



Preliminary evaluations of the environmental impact for the production of ^{99}Mo by fusion neutrons

G. M. Contessa^a , M. D'Arienzo, M. Frisoni, P. Ferrari, R. Panichi, F. Moro, A. Pietropaolo

ENEA, Italian National Agency for New Technologies, Energy and Sustainable Economic Development, Lungotevere Thaon di Revel 76, 00196 Rome, Italy

Received: 10 January 2021 / Accepted: 9 April 2021
© The Author(s) 2021

Abstract ENEA is developing an accelerator-driven 14 MeV neutron source exploiting the deuterium–tritium fusion reaction to produce ^{99}Mo medical radioisotope as an alternative production route not based on fission reactors. It is expected that, during normal operation, a number of radionuclides, generated by means of neutron irradiation on the raw material (natural Molybdenum), will be produced and managed. The present manuscript, as foreseen by national law, discusses a hypothetical scenario to test the environmental screening models, in turn evaluating the mechanisms and parameters which affect and control the path of liquid effluents potentially released during normal operation of the facility. The aim is to estimate the amount of radioactivity to be operated and the fraction potentially discharged in this hypothetical scenario, so as to ensure that the radioactive material can be managed without any risk for the population and the environment, according to national regulations and thoroughly fulfilling the international guidelines.

1 Introduction

The production of ^{99}Mo medical radioisotope is strategic in nuclear medicine as it is the precursor of $^{99\text{m}}\text{Tc}$, the most used tracer in SPECT (Single Photon Emission Computed Tomography) nuclear diagnostics, covering more than 80% of all the procedures worldwide.

^{99}Mo is currently produced in nuclear fission research reactors by irradiating ^{235}U -containing samples. After the 2009 global ^{99}Mo crisis due to the use of aged fission reactors [1, 2], international organizations such as IAEA and OECD asked the scientific community to propose a series of alternative ^{99}Mo production routes not based on fission reactors and not using ^{235}U for non-proliferation issues. This would permit to integrate the irradiation capability of the present plants in the short and midterms, in turn providing suitable, feasible and cost-effective solutions in the long term [3, 4].

One of the proposed solutions relies on the use of 14 MeV neutrons from the deuterium–tritium fusion reaction ($\text{D} + \text{T} \rightarrow {}^4\text{He} + \text{n} + 17.6 \text{ MeV}$) and the inelastic channel ${}^{100}\text{Mo}(\text{n}, 2\text{n}){}^{99}\text{Mo}$.

^a e-mail: gianmarco.contessa@enea.it (corresponding author)

In this context, ENEA aims at developing an accelerator-driven 14 MeV neutron source to be used as irradiation facility for natural Molybdenum pellets, according to a previous study developed at the ENEA Frascati Research Centre [5].

Conceptually, the source is composed by a rotating target where a mixed beam of Deuterium and Tritium, delivering a power of about 250 kW, produces mostly 14 MeV neutrons with a neutron yield in the range $5\text{--}7 \times 10^{13} \text{ s}^{-1}$. These neutrons irradiate a sample of metallic natural Molybdenum, where ^{100}Mo has 10% abundance. First, calculations and projections from a dedicated study performed on a higher power source [6] indicate that the end of irradiation (EoI) activity of ^{99}Mo is in the range 2–5 Ci after 24 h continuous irradiation.

The irradiation plant will be connected to a radiochemical laboratory where the irradiated material is manipulated and chemically treated to produce a liquid solution named sodium Molybdate that is the precursor of the sodium Pertechnetate, *i.e.* the radiopharmaceutical used in the SPECT.

Dedicated experiments have already shown that the production of pure Pertechnetate, compliant to the present requirement of the European Pharmacopoeia, can be obtained starting from the $^{100}\text{Mo}(n,2n)^{99}\text{Mo}$ reaction, as described in Ref. [5].

In order to test the environmental screening models and evaluate the exposure of the population due to the potential liquid effluents of such a facility, as committed by national regulation for every activity carried out with radioactive materials, a case study is carried out taking into account a hypothetical scenario that foresees the discharge of the radioactive liquid effluents into the environment.

Directive 2013/59/Euratom [7], laying down basic safety standards for protection against the dangers arising from exposure to ionizing radiation, provides that “the disposal, recycling or reuse of radioactive materials arising from any authorized practice is subject to authorization”. However, this kind of materials may (the choice is left to the Member States) be released from regulatory control provided that the activity concentrations do not exceed the clearance levels set out in Table A of Annex VII, regarding solid materials, or comply with specific clearance levels established in national legislation following the general exemption and clearance criteria.

In particular, regarding radioactive liquid effluents, plans for the discharge have to be approved, and the competent authority can establish authorized limits as part of the discharge authorization and conditions for discharging radioactive effluents, which consider the results of the optimization of radiation protection. The discharge authorization can take into account the results of screening assessments relying on international guidelines. Deliberate dilution of the radioactive wastes for the purpose of disposal is not allowed.

In Italy, Directive 59 was implemented by Decree 101/2020 [8], which entered into force a few months before the draft of this article.

The Italian legislator has established that any activity involving disposal, recycling or reuse of materials containing radioactive substances in any form (solid, liquid, and gaseous) is subject to authorization.

Therefore, in Italy there is no exemption for disposal either for solid materials or for liquids or for effluents (liquids or gaseous), and any practice that provides for a disposal, of any extent, must be authorized. The authorization may establish specific requirements relating to the conditions for disposal or release, including restrictions on the total activity removed in a given time interval.

Furthermore, no material can be released into the environment, recycled or reused if the clearance levels, established on the basis of compliance with the radiological insignificance criterion and included in the authorization, are exceeded. Therefore, Decree 101/2020 always

requires, differently from Directive 59, the demonstration of compliance with the criterion¹ of radiological insignificance, which basically remains the annual individual dose of $10 \mu\text{Sv/y}$.

In particular, the disposal of liquid effluents into the environment is subject to compliance with an environmental discharge model in terms of activity released into the environment in a time interval that demonstrates compliance with the threshold of radiological insignificance (on an annual basis). The clearance levels are therefore not applicable to liquid and gaseous discharges, which are regulated in terms of maximum activity released annually or in predetermined periods, established with specific prescriptions (precisely the environmental discharge model) contained in the authorization. As in the Directive, deliberate dilution for disposal in the environment, transfer to third parties, or removal for recycling is not allowed.

In conclusion, in Italy it is necessary to verify the radiological insignificance of the liquid discharges to obtain the authorization, using models capable of assessing the impact of disposal into the environment.

Finally, Decree 101/2020 expressly states that the authorization, depending on the type or extent of the risk, includes the examination and approval of the proposed site of the facility, without prejudice to the provisions on environmental impact assessment. The preventive examination of the proposed site for new installations must be carried out in particular from the point of view of protection against undue exposures or contamination, which may affect the subsoil of the installation or even the surrounding environment, taking into account the environmental context, and therefore demographic, meteorological, geological, hydrological and ecological conditions. These requirements are in full compliance with the Directive 59, which introduces an increased attention to the protection of environmental matrices and regards environmental contamination not only as a route of exposure of the population affected by the discharges, but also as a threat to human health.

2 Materials and methods

2.1 Preliminary planning of the work activities of the irradiation plant

According to the work plan envisaged for the facility, every day about 10 kg of natural Mo are irradiated, corresponding to the production of about 5 Ci of ^{99}Mo .

The operational plan has still to be defined in detail, however, for the sake of simplicity, a working load of 2 full days per week has been taken into account, resulting in about 100 days a year of operations. During each irradiation session, a single sample is irradiated for 24 h, and then processed (and never used again); Mo isotopes are recovered from the effluents. Therefore, about 1000 kg of natural Mo per year are processed, and the liquids produced by the processing, not usable for the scope of the facility, can be obtained from the inventory of radionuclides 12 h after shutdown, *i.e.* after the time necessary for the chemical treatment in the radiochemical laboratory.

2.2 Monte Carlo calculations

Neutronic calculations were performed in order to assess the neutron spectra in the Molybdenum target. As the facility is at the planning stage, a preliminary design of the target was considered.

¹ The criterion of radiological insignificance (taken from Annex I of the Directive 96/29/EURATOM of 13 May 1996) establishes that a practice can be overlooked without further consideration if under any circumstances the effective dose released to any member of the public does not exceed $10 \mu\text{Sv/h}$.

The neutron transport calculations were performed by using the MCNP6.2 Monte Carlo code [9]. A preliminary CAD (Computer Aided Design) model of the Molybdenum container located around the neutron source has been converted into the equivalent MCNP geometrical description by means of the CAD-to-MCNP interface embedded in the SuperMC code [10].

In MCNP calculations, an isotropic mono-energetic neutron source of 14 MeV has been used with a $7.5 \times 10^{13} \text{ s}^{-1}$ yield. The neutron source has been modeled as a 10 cm diameter circle, representing the area where the D^+/T^+ beam impinges on the rotating target.

The exact configuration of the Molybdenum target is still under development, so a simplified model has been used, which, in any case, guarantees a reasonably realistic estimate of the activity produced at the EoI. The Mo target was divided in four layers, and in each one the average neutron spectrum was evaluated in the 175 energy groups structure (VITAMIN-J).

The neutron-induced cross section data files used in the transport calculations have been taken from the Fusion Evaluated Nuclear Data Library (FENDL) version 3.1d [11].

2.3 Evaluation of the radiochemical composition of liquid effluents

The neutron spectra calculated by means of the Monte Carlo simulations have been provided as input to assess the inventory of radionuclides obtained after 24 h of irradiation of 10 kg of natural Mo.

An activation calculation has been performed by means of the FISPACT-II code [12] coupled with the EAF-2010 activation nuclear data libraries [13], using as input the neutron spectrum of the most irradiated zone of the Mo target (total flux: $1.31 \cdot 10^{11} \text{ neutrons cm}^{-2} \text{ s}^{-1}$). This assumption is conservative since the considered spectrum, evaluated in the area closer to the neutron source, has been arbitrarily selected as representative for the whole Molybdenum mass: taking into account the uncertainties due to the ongoing design stage of the machine, this approach has been chosen in order to avoid any possible underestimation of the radionuclide inventory.

The activation calculation was performed considering a continuous irradiation scenario of 1 day. Table 1 shows the inventory after 12 additional hours of processing of the irradiated sample in the radiochemical laboratory.

2.4 Examination of a hypothetical site of the facility

To verify the impact on the environment, a worst-case scenario is analyzed: all the liquids after radiochemical processing are discharged without any filtering or preliminary treatment. The results obtained will certainly be an overestimation and, probably, a system of temporary containment and/or treatment will be realized.

In the hypothetical scenario considered for this case study, the liquid effluents from the radiochemical processing of the irradiated molybdenum are discharged into a generic lake named lake A (at a given altitude), which is connected by a reversible hydroelectric plant to a second lake B (at a lower altitude, *e.g.* half of the height of lake A), as it is shown in Fig. 1.

During the day, water flows from lake A to lake B for the production of energy, while overnight water is pumped back to A from lake B through some penstocks, thereby producing mixing of water contained in the two basins.

Just to have some reference parameters in order to carry out the calculations in this hypothetical reference scenario, the characteristics of a system of lakes typical of the Italian Apennines are taken into account: the power of the pumps of about 150 MW each, and a pumping rate of about $72 \text{ m}^3/\text{s}$ of water toward lake A, which is therefore able to fill up in 6 h

Table 1 Inventory of radionuclides relevant from a radiation protection point of view after processing

Radionuclide	Inventory (Bq/y)	λ (1/s)	$T_{1/2}$ (s)
H-3	2.37×10^6	1.78×10^{-9}	3.89×10^8
Sr-89	3.57×10^2	1.59×10^{-7}	4.37×10^6
Sr-90	6.19×10^{-1}	7.63×10^{-10}	9.09×10^8
Y-88	4.44×10^5	7.52×10^{-8}	9.21×10^6
Y-90	2.22×10^5	3.01×10^{-6}	2.30×10^5
Y-90 m	7.86×10^3	6.04×10^{-5}	1.15×10^4
Y-91	1.42×10^4	1.37×10^{-7}	5.06×10^6
Zr-88	4.24×10^7	9.67×10^{-8}	7.17×10^6
Zr-89	3.30×10^{11}	2.46×10^{-6}	2.82×10^5
Zr-93	1.11×10^3	1.44×10^{-14}	4.83×10^{13}
Zr-95	7.96×10^9	1.25×10^{-7}	5.53×10^6
Zr-97	5.40×10^{10}	1.15×10^{-5}	6.03×10^4
Nb-90	4.39×10^4	1.32×10^{-5}	5.25×10^4
Nb-91	1.87×10^8	3.23×10^{-11}	2.15×10^{10}
Nb-91 m	1.38×10^{11}	1.32×10^{-7}	5.26×10^6
Nb-92	3.09×10^2	6.27×10^{-16}	1.11×10^{15}
Nb-92 m	4.21×10^{11}	7.90×10^{-7}	8.77×10^5
Nb-93 m	2.74×10^7	1.36×10^{-9}	5.09×10^8
Nb-94	3.19×10^5	1.10×10^{-12}	6.31×10^{11}
Nb-95	6.16×10^{10}	2.29×10^{-7}	3.02×10^6
Nb-95 m	1.10×10^{11}	2.22×10^{-6}	3.12×10^5
Nb-96	6.77×10^{11}	8.25×10^{-6}	8.41×10^4
Nb-97	5.90×10^{10}	1.60×10^{-4}	4.33×10^3
Nb-98 m	1.75×10^7	2.25×10^{-4}	3.08×10^3
Tc-98	1.48×10^{-5}	5.11×10^{-15}	1.36×10^{14}
Tc-99	1.14×10^5	1.03×10^{-13}	6.75×10^{12}
Tc-99 m	1.37×10^{13}	3.20×10^{-5}	2.16×10^4

with the plant at full operation. The average flow rate of the penstocks over 12 h is therefore $36 \text{ m}^3/\text{s}$.

Let us suppose that lake B is mainly fed by the penstocks connecting it to a lake C with a flow rate of $38 \text{ m}^3/\text{s}$, and itself feeds the hydroelectric power station through penstocks with a flow rate of $42 \text{ m}^3/\text{s}$.

Moreover, let us suppose that lake A has generic dimensions of a small lake (0.55 km^2 ; $6.6 \times 10^6 \text{ m}^3$) and is much smaller than lake B (1.5 km^2 ; $46 \times 10^6 \text{ m}^3$), and both lakes supply drinking water to the neighboring area.

In order to estimate the doses to the members of the public due to the disposal of radioactive materials in the environment, and verify the compliance with the criterion of radiological insignificance of $10 \mu\text{Sv}/\text{y}$ per person, it is necessary to identify the “representative person”, *i.e.* the individual receiving a dose that is representative of the more highly exposed individuals in the population.

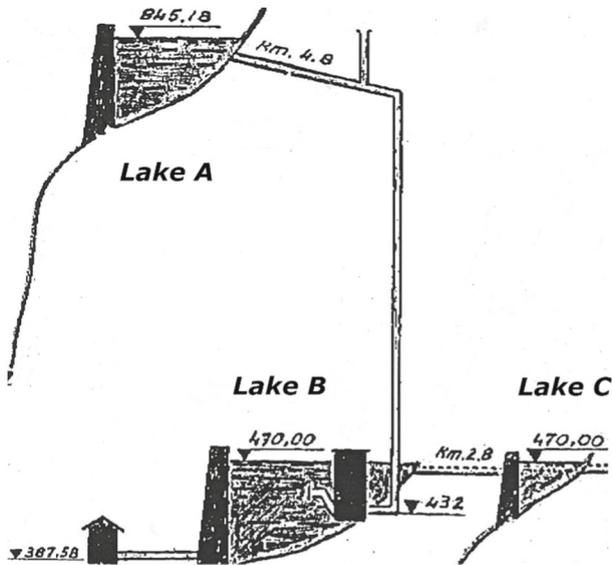


Fig. 1 scheme of the lakes in the scenario considered

In the case study under consideration in this hypothetical scenario, the employee of the facility is chosen as the “representative person” in the population, as the workers are supposed to drink water taken directly from the nearby lake A and then they are the nearest potentially exposed individuals living along the lake A.

Let us suppose that the discharge point of the liquid effluents into the lake A, for which no containment system is currently planned, is several hundreds of meters away from the drinking water collection point and is placed at a greater depth, as usual for this kind of plants. This way, the risk of direct withdrawal of the discharged radioactive effluents can be excluded and the concentration of the contaminant in the water of the lake is assessed through the physical models described below, in order to estimate the dose from ingestion of drinking water, which has been chosen as the effective pathway for transmission of the radioactive substances.

In this model, other pathways of transfer of radionuclides are considered negligible, such as ingestion of vegetables, milk and meat, as the water of the lakes A and B are supposed to be used for irrigation of the surrounding lands only in the summer months, as usually happens.

To support this hypothesis, a calculation of food concentration from contaminated water has been conducted for ^{99m}Tc , which is the radionuclide with the highest concentration in the lake A. Using the model proposed by IAEA [14], the calculated annual dose due to ingestion of contaminated food is of the same order of magnitude as the one due to ingestion of drinking water, yet the former takes into account more conservative assumptions.

2.5 The physical models for radionuclide transport in surface waters

In order to assess the impact of discharges, the environmental screening methodology of IAEA [14] and NCRP [15] is used, that takes into account dilution and dispersion of discharges into the environment, modelling the transport of radioactive materials in surface waters. For this

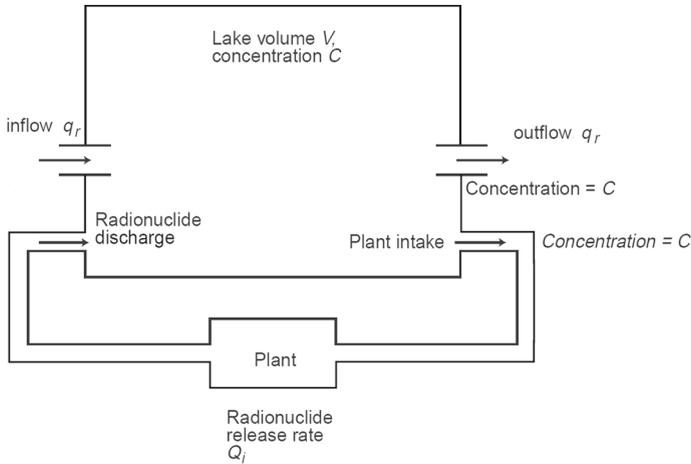


Fig. 2 Scheme of the assumed conditions of release into a small lake [14]

case study, as the surface area of lake A is $0.55 \text{ km}^2 \ll 400 \text{ km}^2$, the model of a small lake or reservoir is exploited (Fig. 2), assuming that radionuclides in water, under the conditions of a routine, long-term release, can be considered to be in equilibrium.

Assuming that radionuclides concentration in lake A is zero at the time of the start of the activities of the facility, the radionuclide equilibrium activity concentration for any generic radionuclide i is given by:

$$C_i = \frac{Q_i}{q_r + \lambda_i V}$$

where: C_i (Bq/m^3) is the activity concentration in lake A of radionuclide i ; Q_i (Bq/s) is the routine discharge into lake A for radionuclide i ; q_r (m^3/s) is the average flow rate into and out of lake A through the penstocks connecting it to lake B; λ_i (s^{-1}) is the decay constant of radionuclide i ; V (m^3) is the lake volume.

The radionuclides activity concentration can be considered not time dependent considering that, as this method aims to assess the potential long-term radiological impact of the operation of the facility over 30 years of lifetime, for all radionuclides involved the following holds:

$$\left(\frac{q_r}{V} + \lambda_i\right) > 10^{-8} \text{ s}^{-1}$$

Regarding lake A, since there is no radionuclide contribution to the lake from its watershed, the deposition rate from the atmosphere is zero.

The calculated concentration of radioactive contaminant in water is used to estimate the ingestion annual dose for adults over the assumed operational period of the facility of 30 years:

$$E_{\text{ing}} = C_i \cdot H \cdot DF_{\text{ing}}$$

where: E_{ing} is the annual effective dose from consumption of nuclide i (Sv/y); C_i is the concentration of radionuclide i at the time of consumption (Bq/m^3); H is the default consumption rate of water, equal to $0.6 \text{ m}^3/\text{y}$ [14] (assuming that the “representative person” drinks the water of lake A even during non-working hours); DF_{ing} is the dose coefficient for ingestion of radionuclide i (Sv/Bq) [14].

A more complex and detailed mathematical mixing model has been developed to simulate the water exchanges between the basin of lake A, where the radioactive contaminant is directly discharged in this hypothetical scenario, and that of lake B. This model is described in detail in another paper [16]. It considers also the timing of the liquid discharges from the facility and water exchanges between the two basins, and then it can evaluate the amount of radioactivity in lakes A and B at any time t , by means of a system of differential equations. It has been used to estimate the internal dose to the employee of the facility and to an individual of the population of the surrounding area following the intake of the radioactive contaminants, allowing to check if the previous choice of the employee as “representative person” in the population is correct. As mentioned, it takes into account also for the contribution to the concentration of contaminant in lake A due to the return flow from lake B (and vice versa), and, given the long half-lives of the radionuclides involved, it is useful to analyze the time-course of the effective dose from intake.

The considered (conservative yet plausible) release scenario consists in 100 discharges of radionuclides on annual basis, *i.e.* approximately once every three days, assuming 60-day experiment shutdown over one year of operation. In the model, the three-day cycle is assumed to consist in 12-h phases during which water flows alternatively from lake A to lake B and vice versa. The release of radionuclides is assumed to take place at the beginning of each cycle, from 8.00 p.m. to 9.00 p.m. The mixing model provides the activity concentration in the two lakes at any time t . For more details on the proposed model we refer the reader to ref. [16].

3 Results and discussion

3.1 Analysis of the radiological impact of liquid discharges

The effective dose to the representative person due to the liquid radioactive discharges of the facility, according to the model proposed by IAEA [14], is preliminarily evaluated to be 0.6 $\mu\text{Sv/y}$. This value is calculated considering the employee of the facility as the representative person of the population and ingestion of drinking water as the effective pathway for transmission of the radioactive substances, and disregarding from the effluents all Mo isotopes, which are recovered during the processing.

Then, according to this model, the amount of radioactivity released into the environment by the facility considered in this hypothetical scenario complies with the threshold of radiological insignificance (on an annual basis), and therefore the practice would result to be compliant with the requirements of the Italian Decree 101/2020 for the purpose of obtaining the license to practice.

The most relevant radionuclides for exposure purposes are ^{89}Zr , $^{92\text{m}}\text{Nb}$ and ^{96}Nb , which alone are responsible for almost 80% of the effective dose to the representative person.

The application of the customized mathematical model makes it possible to study the course of the dose rate over time due to ingestion of drinking water of both lakes.

Figure 3 shows the time course of the dose rate to an employee of the facility for the first 400 h. Again, the most relevant radionuclides from the radiation protection point of view are ^{89}Zr , $^{92\text{m}}\text{Nb}$ and ^{96}Nb , due to their high concentration in the discharges and their longer half-life.

After 400 h of operation, the activity concentrations of ^{89}Zr , $^{92\text{m}}\text{Nb}$ and ^{96}Nb in the lake A increase up to 0.59 Bq/L, 1.6 Bq/L and 0.46 Bq/L, respectively [16]. $^{91\text{m}}\text{Nb}$, which contributes to an increase in the activity concentration over time, is less relevant from the

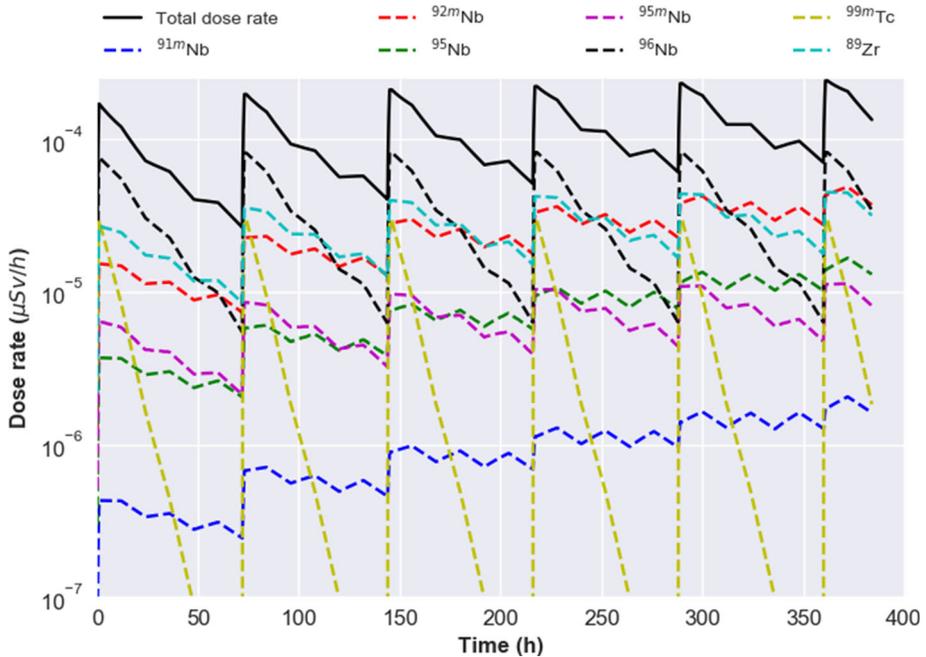


Fig. 3 Time course of the dose rate to an employee of the facility due to ingestion of drinking water from nearby lake A (log scale). The contribution is also shown of the most relevant radionuclides from the radiation protection point of view

radiation protection point of view because of its lower dose coefficient with respect to ^{89}Zr , $^{92\text{m}}\text{Nb}$ and ^{96}Nb .

According to this approach, the annual effective dose per person is about $2.5 \mu\text{Sv}$, using the same assumptions for the representative person of the population and the same inventory of radionuclides.

In lake B, the total radioactivity concentration would increase over time due to the continuous inflow of concentrated contaminant from lake A; for the same radionuclides discussed above, after 400 h of operation the activity concentration in lake B is 0.068 Bq/L (^{89}Zr), 0.26 Bq/L ($^{92\text{m}}\text{Nb}$) and 0.024 Bq/L (^{96}Nb). The dose rate to an individual of the population in the nearby area due to ingestion of drinking water from the lake would increase accordingly (Fig. 4 shows the trend over time for the first 400 h). The growth rate decreases over time because of the physical decay of the radionuclides.

Anyway, the dose rate in the case of lake B never reaches that related to lake A and is an order of magnitude lower, thus confirming the choice of the employee of the facility as the representative person in the population (Fig. 5).

In order to estimate the vulnerability of the activities carried out in the facility to extreme weather events [17, 18], the radiological impact of the discharges in the case of drought is assessed, considering the lowest possible water level in lake A. Under these assumptions, considering a volume of the lake A of about $3 \times 10^6 \text{ m}^3$, and according to the methodology of IAEA, the effective dose to the employee of the facility by drinking water directly from the lake is evaluated equal to $0.9 \mu\text{Sv/y}$. Then, even in an exceptional case of long-lasting drought, the dose to the representative person is lower than $10 \mu\text{Sv/y}$.

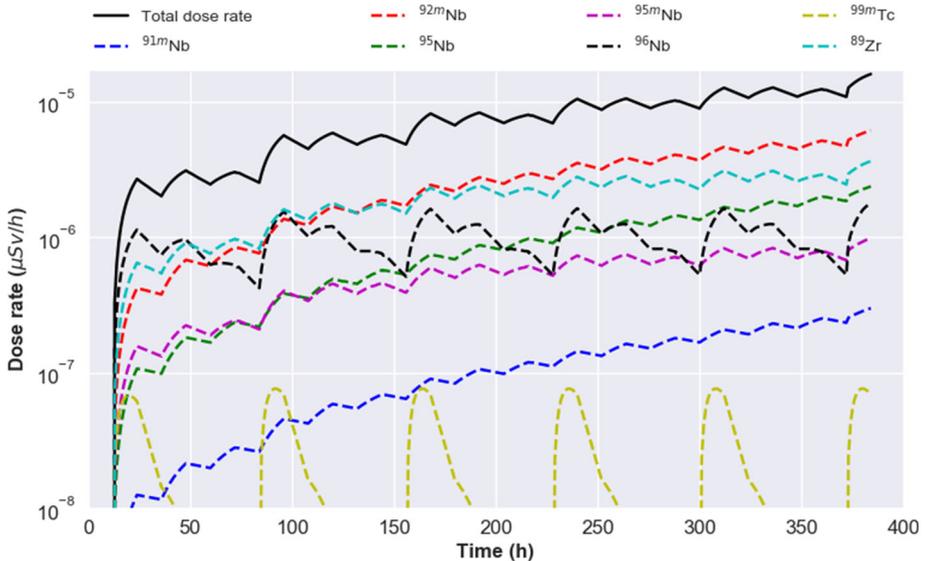


Fig. 4 Time course of the dose rate due to ingestion of drinking water from lake B (log scale). The contribution is also shown of the most relevant radionuclides from the radiation protection point of view

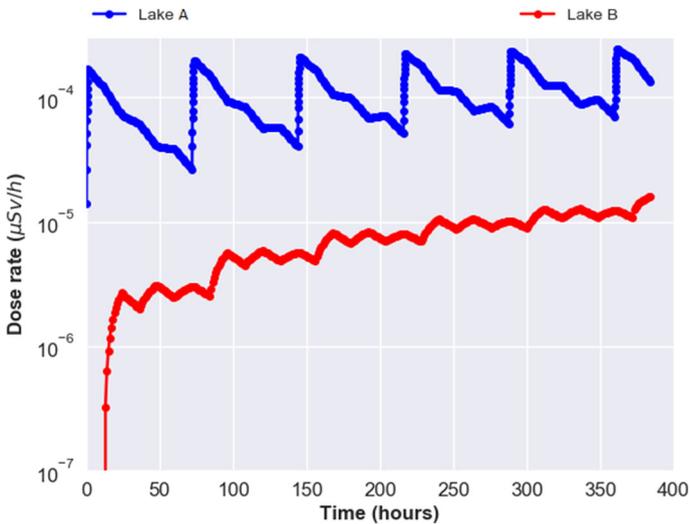


Fig. 5 Dose rate due to ingestion of drinking water from lake A vs dose rate due to ingestion of drinking water from lake B (log scale)

4 Conclusions

The European Directive 2013/59 and the Italian Decree 101/2020, which is its transposition, state that the disposal of radioactive materials arising from any radiological practice is subject to authorization [7, 8]. Therefore, the impact of the liquid discharges of the ENEA's fusion facility developed on the basis of a previous study [5], which is in the planning stage, has

been evaluated in a hypothetical scenario using models of disposal into the environment, according to internationally recognized guidelines.

By means of the environmental screening methodology of IAEA [14], the effective dose to the employee of the facility, chosen as the “representative person” in the population, is assessed equal to $0.6 \mu\text{Sv/y}$, lower than $10 \mu\text{Sv/y}$, as requested by the Italian law [8].

All environmental dose assessment models are subject to some degree of uncertainty, however the doses evaluated using these screening procedures should not underestimate the real doses by more than one order of magnitude under any circumstances [14, 15], thus ensuring compliance with the $10 \mu\text{Sv/y}$ threshold in the case study under consideration. On the contrary, they are generally considered likely to overestimate received doses [14].

Anyway, uncertainty in these methodologies could arise from departures of more complex situations from the generic assumptions taken into consideration.

Therefore, a mathematical model has been developed ad hoc to describe the specificity of the water exchanges between two different but connected small lakes, in only one of which the liquid effluents are discharged [16]. According to this more complex approach, the effective dose to the representative person is equal to $2.5 \mu\text{Sv/y}$.

Probably due both to the complexity of this second water system taken into consideration and the relative water flows between the two basins and to the long half-lives of the involved radionuclides, this case study shows that the screening procedures of international guidelines are highly effective but may not always be conservative. However, the difference between the results of the two approaches is within the uncertainty associated with the procedures proposed by these guidelines. More precise evaluations will be carried out in the future to confirm this difference.

Regarding the organization of the activities according to the radiation protection criteria, in the case of liquid effluents the Italian law is different from Directive 59/2013, which provides that the discharge authorization and conditions take into account the results of the optimization of radiation protection [7]. The fundamental “criterion of radiological insignificance” of Italian Decree 101/2020 [8] remains in fact that of $10 \mu\text{Sv}$ as an annual individual dose, a constraint that makes the application of the optimization principle impracticable [19].

However, 3–4 weeks of plant shutdown per year could be planned, for instance to carry out maintenance operations, so as to reduce the dose contribution of long half-life radionuclides. Moreover, if the activities of the facility should involve a workload higher than the preliminary one considered in this article, the opportunity of a system of temporary delay tanks mainly for ^{89}Zr , $^{92\text{m}}\text{Nb}$ and ^{96}Nb can be foreseen.

In any case, the theoretical methodologies applied to model the discharge of effluents consider that there is complete mixing of the radionuclides in the lakes, which, for instance, is unlikely to occur near the release point. Actually, for small lakes, like the ones considered in this case study, radionuclides concentration can be considered uniform within the entire impoundment [15].

The radionuclide with the highest concentration in the discharges produced by the processing of ^{99}Mo is $^{99\text{m}}\text{Tc}$: in the hypothetical scenario considered in this case study, every year about 10 TBq of this radionuclide are discharged in a lake. For reference, this amount is just two orders of magnitude greater than the activity discharged in the same time interval by a Nuclear Medicine Diagnostic Department [20]. Therefore, the resulting concentration in a lake whose extension is like that of lake A in the presented scenario is of the order of 1 Bq/L. The concentration of the other most relevant radionuclides (^{89}Zr , $^{92\text{m}}\text{Nb}$ and ^{96}Nb) is of the order of hundreds of mBq/L.

Hence, an environmental monitoring of the level of radioactivity both at a preliminary stage and during normal operations is essential, in order to assess the level of dose rates and

to implement specific operational and radiation protection choices, taking into consideration the different contributions of the various isotopes. Among the obligations of the employer, the Italian law [8] also provides for the assessment of radioactive contamination, with indication of the nature, physical and chemical state of the radioactive materials, as well as their concentration in environmental matrices.

Acknowledgements The authors warmly thank Dr A. Dodaro (ENEA), Dr S.A. De Crescenzo (A.S.S.T. Papa Giovanni XXIII) and Dr P. Rossi (INAIL) for their insightful suggestions.

Funding Open access funding provided by Ente per le Nuove Tecnologie, l'Energia e l'Ambiente within the CRUI-CARE Agreement.

Availability of data and material The data that support the findings of this study are available from the corresponding author (G.M.C.), upon reasonable request.

Declarations

Conflict of interest The author declare that they have no conflict of interest.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

1. P. Gould, *Nature* **460**, 312 (2009)
2. R. Van Noorden, *Nature* **504**, 202 (2013)
3. International Atomic Energy Agency. Non-HEU Production Technologies for Molybdenum-99 and Technetium-99m; IAEA Nuclear Energy Series No. NF-T-5.4; Technical Reports; IAEA: Vienna, Austria (2013)
4. OECD Nuclear Energy Agency, *NEA-OECD, Report on the Supply of Medical Radioisotopes: An Assessment of Long-Term Global Demand for Technetium-99m* (OECD, Paris, France, 2011)
5. M. Capogni, A. Pietropaolo, L. Quintieri, M. Angelone, A. Boschi, M. Capone, N. Cherubini, P. De Felice, A. Dodaro, A. Duatti, A. Fazio, S. Loreti, P. Martini, G. Pagano, M. Pasquali, M. Pillon, L. Uccelli, A. Pizzuto, *Molecules* **23**(8), 1872 (2018)
6. M. Capogni, A. Pietropaolo, L. Quintieri, ^{99m}Tc production via $^{100}\text{Mo}(n,2n)^{99}\text{Mo}$ using 14 MeV neutrons from a D-T neutron source: discussion for a scientific case. RT/2016/32/ENEA; ENEA: Rome, Italy (2016) <http://hdl.handle.net/10840/8208>. Accessed on 28 May 2018
7. European Council Directive 2013/59/Euratom on basic safety standards for protection against the dangers arising from exposure to ionising radiation and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom and 2003/122/Euratom. OJ of the EU. L13; 57: 1–73 (2014)
8. Legislative Decree 31 July 2020 No. 101 implementing Directive 2013/59/Euratom – “Attuazione della direttiva 2013/59/Euratom, che stabilisce norme fondamentali di sicurezza relative alla protezione contro i pericoli derivanti dall'esposizione alle radiazioni ionizzanti, e che abroga le direttive 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom e 2003/122/Euratom e riordino della normativa di settore in attuazione dell'articolo 20, comma 1, lettera a), della legge 4 ottobre 2019, n. 117”. G.U. n. 201 del 12/08/2020, Supplemento ordinario n. 29/L
9. MCNP User's Manual - Code Version 6.2, LA-UR-17-29981, (2017)

10. Y. Wu, FDS Team, Fusion Eng. Des. **84**(7–11), 1987–1992 (2009)
11. Fusion Evaluated Nuclear Data Library FENDL-3.1d, IAEA website. <https://wwwnds.iaea.org/fendl/index.html>
12. The FISPACT-II User Manual, UKAEA-R(18)001 February 2018 Editors: M. Fleming, T. Stainer, M. Gilbert
13. J.-C. Sublet, L.W. Packer, J. Kopecky, R. A. Forrest, A. J. Koning and D. A. Rochman, The European Activation File: EAF-2010 neutron-induced cross section library, EASY Documentation Series CCFE-R (10) 05
14. INTERNATIONAL ATOMIC ENERGY AGENCY, Generic Models for Use in Assessing the Impact of Discharges of Radioactive Substances to the Environment, Safety Reports Series No. 19, IAEA, Vienna (2001)
15. NCRP Report No. 123. Screening Models for Releases of Radionuclides to Atmosphere, Surface Water, and Ground - Vol. I and Vol II. NCRP, Bethesda (1996)
16. M. D’Arienzo, G.M. Contessa, M. Frisoni, P. Ferrari, R. Panichi, F. Moro, A. Malizia, A. Pietropaolo, Eur. Phys. J. Plus (2021) *in publication in the Special Issue “New technologies for detection, protection, decontamination and developments of the decision support systems in case of CBRNe events”*
17. G.M. Contessa, C. Grandi, M. Scognamiglio, E. Genovese, S. Sandri, Ann. Ist. Super Sanità **52**(3), 386–396 (2016)
18. G.M. Contessa, M. Guardati, M. D’Arienzo, C. Poggi, S. Sandri, M.C. D’Auria, E. Genovese, V. Cannatà, Eur. Phys. J. Plus **133**, 380 (2018)
19. D. Giuffrida, Proceedings of the National Congress of the Italian Radiation Protection Association (AIRP) “Il nuovo sistema radiologico: situazioni di esposizione programmate, esistenti e di emergenza” (2013)
20. R. Barquero, M.M. Agulla, A. Ruiz, J. Environ. Radioact. **99**, 1535–1538 (2008)