the nuclide contributions to cumulative dose E(t), it appears that over a 50 y range ^{172}Hf will be the most significant contributor to the cumulative effective dose by means of its daughter product ^{172}Lu, followed by ^{178n}Hf and then ^{182}Ta, respectively. Even though the nuclide-specific relative contribution to the 50-y effective dose will be dependent on the initial ground penetration (wet or dry deposition event), the order of importance of the radionuclides appears to be the same given the simplistic assumption, applied here, of uniform *Eco*(t)-functions for all elements. When using an alternative source-term based on FLUKA simulations of spallation reactions in the W-target [6], the relative distributions presented in Fig. 2c and d are obtained. The relative contribution to the E(t = 50 y) from build-up of ^{172}Lu is then even larger than for the SSM source term.

Figure 3 shows the nuclide-specific relative contribution to the effective dose-rate as a function of time for the SSM source term. Calculations of the development of the relative dose-contribution with time, show that ^{182}Ta is significant within a time-window of a few years, after which ^{172}Hf supported ^{172}Lu will dominate. Using the alternative source-term from Baurkaukas and Stenström [6], the relative contribution from ^{182}Ta will be significantly less than for the SSM source term. These results show that gamma-ray emitting radionuclides such as ^{172}Lu or ^{178n}Hf to be more relevant than ^{182}Ta as key-nuclei, to follow-up dose prognoses over the long term. Additional research is currently being conducted [17–19], to investigate the detectability of the gamma-ray lines of the most strongly contributing radionuclides during the time-window 0 to 1 y after a fallout. This ongoing research will shed further light on the necessary detection conditions for *in-situ* gamma-ray spectrometry, following a possible accident at ESS.

Variability in E(50-y) estimates

A preliminary evaluation of the variability in the results was performed by repeated calculations for a combination of settings, applying a range for $T_{eco,long,i} = 10 \pm 5$ y, $T_{eco,short,i} = 1 \pm 0.5$ y and $c_{short} = 0.5 \pm 0.25$. The estimated $E(50$ y) for dry deposition of the SSM source term then ranged from

Table 1. Source terms used for computation of external doses from ground deposition.

Radionuclide	SSM [4]		Barkauskas and Stenström [6]	
	Inventory	Normalized to ^{182}Ta	Inventory	Normalized to ^{182}Ta
Cd-109	3.8E+11	0.0514	1.65E+13	0.0314
Ce-139	8.8E+11	0.119	8.8E+13	0.167
Eu-147	1.2E+12	0.162	2.14E+14	0.406
Eu-147	1.2E+12	0.162	2.14E+14	0.406
Gd-146[a]	9.7E+11	0.131	2.07E+14	0.393
Gd-153	8.2E+11	0.111	1.32E+14	0.251
Hf-170	5.8E+12	0.784	5.61E+14	1.07
Hf-172	4.1E+12	0.554	6.8E+14	1.29
Hf-173	1.1E+13	1.49	1.04E+15	1.97
Hf-175	1.15E+13	1.55	1.52E+15	2.89
Hf-178n[a]	2.5E+11	0.0338	2.5E+13[b]	0.0475
Hf181	9.5E+11	0.128	3.11E+13	0.0590
I-120	5.8E+11	0.0784	2.67E+13	0.0508
I-121	6.9E+11	0.0932	4.29E+13	0.0815
I-122	6.5E+11	0.0878	4.64E+13	0.0881
I-123	8.4E+11	0.114	4.9E+13	0.0932
I-124	1.1E+11	0.0149	1.2E+12	0.00228
I-125	7.5E+11	0.101	5.65E+13	0.107
I-126	2.9E+10	0.00392	2.07E+11	0.000393
Lu-169	5.9E+12	0.797	5.2E+14	0.988
Lu-170	7.8E+12	1.054	6.43E+14	1.22
Lu-171	9.3E+12	1.26	7.52E+14	1.43
Lu-172[a]	5.2E+12	0.702	7.3E+14	1.39
Lu-173	6.7E+12	0.905	9.38E+14	1.78
Lu-177	0	0	5.96E+12	0.0113
Re-182	9.3E+11	0.126	8.33E+13	0.158
Re-184	6.8E+11	0.0919	5.95E+13	0.113
Re-186	1.1E+13	1.49	3.37E+12	0.00641
Re-188	8.3E+12	1.12	8.3E+14	1.58
Ta-173	8.4E+12	1.14	8.21E+14	1.56
Ta-174	8.9E+12	1.20	1.1E+15	2.08
Ta-175	1.3E+13	1.76	1.34E+15	2.55
Ta-176	1.6E+13	2.16	1.79E+15	3.41
Ta-177	2.2E+13	2.97	2.17E+15	4.12
Ta-179	2.2E+13	2.97	2.83E+15	5.38
Ta-180	7.2E+12	0.973	3.35E+14	0.637
Ta-182	7.4E+12	1	5.26E+14	1
Ta-183	1.1E+13	1.49	4.6E+14	0.874
Ta-184	4E+12	0.541	2.11E+14	0.401
Tb-149	8.1E+11	0.109	1.32E+14	0.252
Te-118	8.8E+11	0.119	2.43E+13	0.0462
Tm-166	3.6E+12	0.486	4.07E+14	0.774
Tm-167	4.3E+12	0.581	4.28E+14	0.814
W-177	1.3E+13	1.76	1.43E+15	2.72
W-178	2.2E+12	0.297	2.4E+15	4.57
W-181	8.6E+13	11.6	7.25E+15	13.8
W-185	2.6E+14	35.1	9.26E+15	17.6
W-187	6.7E+14	90.5	2.21E+16	42.1
Wb-183 m	1.2E+12	0.162	1.2E+14	0.228
Yb-166	3.4E+12	0.459	3.96E+14	0.752
Yb-169	7E+12	0.946	5.42E+14	1.03

The source terms are taken from the cited references. The SSM-source term refers to assumed dispersed radionuclides from the damaged tungsten source whereas the source term from [6] refers to the total radionuclide inventory in the tungsten source after 5 y operation. Since all doses are normalized to the deposited ^{182}Ta (Eq. 1), it will be the relative abundance with respect to ^{182}Ta that influences the estimates. [a]Build-up of daughter nuclide has been accounted for in the dose calculations. [b]Added activity in the same proportion to ^{182}Ta as for the SSM-source term [4].

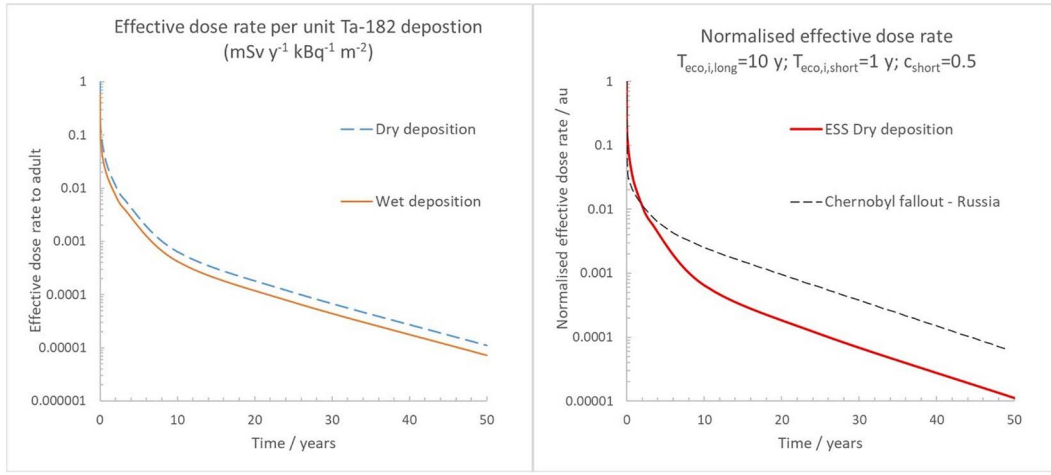

Figure 1. Left: Normalized effective dose rate to time = 0, using *eco*(*t*)-settings of $T_{eco.long,i} = 10$ y and $T_{eco,short,i} = 1$ y, $c_{short,i} = 0.5$, for an initial ground deposition of zero (mimicking dry deposition) and 0.5 g cm^2 (mimicking an initial wet deposition event). Right: Comparative plot between a dry deposition of ESS accident release and a corresponding source term for the Chernobyl fallout in Russia [16], using the same *eco*(t)-settings.

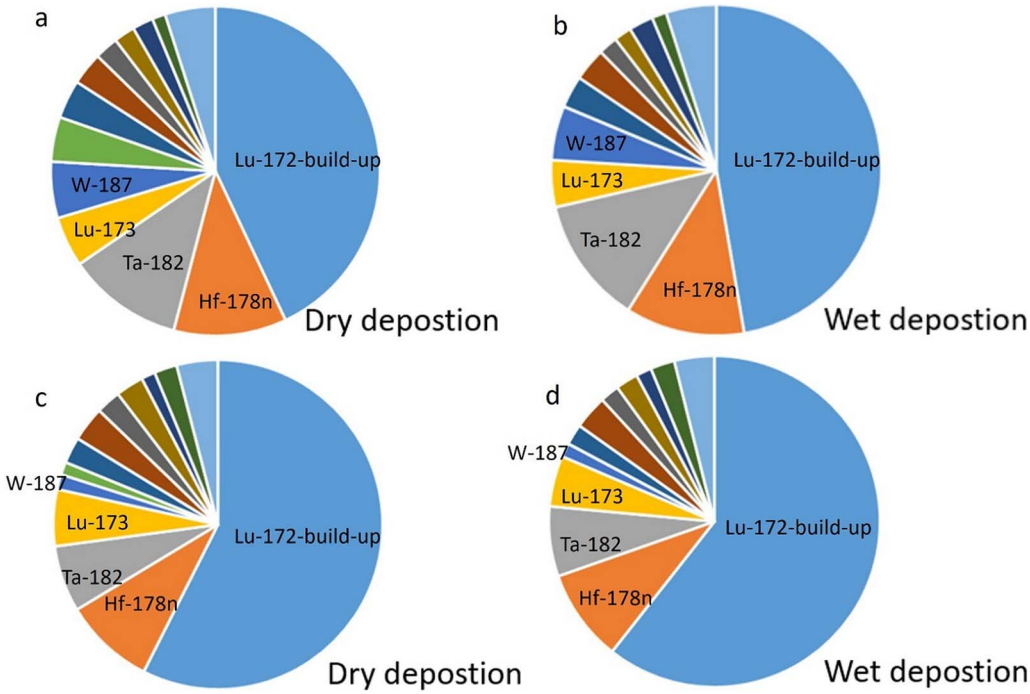

Figure 2. The nuclide specific relative contributions to the cumulative 50-y effective dose from ground deposition of an accident release of tungsten target particles calculated from the SSM source term for 5 y operation W-target for dry (a) and wet (b) deposition or the alternative source-term [6] for dry (c) and wet (d) deposition. Five most prominent radionuclides are marked in the pie-charts.

0.057 to 0.14 mSv/(kBq m^{-2}^{182}Ta). The variation in the c_{short} will account for about 75% of the variance decomposition. However, if also considering the influence of the initial ground penetration as well as the alternative source term, the E(50 y) may be as high as 0.30 mSv/(kBq m^{-2}^{182}Ta) for a dry deposited fallout. A preliminary variogram (Fig. 4) indicates that half of the variability is on account of which source term is used and a third is attributed to the initial ground penetration depth (Fig. 4). The reason for the

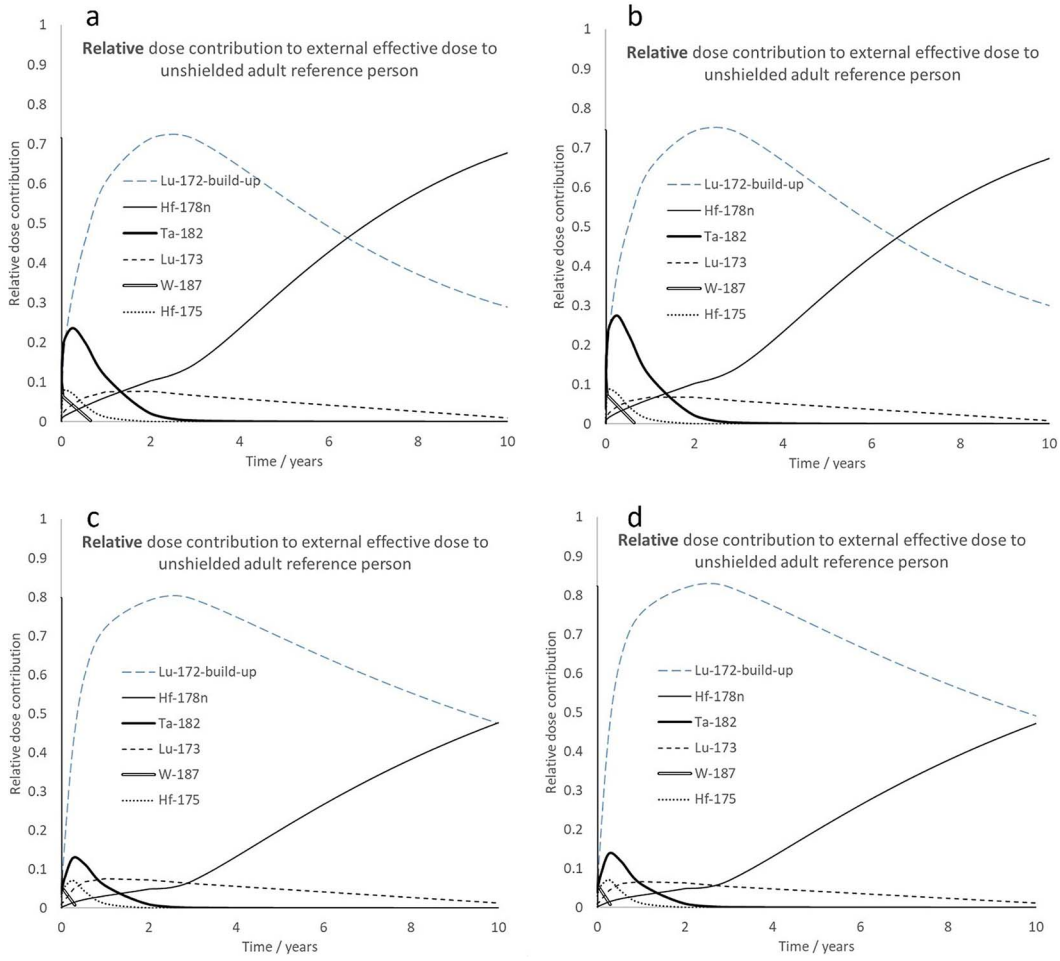

Figure 3. The nuclide specific relative contribution to the effective dose-rate from ground deposition of an accident release of tungsten target particles calculated using the SSM source term [4] for 5 y operation of the W-target for dry (a) and wet (b) deposition or the alternative source term [6] for dry (c) and wet (d) deposition.

large impact of the latter two factors on the E(50-y) estimates is attributed to the higher proportion of this dose being delivered so shortly after the fallout, in comparison to releases containing debris of nuclear power fuel. Albeit the considerable variability in the ESS target inventory, estimates based on the inventories used in this work point to ^{172}Hf/^{172}Lu being the most predominant external dose pathway from an accident release, in both cases.

Conclusions

Based on a hypothetical worst-case accident scenario at ESS, the effective dose per unit ground deposition of the spallation product ^{182}Ta has been calculated by using dose conversion factors retrieved from the ICRP [11] and simulated ESS target inventories [4, 6]. The objective has been to identify which of the released spallation-products will be the greatest contributor to external radiation exposure, both over short- and long-term (up to 50 y) after the accident scenario. The outcome of the calculations presented here also indicates which radionuclides will be the most important for radiometrically surveying the doses to members of the public by means of e.g. *in-situ* gamma-ray spectrometry. Although on-going studies are being conducted regarding the detectability of the released spallation-target products [17–21] the following preliminary conclusions can be drawn:

- The estimated 50-y effective dose, per unit of initial ^{182}Ta deposition, ranges from 0.082 to 0.23 mSv/kBq m^{-2} depending on which target inventory is used for the calculation and whether a dry- or wet-deposition event is assumed.

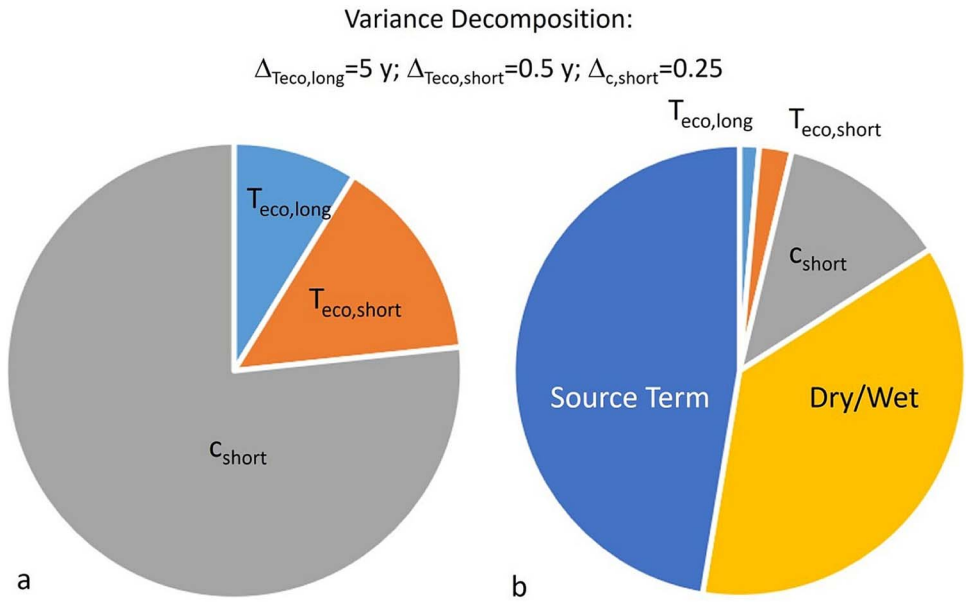

Figure 4. Variogram for E(50 y) for a dry deposition (0 initial soil penetration depth) scenario of the SSM source term [4] (a). Corresponding variogram when also accounting for two different source terms [4, 6] and initial ground penetration depths (dry = 0 g cm^{-2} and wet = 0.5 g cm^{-2}) (b).

- A significantly larger proportion of the 50-y cumulative external-dose, from fallout following an ESS accident, is distributed over the first 10 y, when compared to fallout from a nuclear power-plant accident. The effective dose rate 10 y after a worst-case ESS accident will be less than 1/1500 of the initial dose rate, whereas the corresponding ratio for the Chernobyl fallout in Sweden was ∼1/300.
- ^{172}Hf, together with the build-up of daughter ^{172}Lu, accounts for the highest portion (∼50%) of the 50-y time-integrated effective dose, followed by ^{178n}Hf (∼15%) and ^{182}Ta (∼10%). This result is consistent for both source terms studied.
- It is recommended based on the existing results that ^{172}Lu be used as a key nuclide for long-term dose surveillance, following an ESS accident, instead of the previously suggested ^{182}Ta. On-going studies, on simulated in-situ gamma-ray spectra, will show the time-window of detectability of these two radionuclides to fully test this hypothesis.

For a thorough understanding of the radiological consequences of major spallation-product release from ESS, additional studies are needed to investigate (i) how the ecological behavior of the spallation-products influences the long-term external doses and (ii) the potential variability of the ESS target inventory and the impact this variability has on the uncertainty in projecting accident exposures. Assessing the variability of the source term is also crucial for predicting the performance of existing methods for surveying the long-term external gamma-ray doses from an accidental release.

Author contributions

C. L. Rääf (Principal investigator, data generation and editor of the manuscript), R Frost (Data generation and analysis, review of manuscript), C Bernhardsson (Data generation and analysis, review of draft), G Pedehontaa-Hiaa (Data generation and analysis, review of draft).

Conflict of interest

None declared.

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