Impact of medical radionuclide discharges on people and the environment

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ABSTRACT

We present a novel methodology to dynamic calculate dose rates to non-human biota from hospital-released radionuclides reaching the environment through waste water treatment plants (WWTPs), using the biokinetic model D-DAT for aquatic biota, applied to ^{18}F, ^{123}I, ^{131}I, ^{153}Sm, ^{99m}Tc and ^{201}Tl. We have also developed a method to calculate doses to WWTP workers and farmers from agricultural practices. This proof-of-concept study uses a calculated generic source term of radionuclide levels in the Belgian Molse Nete River during the year 2018. The dose rates to non-human biota calculated for this scenario, which are calculated under conservative assumptions, are well below ERICA predicted no effects dose rate to wildlife of 10 µGy h^{-1}. For humans, the exposures are also very low, approaching a trivial value of the dose rate of 10 µSv y^{-1} in most cases. This work identifies important data gaps and areas of uncertainty in the assessment of radiopharmaceutical effluents. The study, which is part of the EC project SINFONIA, paves the way for a possible pan-European screening assessment methodology with the possibility to perform consistently assessments of the impact of radiopharmaceuticals on people and the environment. This is particularly relevant since discharges of radiopharmaceuticals in rivers are on the increase and it is necessary to explicitly demonstrate that people and the environment are adequately protected.

Keywords: Hospitals; Radiological impact assessment; Molse Nete; Radiopharmaceuticals; Wildlife; Water treatment plant.

1. INTRODUCTION

Radiopharmaceuticals are broadly used for diagnostic purposes, for treatment of cancer and other diseases. After their use in hospitals, radioactive effluents are collected in special tanks until acceptable activity levels are reached for their release into the sewer system. Then, they are conveyed through the sewer system to wastewater treatment plants (WWTPs) for further standard 24 - 48 h treatment, whereupon they are released into rivers. There they come in contact with aquatic life and people via ingestion and the external exposure pathways.

With the general increase of radiopharmaceutical use, including the approval of new treatments, releases to WWTPs and watercourses are on the increase. The radionuclides involved typically include short-lived γ-ray emitters used for imaging and diagnostics, such as ^{99}Tc (IAEA, 2009), but also longer-lived β-emitting radionuclides that are used in therapeutic treatments, such as ^{131}I (Veliscek Carolan et al., 2011), to which one must add a variety of novel isotopes (including α-emitters) arising from recently approved radiopharmaceuticals and novel techniques in nuclear medicine, which have experienced a remarkable increase in recent times (Vives i Batlle et al., 2022).

Radiological impact assessments for these environmental releases are seldom conducted and environmental parameter data for specific radionuclides are lacking. Some unusual sewer-WWTP-aquatic dispersion pathways are involved, and this leads to both over-conservatism and uncertainty in assessments. Assessment uncertainties are contributed to by the fact that environmental monitoring is not often performed, and there is a lack of environmental parameter values to represent the relevant uptake, bioaccumulation and dispersion pathways. Radiation doses to WWTP workers, the public and the environment are presumed to be low due to the short half-life of most of the radionuclides (Martínez et al., 2018), but this is not a given for all situations/radionuclides and there is a need to explicitly prove protection of people and the environment for a wider variety of radionuclides and assessment cases, including routine and accidental releases.

From the environmental perspective, the highest priority is to produce special models for dose assessment of radionuclide releases from hospitals to the environment via WWTPs, not only for impact on members of the public but also for wildlife. The reason to include wildlife is that the International Commission on Radiation Protection (ICRP) has established that, in addition to humans, the environment should similarly be protected from deleterious effects of radiation (ICRP, 2008, 2014, 2017). The goal to protect the environment is motivated by a significant evolution of thought based on both moral and scientific grounds, debunking the old statement that “if humans are protected, the environment is also protected”.

In Belgium, there is a lack of radiological impact assessment of current as well as future medical releases, and hospitals are not always forthcoming with information on their waste disposal practices. This is because licensing usually does not require hospitals to sample or do monitoring of the

environment and there is no notification procedure for outside releases. To address this, between 2012 and 2014, automatic measuring stations were installed by the Federal Agency for Nuclear Control in some of the treatment plants that receive wastewater containing radiopharmaceuticals (FANC, 2015). The measurements show that, in the influents at the inlet of the WWTPs, ^{99m}Tc and ^{131}I (which typically appear as discontinuous discharges, or "spikes") are often detected while ^{18}F, ^{123}I, ^{192}Ir and ^{153}Sm are occasionally detected or not depending of the WWTP (FANC, 2015). In many instances, the activity levels in the effluents were below the detection limit.

According to these measurements, the current releases do not represent a risk for people or the environment; notwithstanding this, they exemplify that hospital discharges should be the subject of monitoring and regular control. The data gives information not only about activity levels but also about the effluent's isotopic composition and the release schedule, providing an opportunity for making a pilot study and to identify the data gaps present in assessments of this type.

The present study aligns with the Radioecology ALLIANCE recommendations to EURAMED in the EC Rocc-n-Roll project (Vives i Batlle et al., 2022), which indicated the general research need to (a) identify the behaviour of relevant radionuclides and exposure pathways, (b) improve datasets and assessment methods, identifying the relevant data gaps and (b) provide advice to operators and regulators leading to a future European-level assessment approach. Following these recommendations, the objective of this project, performed within Work Package 3 of the EC project SINFONIA (https://cordis.europa.eu/project/id/945196), is to show how to assess the impact of environmental releases of radiopharmaceuticals from hospitals on the public and the environment, using the aforesaid Belgian test case as an example.

Our study involves estimating radionuclide uptake in freshwater wildlife at the outlet of a WWTP in Belgium and resulting uptake and internal/external exposures to aquatic wildlife for a conservative and generic discharge scenario that is used to test the assessment method and to identify knowledge gaps. The data on which this study is based are measurements previously performed at several WWTPs receiving discharges from hospitals across the country. For the human part of the assessment, we consider human ingestion of aquatic biota and drinking of contaminated water as well as external exposures at the riverbank and swimming, including also an indirect mechanism, namely internal doses arising from the consumption of agricultural foodstuffs after irrigation or fertilisation with contaminated sludge.

It is therefore not the purpose to assess the releases of a particular hospital, but to use this semi-realistic scenario to put to the test the different steps of the assessment chain and highlight where more data and process information are required, deriving recommendations for a the development of a more integrated and pan-European assessment system.

2. MATERIALS AND METHODS

2.1 Case study definition

Our case study is a fictitious situation with a WWTP in Mol releasing into the Molse Nete river in Belgium, but using a generic source term based on the river releases from the hospital of Leuven, given the lack of discharge data for the (smaller) hospitals in the Mol-Geel region of interest. The modelling of the impact of radiopharmaceuticals is based on a series of conservative assumptions, explained in turn below.

The first assumption is selecting the radionuclides at the inlet of WWTP Leuven as a generic test case to provide radionuclide data for the assessment. Our assumption is reasonable because this is the most important centre for cancer research and treatment in Belgium. Additionally, the proportions of this hospital with regard to the number of patients and treatments lead us to presume that the rest of the hospitals in the country could increase their releases in the future up to similar levels, given the growing demand for medical treatments of this nature. The activity concentrations at the inlet of WWTP Leuven are periodic and change over the day, so this source term captures the typical variability in the dilution of the activity concentration, making it an essentially dynamic problem.

Secondly, for the assessment of impact to the environment and members of the public, we made the penalising assumption that the hospital effluents from the decay tanks reaching the WWTP inlet are bypassed directly into the river. This could occur under extraordinary circumstances such as maintenance or expansion of the WWTP, so it is necessary to include it in our (conservative) assessment.

Thirdly, we assume that the aquatic biota and the members of the public are located directly at the outlet of the WWTP where the concentrations are maximal. The released radionuclides would spread downstream the release point, travelling across the river network which has a long hydrological simulation domain, but concentrations and the resulting dose rates would be much lower at increasing distances downstream from the WWTP.

In fourth place, we selected the low-flow year 2018 as reference year because the highest concentration in the river occurs during the periods where the river discharge is very low. In principle, it would be sufficient to just calculate the activity concentration in the river for the driest historical period, but we were also interested to investigate the fluctuations within a year to assess the variability of dose in relation to the accepted limits. Therefore, the release time series were cycled to have a registry for a full year. Using the available hydrometric data, we concluded that the year 2018 was one of the driest in the last decennia, especially in summer, leading to selecting this year as the test case for subsequent study. Specifically, we used the 7-day, 10-year low flow (Q7, 10) statistic as an indicator. The (Q7,10) is the 7-day minimum flow that is expected to occur every 10 years (Chapra, 1997). The calculation of the (Q7, 10) was done based on the last 35 years of flow registries. Our calculation shows that the summer

of 2018 was one of the driest summers in the last decennia, justifying the selection of 2018 as the representative year for this study. In addition, the number of low flows around the minimum flow observed in 2018 is higher in comparison to other years.

Lastly, we made the conservative assumption for workers of the WWTP in Mol that the plant is actively taking-up radionuclides from the water according to literature-based retention efficiencies, leading to exposures to plant operating and maintenance workers and doses from agricultural application of water and sludge. This is deliberately opposite to the assumption of free release for the non-human biota and members of the public.

The high conservatism of our assumptions means that the results must not be interpreted as an impact assessment of the UZ Leuven hospital as such, which would be much lower than calculated here, but rather as a generic, approximated benchmark simulation which, in so far as possible, is used to understand the assessment chain for radiological discharges from hospitals to rivers, and to give a conservative estimation of the potential impact of similar discharges in the Molse Nete region.

Our model simulations led to the calculation of activity concentrations of ^{18}F, ^{123}I, ^{131}I, ^{153}Sm, ^{99m}Tc and ^{201}Tl in 10-minute intervals taking into account their decay half-lives, their distribution between solid and liquid phases, the volumes and activity levels, the high discharge periodicity of the radioactive effluents and the river's flow regime. We refer to a previous report (Fiengo Pérez et al., 2022) for details on the hydrological simulations performed. In short, the model used for the dispersion simulation is DHI MIKE 11-ECO Lab framework. This model solves the full, dynamic, 1-D shallow-water equations in unidirectional form, also called Saint Venant equations (Hervouet, 2007). Direct verification of the model's hydrological predictions across the whole simulation domain could not be fully carried out with the data available to represent a highly complex system, but the model's capacity to simulate a complex river network has been verified separately during an ongoing model evaluation for tritium in collaboration with the IRSN (France) for the Rhône River (Fiengo Pérez et al., 2022). Nevertheless, our predictions of flow rate and activity concentrations in this exercise matched the limited data available.

We also calculated an accidental scenario involving the release into the sewer system of a 1-MBq ^{131}I therapeutic capsule as used for thyroid cancer treatment. The resulting doses can be scaled up or down to fit a range of possible radionuclide activities.

2.2 Basis of the impact assessment approach

The sequence followed to develop the assessment is as follows:

1. Devising a list of relevant radionuclides and creating a database of radioecological parameters for freshwater wildlife: expected chemical form, decay half-life, distribution coefficient (K_d), transfer factor and biological half-life information. We used mostly data from the ERICA assessment tool for wildlife impact assessment (Brown et al., 2008; Brown et al., 2013) and associated data collection

approaches (Beresford et al., 2015b) where data was not directly available, including extrapolation methods to fill in the data gaps.

2. Adaptation of the Dynamic Dose Assessment Tool (D-DAT) model for marine wildlife (Vives Batlle et al., 2008; Vives i Batlle, 2016) to cover the freshwater environment, by re-parameterising the model with information from the aforesaid database and introducing the relevant radionuclides. This task included also the incorporation of a new human exposures post-processor into D-DAT to calculate doses to people arising from the consumption of aquatic wildlife or exposure to the contaminated water.

3. Production of a simple model to calculate doses to waste treatment plant workers, sewer maintenance workers and the public drinking the water and eating from the terrestrial food chain. This model was grounded on a previous example of assessment of liquid pharmaceutical discharges in sewers (McDonnell, 2004; Titley et al., 2000) but with significant methodological and functional differences and customised with the relevant radionuclides and assessment parameters for the Belgian situation.

2.3 Database of radioecological parameters

The radionuclide parameters listed in the database cover ^{18}F, ^{89}Zr, ^{90}Y, ^{99}Mo, ^{99m}Tc, ^{123}I, ^{131}I, ^{131m}Xe, ^{133}Xe, ^{153}Sm, ^{177}Lu, ^{177m}Lu, ^{201}Tl , ^{223}Ra, ^{225}Ac, ^{226}Ra and ^{227}Th, although in practice this study covers only for ^{18}F, ^{123}I, ^{131}I, ^{153}Sm, ^{99m}Tc and ^{201}Tl because these are the only radionuclides detected in the Leuven WWTP. Data for the remaining radionuclides are kept in readiness for future studies.

This database contains the following information: radionuclide chemical form, half-life, the solid-liquid distribution coefficient K_d, and the concentration factor CF and biological half-lives of elimination $T_{B1/2}$ for multiple processes. This information, covering general radionuclide information and biological half-lives, is presented in Tables 1 and 2, respectively.

We conducted detailed reviews to obtain much of the data but, when not available, we used a simple analogy with closest chemical element to fill parameter gaps. Since CFs for ^{99}Mo and ^{177}Lu could not be found in the ERICA database, we used a conservative value from another transition element for ^{99}Mo and a lanthanide for ^{177}Lu: Tc was used as an analogue for Mo as it is the nearest transition metal in terms of closest atomic number.

For lutetium, the situation is more difficult; The ERICA Tool has data for La, Ce and Eu with La and Ce having high K_ds and Eu behaving more as a soluble element. It is stated that K_ds for lanthanides vary in the order Eu < Ho < Gd < Er < Dy < La (Tomczak et al., 2019); clearly it is better to take a high value for Lu from among the lanthanides, which signals that our most appropriate analogue for Lu and Sm radionuclides is Eu. In similar fashion, we used Cl data as an analogue for F, given the closeness of these elements in the periodic table. For the CF and K_d specifically, additional sources of information and data extrapolation included data from the ongoing new revision of the IAEA SRS-19 report (IAEA,

2001), and other sources describing Tc uptake experiments for crustaceans and molluscs, performed at Oregon State University (Hevland, 1981; McKenzie-Carter, 1985).

The thallium K_d is a sensitive parameter in our study, given the activities and retention times involved. Here, a single suitable source was found (Seaman and Kaplan, 2010). For the CR data gaps encountered for ^{201}Tl we used published measurement data (Zitko, 1975) for fish and we used Pb as an analogue for macroalgae. The principal source available for the biological half-lives is the freely available international database of radionuclide biological half-life values developed during the IAEA project MODARIA (https://www-ns.iaea.org/projects/modaria/modaria2.asp?s=8&l=129, which includes 1907 entries for 52 elements for terrestrial, freshwater, riparian and marine organisms (Beresford et al., 2015a). Additional biological half-lives were taken from the IAEA INIS database (https://www.iaea.org/resources/databases/inis), with some success for direct data for lanthanides and thorium. We also performed data gap extrapolation by finding the nearest radionuclide or biological analogues as described previously (these data are appropriately colour-coded in the database).

In some cases, our scientific judgement indicated that it was more adequate to use information from other sources not linked to the IAEA biological half-life database, and that additional literature was more suitable. Moreover, we used data for carp (*Cyprinus carpio*) and mosquitofish (*Gambusia affinis*) (Blaylock and Frank, 1981). For thorium in fish, we used additional publish data (Mahmood et al., 2014).

The extrapolation method indicated above, based on consideration of nearest chemical analogue, has the potential to be improved in future using chemical speciation knowledge. For example, iodate could be used to predict the long-term behaviour of ^{99}Tc, which is present in the aquatic environment mainly as the pertechnetate ion TcO_4^-.

Table 1: Database of radionuclide parameters and related information

RN	Relevance	Chemical (administered)	Chemical form (speciation) As administered	Environment	λ (d^{-1})	Kd (m^3 kg^{-1})	Concentration ratio (Bq kg^{-1} FW per Bq m^{-3}) Fish	Crust.	Bivalve	Aq. plant	Phytopl.	Zoopl.
^{89}Zr	Used in positron emission tomography (PET)	Radiolabeled monoclonal antibodies		Soluble	2.1E-01	1.7E+02	1.3E+00	8.2E-01	8.2E-01	9.7E-02	8.2E-01	1.5E+00
^{90}Y	Used in cancer radiotherapy	$^{90}YCl_3$ labelled particles injection	Y^{+3} (ionic)	Soluble	2.6E-01	4.0E+00	7.9E-02	2.3E+00	2.3E+00	3.8E-01	2.3E+00	6.9E+00
^{99}Mo	^{99m}Tc production	Not administered		Highly soluble	2.5E-01	2.6E-02	9.9E-02	9.9E-02	9.9E-02	9.9E-02	9.9E-02	9.9E-02
^{99m}Tc	Nuclear medicine diagnostics	$^{99m}TcO_4^-$ (VII), other III or IV reduced complexes	$^{99m}TcO_4^-$	Highly soluble	2.8E+00	2.6E-02	9.9E-02	9.9E-02	9.9E-02	9.9E-02	9.9E-02	9.9E-02
^{131}I	^{99}Mo byproduct, thyroid radiotherapy, diagnostic cameras	sodium iodide ($Na^{131}I$) and metaiodobenzyguanidine	I^-	Moderately insoluble	8.6E-02	1.1E+00	3.1E-01	8.0E-02	8.0E-02	5.3E-02	8.0E-02	5.3E-02
^{131m}Xe	By-product of ^{99}Mo production	Noble gas (not administered)	Free element	Gas	5.9E-02	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
^{133}Xe	By-product of ^{99}Mo production	Noble gas (not administered)	Free element	Gas	1.3E-01	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
^{177}Lu	Radiopharmaceutical percursor used for radiolabelling medicines	$^{177}LuCl_3$, Lutathera lutetium (177Lu)-oxodotreotide	Akaline, $Lu(OH)_3$	Highly soluble	1.0E-01	2.9E+02	6.2E-02	1.6E+00	1.6E+00	2.2E-01	1.6E+00	8.3E+00
^{177m}Lu	Production of ^{177}Lu from ^{177m}Lu	Not administered	Akaline, $Lu(OH)_3$	Highly soluble	4.3E-03	2.9E+02	6.2E-02	1.6E+00	1.6E+00	2.2E-01	1.6E+00	8.3E+00
^{223}Ra	Xofigo therapy with ^{223}Ra to treat bone tumours.	$^{223}RaCl_2$		Moderately soluble	6.1E-02	8.5E+00	1.0E+00	2.8E-01	5.2E+01	8.7E-01	5.2E+01	5.2E-01
^{225}Ac	Targeted α-particle cancer therapy	Free metal or chelating/ complexing agents		Highly insoluble	7.0E-02	2.0E+04	1.1E+00	3.4E+01	3.4E+01	4.4E+01	3.4E+01	1.2E+01
^{226}Ra	^{225}Ac production in LINAC by bombarding ^{226}Ra	Not administered	Insoluble	Moderately soluble	1.2E-06	8.5E+00	1.0E+00	2.8E-01	5.2E+01	8.7E-01	5.2E+01	5.2E-01
^{227}Th	Targeted thorium conjugates (TTC)	Attached to targeting proteins for tumor delivery		Highly insoluble	3.7E-02	2.7E+02	7.2E-01	1.7E+01	1.7E+01	4.4E+01	1.7E+01	1.2E+01
^{18}F	Tracer for PET.	NAF and fluorodeoxyglucose	F^-	F^- (soluble)	9.1E+00	1.0E-03	1.0E+00	1.0E+00	1.0E+00	2.8E-01	1.0E+00	2.8E-01
^{123}I	Single photon emission computed tomography (SPECT)	Supplied as NaI in 0.1 M sodium hydroxide solution	I^-	Moderately insoluble	1.3E+00	1.1E+00	3.1E-01	8.0E-02	8.0E-02	5.3E-02	8.0E-02	5.3E-02
^{153}Sm	Bone cancer palliation	Component of samarium lexidronam.	Chelated complex	Ca analog – bone-seeker	3.6E-01	2.9E+02	6.2E-02	1.6E+00	1.6E+00	2.2E-01	1.6E+00	8.3E+00
^{201}Tl	Myocardial perfusion imaging, SPECT for heart diagnosis	Thallous (I) chloride (TlCl) injection	Tl^+ (ionic)	Soluble. Tl(III) sorbs stronger	2.3E-01	1.7E+00	6.5E-01	9.8E-01	9.8E-01	4.7E-01	9.8E-01	8.6E-01

IAEA live chart of nuclides (https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html)
Using Tc as an analogue for Mo - nearest transition metal in terms of atomic number.
Data from new revision of IAEA SRS-19 report (IAEA, 2001).
Using Eu as an analogue for Lu and Sm as it maximises Kd for the three available candidates (La, Ce and Eu).
Assumption of Kd = 0 and CR = 0 for noble gases.
Ac transfer factors from the ERICA Tool (Brown et al., 2016), mostly extrapolated from Pu, Am and Th and some of the CRs are even based on data.
Y transfer factors from the latest ERICA version, originating from the IAEA wildlife transfer parameter database (https://www.wildlifetransferdatabase.org/).
Primary value from the ERICA tool with or without extrapolation.
From direct sources describing uptake experiments (Hevland, 1981; McKenzie-Carter, 1985).
Using Cl as an analogue for F.
Using Pb as analogue (data from the ERICA Tool).
Published data on thallium toxicity (Zitko, 1975).

Table 2: Database of biological half-lives). Note the analogies between the following pairs of elements: Ac and Ra, F and I, Sm, Lu and Eu. Values in red are extrapolated from a related species (plant to phytoplankton or crustacean to zooplankton) or radionuclide analogues (Y as analogue for Zr).

Rn	Biological half-life (d) – primary data						Short-term biological half-life with data extrapolation (d)						Long-term ibological half-life with data extrapolation (d)					
	Fish	Crust.	Bivalve	Aq. plant	Phytopl.	Zoopl.	Fish	Crust.	Bivalve	Aq. plant	Phytopl.	Zoopl.	Fish	Crust.	Bivalve	Aq. plant	Phytopl.	Zoopl.
^{89}Zr				5.2E+00			8.4E+00	1.4E+00	2.0E+00	6.9E-03	6.9E-03	1.4E+00	6.9E+01	8.5E+01	6.9E+01	5.2E+00	2.7E+00	6.2E+01
^{90}Y				5.2E+00			8.4E+00	1.4E+00	2.0E+00	6.9E-03	6.9E-03	1.4E+00	6.9E+01	8.5E+01	3.4E+01	5.2E+00	2.7E+00	6.2E+01
^{99}Mo	3.4E+00	3.0E+00 (14%) 1.4E+02 (83%)	1.0E+02			3.0E+00 (14%) 1.4E+02 (83%)	8.4E+00	3.0E+00	2.0E+00	6.9E-03	6.9E-03	3.0E+00	3.4E+00	1.4E+02	1.0E+02	3.6E+00	2.7E+00	1.4E+02
^{99m}Tc	3.4E+00	3.0E+00 (14%) 1.4E+02 (83%)	1.0E+02			3.0E+00 ((14%) 1.4E+02 (83%)	8.4E+00	3.0E+00	2.3E+03	6.9E-03	6.9E-03	3.0E+00	3.4E+00	1.4E+02	1.0E+02	3.6E+00	3.0E+00	1.4E+02
^{131}I	3.4E+00	3.0E+00 (14%) 1.4E+02 (83%)	1.0E+02			3.0E+00 (14%) 1.4E+02 (83%)	8.4E+00	3.0E+00	2.3E+03	6.9E-03	6.9E-03	3.0E+00	3.4E+00	1.4E+02	1.0E+02	3.6E+00	3.0E+00	1.4E+02
^{131m}Xe	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
^{133}Xe	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00	0.0E+00
^{177}Lu			2.0E+00 (61%) 3.4E+01 (39%)	2.7E+00	2.7E+00		8.4E+00	1.4E+00	2.0E+00	6.9E-03	6.9E-03	1.4E+00	6.9E+01	8.5E+01	3.4E+01	2.7E+00	2.7E+00	6.2E+01
^{177m}Lu			2.0E+00 (61%) 3.4E+01 (39%)	2.7E+00	2.7E+00		8.4E+00	1.4E+00	2.0E+00	6.9E-03	6.9E-03	1.4E+00	6.9E+01	8.5E+01	3.4E+01	2.7E+00	2.7E+00	6.2E+01
^{223}Ra	1.0E+01 2.7E+02	1.7E-01 4.3E+01	4.0E+03	6.9E-03 3.2E+00	6.9E-03 3.2E+00	2.5E-01 2.3E+00	1.0E+01	1.7E-01	4.0E+03	6.9E-03	6.9E-03	2.5E-01	2.7E+02	4.3E+01	4.0E+03	3.2E+00	3.2E+00	2.3E+00
^{225}Ac							8.4E+00	1.7E-01	4.0E+03	6.9E-03	6.9E-03	2.5E-01	1.6E+02	4.3E+01	4.0E+03	3.2E+00	3.2E+00	2.3E+00
^{226}Ra	6.7E+00 4.9E+01	1.7E-01 4.3E+01	4.0E+03	6.9E-03 3.2E+00	6.9E-03 3.2E+00	2.5E-01 2.3E+00	6.7E+00	1.7E-01	4.0E+03	6.9E-03	6.9E-03	2.5E-01	5.0E+01	4.3E+01	4.0E+03	3.2E+00	3.2E+00	2.3E+00
^{227}Th	8.8E-01						8.4E+00	1.7E-01	4.0E+03	6.9E-03	6.9E-03	2.5E-01	8.8E-01	4.3E+01	4.0E+03	3.2E+00	3.2E+00	2.3E+00
^{18}F							8.4E+00	3.0E+00	2.3E+03	6.9E-03	6.9E-03	3.0E+00	3.4E+00	1.4E+02	1.0E+02	3.2E+00	3.0E+00	1.4E+02
^{123}I	0.0E+00	0.0E+00	0.0E+00			0.0E+00	8.4E+00	3.0E+00	2.3E+03	6.9E-03	6.9E-03	3.0E+00	3.4E+00	1.4E+02	1.0E+02	3.2E+00	3.0E+00	1.4E+02
^{153}Sm			2.0E+00 (61%) 3.4E+01 (39%)	2.7E+00	2.7E+00		8.4E+00	1.4E+00	2.3E+03	6.9E-03	6.9E-03	1.4E+00	6.9E+01	8.5E+01	3.4E+01	2.7E+00	2.7E+00	6.2E+01
^{201}Tl							8.4E+00	1.4E+00	2.3E+03	6.9E-03	6.9E-03	1.4E+00	6.9E+01	8.5E+01	2.4E+03	3.4E+00	3.1E+00	6.2E+01

From direct sources describing uptake experiments (Hevland, 1981; McKenzie-Carter, 1985).
Using Tc as an analogue for Mo - nearest transition metal in terms of atomic number.
Published data for carp (*Cyprinus carpio*) and mosquitofish (*Gambusia affinis*) (Blaylock and Frank, 1981).
Assumption of zero for noble gases.
Taking Ce as analogue and using the MODARIA WG8 Biological half-life database (http://dx.doi.org/10.1016/j.jenvrad.2015.08.018).
Direct average from MODARIA WG8 Biological half-life database (http://dx.doi.org/10.1016/j.jenvrad.2015.08.018).
Published data (Mahmood et al., 2014).
Published data (Seaman and Kaplan, 2010).

2.4 Adaptation of the D-DAT model for freshwater assessments

2.4.1 Brief description of the D-DAT model

The D-DAT assessment model (Vives i Batlle et al., 2008) calculates aquatic wildlife radionuclide concentrations (fish, crustaceans, molluscs, macroalgae, phytoplankton and zooplankton) and the resulting dose rates to biota using time series of contaminated water concentrations (measured or modelled) as input. In addition, the model contains a sediment sub-model that considers suspended particulates, molecular diffusion, pore water mixing and bioturbation, in order to dynamically calculate sediment activity concentrations and therefore external dose rates to wildlife arising from sediment exposure. This model has been successfully applied to Fukushima studies (Vives i Batlle et al., 2018) and was further developed into an advanced version which was the offspring of the Euratom project COMET (Vives Batlle, 2013; Vives i Batlle et al., 2018). That model implements a dual $T_{B1/2}$ approach, requiring three compartments: water (A_W), as well as a fast (A_{OF}) and slow (A_{OS}) organism, linked to fast and slow routes of uptake and release, respectively:

$$\frac{dA_W}{dt} = -\left(K_{Wf} + K_{WS} + \lambda\right)A_W + \frac{m}{V}\left(K_{OF}A_{OF} + K_{OS}A_{OS}\right)$$

$$\frac{dA_{Of}}{dt} = K_{WF}\frac{m}{V}A_W - \left(K_{Of} + \lambda\right)A_{OF}; \frac{dA_{Os}}{dt} = K_{WS}\frac{m}{V}A_W - \left(K_{OS} + \lambda\right)A_{OF}$$

Where K_{ij} are the rate constants governing transfer from compartment *i* to compartment *j* (where *i* and *j* symbolise the water, organism-fast and organism-slow retention phases *W*, *OF* or *OS*); λ is the radionuclide decay constant; *m* is the mass of the organism; *V* is the volume of the water compartment with $K_{Of} = \frac{ln(2)}{T^F_{B1/2}}$ and $K_{Os} = \frac{ln(2)}{T^S_{B1/2}}$ (where $T^F_{B1/2}$ and $T^S_{B1/2}$ are the two "fast" and "slow" biological half-lives of elimination which can, in the general case, be present simultaneously). The exchange of radionuclides between water and sediment is represented by the dynamic coupling of the above model with a four-compartment (water and 3 layers of sediment) linear, first order kinetic exchange model (Lepicard et al., 2004; Lepicard et al., 1998; Simmonds et al., 2004) which includes the processes of particle scavenging, molecular diffusion, particle mixing, pore water mixing and sedimentation.

To calculate internal and external dose rates to the wildlife for the various radionuclides, activity concentrations (sum of the "slow" and the "fast" component) are multiplied by the dose coefficients (μGy h^{-1} per Bq kg^{-1}), or DCs, for the required organism from ICRP Publication 136 (ICRP, 2017). External dose rates are calculated similarly by using external exposure DCs and purposely-defined occupancy factors that account for hybrid exposure from both water and sediment.

D-DAT is currently implemented in the ModelMaker® 4 software (Adamatzky, 2001; Citra, 1997; Rigas, 2000) and, in this form, it has had a very successful track record of application to a variety of environmental situations involving non-continuous discharges of radionuclides in the marine

environment, as well as having been successfully tested in inter-comparisons with other dynamic models (Vives Batlle et al., 2008; Vives i Batlle, 2016; Vives i Batlle et al., 2016).

The ability of D-DAT to balance incoming activity concentrations of radionuclides in water at the wildlife receptor location, combined with the explicit modelling of the role of sediments as a potential dose-giving reservoir of radionuclides, means that D-DAT is eminently suitable for adaptation to carry out the present study for the freshwater environment, extending the original radionuclide set (the model was originally designed for the long-lived radionuclides ^{90}Sr, ^{99}Tc, ^{129,131}I, ^{134,1237}Cs, ^{239,240}Pu, ^{241}Am and ^{236}U) to the short-lived medical radionuclides considered in medical releases.

2.5 Adaptation of D-DAT to medical radionuclides in freshwater

The D-DAT model performs simultaneous calculations for a suite of radionuclides and organisms thanks to a very compact form of the model's differential equations, which are indexed as a two-dimensional array, with the first index *i* signifying the radionuclide and the index *j* signifying the wildlife group. The model's parameters are also stored in multidimensional array format. Therefore, the first step in the adaptation of D-DAT for freshwater was the re-indexing of all the compartments and fluxes of the model to accommodate the new radionuclides, as shown in Fig. 1.

A significant improvement to the original D-DAT model was adding a human dosimetry post-processing module. This module is capable of calculating as function of time: (a) time-dependent aquatic food ingestion doses, and associated annually-averaged ingestion doses to all age groups, foods and radionuclides; (b) time-dependent and annually-averaged water ingestion dose rates and (c) time-dependent, and also annually averaged, external dose rates from exposure to both sediment (riverbank) and water (swimming) exposure to all radionuclides.

Internal dose rates are calculated conventionally by multiplying the activity concentration in the wildlife used as food (Bq kg^{-1}) per the ingestion rate (kg y^{-1}), the dose per unit via intake via ingestion (Sv Bq^{-1}) from ICRP Publications 72 and 119 (ICRP, 1996, 2012) and the fraction of food that is obtained locally. For external exposures, the model uses the committed effective doses to 70 years of age per unit time and deposited activity of radionuclide on the riverbank in Sv Bq^{-1} s^{-1} m^{2} (Eckerman and Ryman, 1993), maximally assumed to be as contaminated as sediment, so as to estimate doses for riverbank exposure to members of the public. We deduced the modelled activity per unit area of riverbank soil (Bq m^{-2}) by multiplying the activity concentration by a 0.3-m active soil contamination depth and by the external dose coefficient and per the number of seconds in a year, with further application of a factor of 0.5 to account for the geometry of the source/target distribution on the riverbank and application of the occupancy factor, giving the external exposure dose rate.

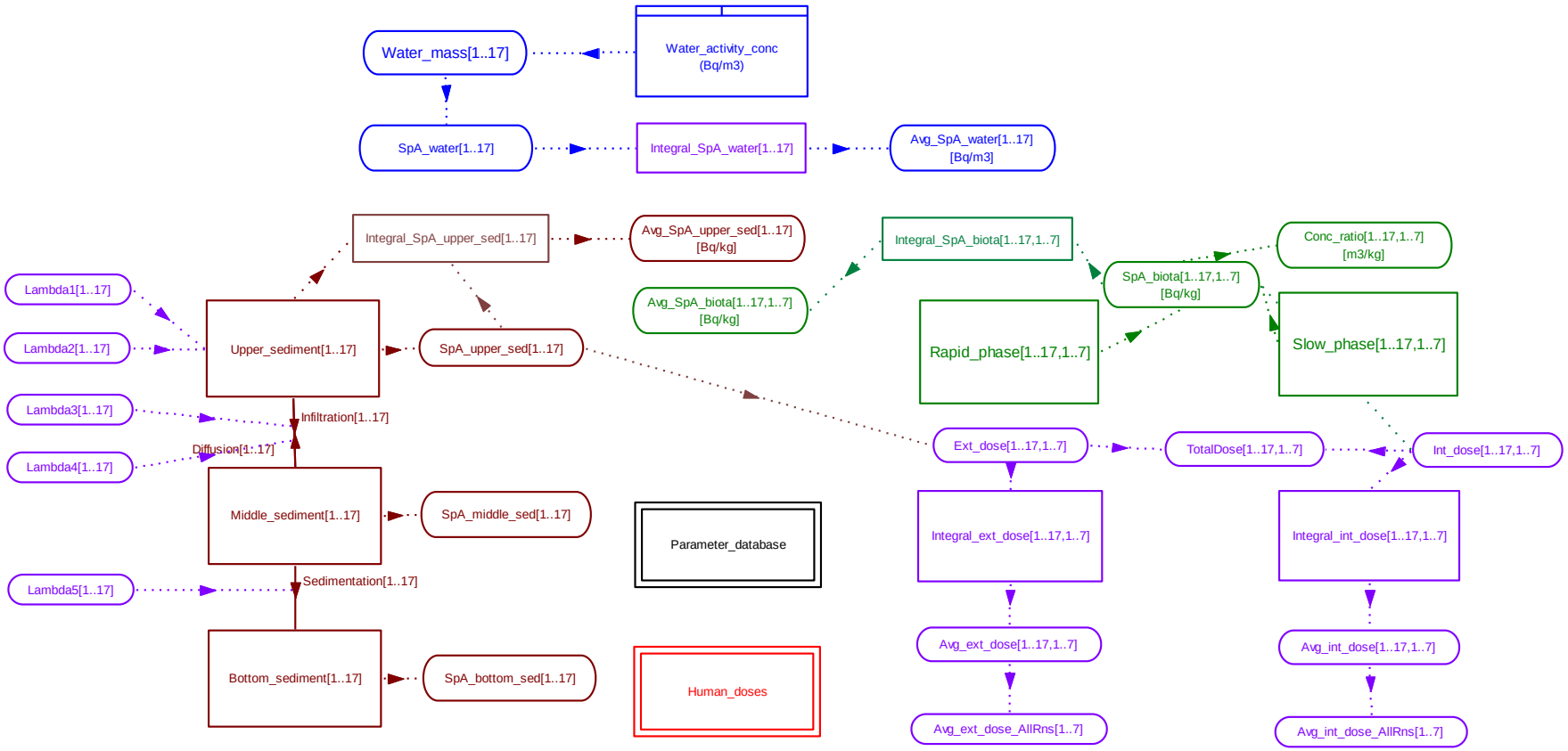

Figure 1: Representation of the new version of D-DAT for freshwater in ModelMaker 4, showing integrating compartments (rectangles), embedded sub-models (double rectangles), variables (rounded rectangles) and influences (dotted arrows). The compartments are indexed as two-dimensional arrays as follows. The sub-indices i = 1 to 17 represent the radionuclides ^{89}Zr, ^{90}Y, ^{99}Mo, ^{99m}Tc, ^{131}I, ^{131m}Xe, ^{133}Xe, ^{177}Lu, ^{177m}Lu, ^{223}Ra, ^{225}Ac, ^{226}Ra, ^{227}Th, ^{18}F, ^{123}I, ^{153}Sm and ^{201}Tl, respectively. The sub-indices j = 1 to 7 represent the freshwater biota organisms Pelagic Fish, Benthic Fish, Crustacean, Mollusc, Macroalgae, Phyrtoplankton and Zooplankton, respectively.

The model uses an average individual shore occupancy rate of around 500 h per year (occupancy fraction of 5.7×10^{-2}), deemed to be sufficiently conservative. Any additional dose from irradiation of the skin due to direct contact with sediment is not included in the methodology because it is not a major contributor to the overall doses.

The above extensions to the model required the addition of new parameters, namely human food ingestion and water drinking rates for infant, child and adult, occupancy factors for external exposure to shoreline sediments, fraction of locally produced food, internal dose coefficients for ingestion and external dose coefficient for ground surface and water immersion.

The model was verified to check that all equations were correct. In particular, the complex input data structure was scrutinised to ensure that the input parameters are read correctly, and the integration algorithm was optimised, given the large number of data points of the input file provided by the hydrological simulations, which give a full year of water activity concentration data at 10-minute intervals. In the end, the Euler solving method was selected, with a random seed of one and running with a fixed step of 5.2×10^{4} user-defined output points.

2.6 Excel dose calculator for water treatment plant workers, sewer maintenance workers and the public

The initial starting point was the UK NRPB methodology for the radiological assessment of liquid pharmaceutical discharges in sewers (McDonnell, 2004; Titley et al., 2000), which has now been developed into the IRAT-2 approach (https://www.gov.uk/government/publications/initial-radiological-assessment-methodology). We used a modified and simplified approach to suit the methodological needs of the Belgian situation, and we implemented the resulting equations in an Excel calculator to perform dose screening to workers and the public drinking the water and eating from the terrestrial foodchain, based on monthly concentration averages.

Calculations are based on the assumption of steady state conditions, where discharges are assumed to take place at a uniform rate and there is little change in the flow rates downstream of the discharge point. This is because there is no simple equivalent of the D-DAT model in the form of a tool to calculate dynamically doses from short-lived radionuclides to humans. This would involve use of complex pharmacokinetic models, with an inevitable lack of associated parameter data for the radionuclides involved. In general, complex models comprise large amount of parameters and variables that are difficult to obtain. Our simplified approach is appropriate for screening assessments, requires relatively few parameters and is adequately conservative.

The calculator includes the following radionuclides: ^{18}F, ^{89}Zr, ^{90}Y, ^{99}Mo, ^{99m}Tc, ^{123}I, ^{131}I, ^{131m}Xe, ^{133}Xe, ^{153}Sm, ^{177}Lu, ^{177m}Lu, ^{201}Tl, ^{223}Ra, ^{225}Ac, ^{226}Ra and ^{227}Th. However, we used only ^{18}F, ^{123}I, ^{131}I, ^{153}Sm, ^{99m}Tc and ^{201}Tl as these are the only radionuclides for which underwater radioactivity measurements (gamma spectrometry) were provided by the authorities.

Using as input annual discharges (calculated as average annual concentration in Bq m^{-3} multiplied by the flow rate in m^3 y^{-1}), our model calculates activity concentrations in the different waste streams at the plant, namely effluent, a blocked sewer and sludge, whereupon doses to general and maintenance plant workers could be calculated. This tool also calculates dose rates for the most exposed members of the public: consumers of locally caught fish, external exposure from frequenting the riverbank, direct drinking of river water, abstraction of river water for irrigation or drinking water and use of sludge as fertiliser for agricultural processes. However, it does so assuming average concentrations in water, thereby making a conservative estimation, whereas the D-DAT model is more appropriate to make a more detailed, dynamic calculation of doses to public and the environment – so exposes to the public can in effect be compared here for different modelling assumptions.

2.6.1 Dosimetry tool description

The tool as developed has the following calculation worksheets: general assessment parameters, basic radionuclide data, source term fractions, calculation of dose rates and additional worksheets for transfer factors, dose factors and assessment assumptions. The general assessment parameters worksheet contains the required parameter data for the assessment: disposal pathway parameters, data for assessment of exposure of sewer workers, data used for terrestrial food chain calculations, data used for soil hydrology calculations and habit data and other parameters for public exposure. Element independent parameters where chosen specific for the Belgian dataset from the Category A Waste Disposal project (Sweeck, 2018) or (when not available) literature values (McDonnell, 2004; Titley et al., 2000) and/or plain expert judgement.

The basic radionuclide data parameters worksheet contains radionuclide half-lives, external dose coefficients for ground surface and water immersion and internal dose coefficients for ingestion and inhalation for their stated lung classes. The source term fractions worksheet contains an estimation of the source term and fractions appearing in sewage, leading to estimation of the average radionuclide concentrations serving as source term for the different assessment locations of interest: blocked sewer scenario, plant water streams, sludge, the river ecosystem at the plant's outlet and the use of river water for irrigation and drinking.

The calculation of dose rates worksheet contains the main calculation block, with embedded equations to calculate dose rates to sewer maintenance workers, general workers at the sewage works, dose rates for the freshwater pathways and dose rates arising from use of river water for irrigation of farmland and use of sewage sludge in agriculture, as detailed in the sub-sections below. Additional worksheets detail the transfer and dosimetry factors used in this model for information and data sourcing purposes.

2.6.1.1 Calculation of activity concentrations in blocked sewer and the river

The mean cumulative discharge over a month in Bq, A_M [Bq] is the main input to the model. We assume that 100% of the radionuclides discharged from the hospital reach the treatment plant, which is a

conservative assumption but one that allows us to obviate site-specific river dispersion modelling calculations between the hospital and the plant in favour of a more generic type of screening methodology, especially in the present case in which (as will be seen below) the radiological impact, even with this assumption, is not significant.

It is assumed that the plant has a certain flow of water going through it, ϕ_{WWTP}, which is less than the total flow of the river - the water going through the plant is in fact 1.1% of the total river flow, if we assume a typical throughput of 1000 m^3 per day in a river of mean flow of 1.0247 m^3 s^{-1} as is typical of the Molse Nete river.

The apportioning of the incoming (average) radionuclide concentration [Bq m^{-3}] between the various compartments is as follows. For a blocked sewer, it is assumed that 1 month-worth of discharges are trapped in a pipe blockage, as previously espoused elsewhere (McDonnell, 2004). We take the following equation:

$$C_{blocked\ sewer}[Bq\ m^{-3}] = \frac{A_M[Bq]}{V_{blockage}[m^3]} \times \frac{1}{\lambda T} \times f_{blocked\ drain}$$

Where λ is the decay constant, T = 30 days (the factor $\frac{1}{\lambda T}$ is the result of averaging $e^{-\lambda t}$ between 0 and T to correct for ongoing decay during the 30 days, assuming that λT >>1: $A_{avg} = \frac{1}{T}\int_0^E A_0 e^{-\lambda t} dt = \frac{A_0}{\lambda T}\left(1 - e^{-\lambda t}\right) \approx \frac{A_0}{\lambda T}$. Moreover, $f_{blocked\ drain}$ is the percentage of activity ending in the blockage divided by 100. In other words, the model assumes that a single month's discharge is contained in a small volume (2 m^3) of sewage at a point where the drains have been blocked, with workers operating in the vicinity, whilst undergoing decay. The volume estimation is a best judgement assumption (Titley et al., 2000).

For the plant water streams, we simply consider:

$$C_{stream}[Bq\ m^{-3}] = \frac{12A_M/s_y[Bq\ y^{-1}]}{\emptyset[m^3 y^{-1}]} \times f_{reaching\ WWTP}$$

Where $12A_M/s_y$ is simply the average annual discharge rate in Bq s^{-1} (A_M is the monthly discharge and s_y is the number of seconds in a year).Therefore, for the sludge:

$$C_{sludge}[Bq\ m^{-3}] = \frac{12A_M/s_y[Bq\ y^{-1}]}{\emptyset[m^3 y^{-1}] \times r_s[-]} \times f_{in\ sludge}$$

Where r_s is the annual rate of sludge production to incoming sewage = sludge yearly production rate [m^3 y^{-1}] per unit of total incoming sewage flow rate [m^3 y^{-1}].

For the aquatic pathways, the average activity concentration in river water after release is:

$$C_{RW}[Bq\ m^{-3}] = \frac{12A_M/s_y[Bq\ y^{-1}]}{\emptyset_{river}[m^3 y^{-1}]} \times (1 - \varepsilon)$$

Where ε is the treatment plant's removal efficiency. There are three cases of river flow rate that in our calculations are assumed to have the same water concentrations: Water for fish and the calculation of the various external exposure pathways such as irrigation and public drinking water supply. Different values for these pathways could be introduced if the need arises. It is assumed here that the measured activity concentrations supplied to us by the authorities, being so close to the plant, reflect the concentration of the undiluted effluent (a conservative assumption).

Note that an improvement to this methodology would be to introduce decay terms to account for the time delay in the sewage plant. Decay during transit is ignored in this study because the dose rates are so low that it does not seem necessary to undergo the complication to calculate to that level of detail. Indicative delays that could be used are 0.5 day for milk consumption, 182 days for root vegetables and 7 days for all other foodstuffs (Titley et al., 2000).

2.6.1.2 Calculation of concentrations for the irrigation and sludge fertiliser pathways

For the irrigation pathway, the starting point is the radionuclide concentration in water C_{RW} [Bq m^{-3}], the irrigation water flux $\emptyset_W$ [m^3 m^{-2} s^{-1}], the active depth of contamination (average root soil depth for food vegetables) d [m] and the surface area of the soil = S [m^{-2}]. The amount of water infiltrating in the soil per unit time is given by the infiltration equation, which takes into account the (higher) pore water velocity ($\upsilon_p = \upsilon/\theta$ where θ is the volumetric water content of the soil) compared with the irrigation water fall-in rate υ: $\frac{dV}{dt} = \frac{\emptyset_W S}{\theta}$ [m^3 s^{-1}]. The rate of change of radionuclide in soil, C_{soil} [Bq] due to infiltration is $\left(\frac{dC}{dt}\right)_{inf} = C_{RW}\frac{dV}{dt} = \frac{C_{RW}\emptyset_W S}{\theta}R$ [Bq s^{-1}]. Here, $R = \left[1 + \frac{\rho K_d}{\theta}\right]^{-1} = \left[1 + \frac{\rho_p(1-\varepsilon)K_d}{\theta}\right]^{-1}$ is the retardation factor, introduced to consider that the radionuclide may be infiltrating at a lower velocity than the water, due to sorption processes as the dissolved radionuclide migrates downwards across the soil column. The additional parameters in this equation are the soil/water distribution coefficient K_d [m^3 kg^{-1}] and the volumetric water content θ[-]. The porosity can be expressed as $\varepsilon = 1 - \frac{\rho}{\rho_0}$ where ρ is the bulk density of the soil and ρ_p is the (higher) particle density (both in kg m^{-3}).

Since the radionuclide is fast decaying, at equilibrium, infiltration balances the loss due to decay which, according to the definition of activity, is proportional to the decay constant: $\left(\frac{dC}{dt}\right)_{decay} = \lambda C_{soil} S d$ [Bq s^{-1}]. Here, C_{PW} is the activity concentration in soil pore water [Bq m^{-3}], S is the surface area and d is the active depth of the contamination. Therefore, $\left(\frac{dC}{dt}\right)_{inf} = \left(\frac{dC}{dt}\right)_{decay} \Longrightarrow \frac{C_{RW}\emptyset_W S}{\theta}R = \lambda C_{PW} S d$. Hence, we arrive at $C_{PW} = \frac{1}{\theta + \rho K_d}\left(\frac{C_{RW}\emptyset_W}{\lambda d}\right)$.

The concentration in soil under conditions of equilibrium can be obtained as $C_{soil} = C_{PW}K_d$, and the concentration in the vegetables is obtained using by further multiplication by the concentration ratio CF [m^3 kg^{-1}]: $C_{veg} = C_{soil}CF = C_{PW}K_d CF$. Thus, the food concentration is $C_{veg} = \frac{1}{\theta + \rho K_d}\left(\frac{C_{RW}\emptyset_W}{\lambda d}\right)K_d CF$,

so we finally obtain a food activity per unit deposited activity ratio R [Bq/kg per 1 Bq/m^2 per second] of $R = \frac{C_{veg}}{C_{RW}\emptyset_W} = \frac{1}{\theta+\rho K_d}\left(\frac{K_d CF}{\lambda d}\right)$.

In this project, we used the following element-independent reference biosphere parameters as input for the equations, consistent with the near-surface disposal project for category A waste at Dessel, Belgium (Sweeck, 2018): Volumetric water content θ = 0.32 (general case); average root soil depth d = 0.3 m; soil bulk density ρ = 1350 kg m^{-3}, soil particle density ρ_p = 2650 kg m^{-3} and thus a porosity $1 - \frac{\rho}{\rho_p}$ = 0.491.

The soil activity per unit deposition arising from a concentration in sludge C_{sl} [Bq kg^{-1}], which is being applied to farmland at a rate $\emptyset_{sl}$ [kg m^{-2} s], is calculated as follows. The rate of change of radionuclide activity in soil, A_{soil} [Bq] due to sludge deposition is: $\frac{dA_{soil}}{dt} = C_{sl}\emptyset_{sl}S$ [Bq s^{-1}], where S is the surface area of the soil. Here again we assume that influx is cancelled by decay, and therefore $C_{sl}\emptyset_{sl}S = \lambda C_{soil}\rho_{soil}Sd \Longrightarrow C_{soil} = \frac{C_{sl}\emptyset_{sl}}{\lambda d\rho_{soil}}$ (here, the term $C_{soil}\rho_{soil}Sd$ is the activity concentration in soil multiplied by the mass of the soil, to convert it to units of absolute activity). Frome here we can define a convenient soil concentration per unit deposition ratio $CPUD_{soil} = \frac{C_{soil}}{C_{sl}\emptyset_{sl}} = \frac{1}{\lambda d\rho_{soil}}$ [Bq kg^{-1} per unit Bq m^{-2} s^{-1}], which can be used for the calculation of irrigation dose rates, as shown below.

2.6.2 Calculation of dose rates to WWTP maintenance and sewage workers

In order to obtain dose rates, one multiplies the radionuclide concentration by the dose coefficient for internal exposure via inhalation or ingestion (internal dose rate) or by the dose coefficient exposure to ground surface or immersion (external dose rate), and by additional factors as described in turn below.

For accidental ingestion of sludge, one must consider the radionuclide concentration in a blocked sewer for maintenance workers, or the average of concentration into sewage works + in sludge for regular workers [Bq m^{-3}]. For the latter case, is assumed that workers spend 50% of their yearly working time in each operation. The pertinent activity concentration in sludge [Bq m^{-3}] is then divided by the density (approximated by the density of water) to convert the concentration to units of Bq kg^{-1}. Then, the result is multiplied by the ingestion rate [kg h^{-1}], the fractional occupancy f_{occ}^{worker} for the relevant type of worker [h y^{-1}] and the internal dose coefficient via ingestion DC_{ing} [Sv Bq^{-1}], leading to:

$$H_{ing} = \frac{C[\mathrm{Bq\ m^{-3}}]}{\rho[\mathrm{kg\ m^{-3}}]} \times DC_{ing}[\mathrm{Sv\ Bq^{-1}}] \times I_R[\mathrm{kg\ h^{-1}}] \times f_{occ}[\mathrm{h\ y^{-1}}]$$

For inhalation, the approach used here takes the activity concentration [Bq m^{-3}] (concentration in untreated sewage/sludge for regular workers – assumed to spent 50% of time in each operation) and divides it by the water density to convert the concentration to units of Bq kg^{-1} of sludge. This is then multiplied by the airborne particulate matter concentration χ_P [kg m^{-3}] assuming conservatively that these particles become inhaled, giving Bq in the particles per unit volume of air. This is then multiplied by

the inhalation rate B_R [m^3 h^{-1}], the fractional occupancy for the relevant type of worker [h y^{-1}] and the internal dose coefficient via inhalation DC_{inh} [Sv Bq^{-1}], leading to the following equation:

$$H_{inh} = \frac{C[\mathrm{Bq\ m^{-3}}]}{\rho[\mathrm{kg\ m^{-3}}]} \times \gamma_p[\mathrm{kg\ m^{-3}}] \times DC_{inh}[\mathrm{Sv\ Bq^{-1}}] \times B_R[\mathrm{m^3\ h^{-1}}] \times f_{occ}^{worker}[\mathrm{h\ y^{-1}}]$$

For external gamma exposure, the dose rate is proportional to the surface density of contamination [Bq m^{-2}]. The dose rate derives from the activity concentration [Bq m^{-3}] (in a blocked sewer for maintenance workers, and mean of plant water and sludge for regular workers – which assumes 50% of time spent in each operation) multiplied by the contamination active depth in the WWTP [m], assumed here to be 5 cm, the occupancy fraction for the worker's task [h y^{-1}], the external dose coefficient [Sv m^2 s^{-1} Bq^{-1}] and time units conversions:

$$H_{ext} = DC_{ext}[\mathrm{Sv\ m^2 s^{-1} Bq^{-1}}] \times C[\mathrm{Bq\ m^{-3}}] \times d[m] \times f_{occ}^{worker}[\mathrm{h\ y^{-1}}] \times y_h[\mathrm{y\ h^{-1}}] \times s_y[\mathrm{s\ y^{-1}}]$$

Where y_h and s_y are the unit conversion factors for hours in a year and seconds in a year.

2.6.3 Calculation of dose rates to the public for the freshwater pathways

In this case, it is necessary to calculate first the river input rate data as shown previously (Fiengo Pérez et al., 2022): $I_{river}[\mathrm{MBq\ y^{-1}}] = C_{river}^{avg}[Bq\ m^{-3}] \times \varphi_{river}[m^3\ s^{-1}] \times s_y[\mathrm{s\ y^{-1}}] \times 10^{-6}$. The external gamma dose rate for riverbank occupancy assumes that the riverbank concentration is the riverbed concentration, obtained by multiplication of the average water concentration by the K_d. The fraction $\frac{1}{1+S_{susp}K_d}$ is a factor used to take water filtration into account:

$$H_{ext}[\mathrm{Sv\ y^{-1}}] = \frac{1}{1+S_{susp}K_d} C_{river}^{avg}[Bq\ m^{-3}] \times K_d[m^3\ kg^{-1}] \times f_{occ}^{riverbank}[\mathrm{h\ y^{-1}}] \times$$
$$DC_{groundsurf}^{ext}[Sv\ Bq^{-1}\ s^{-1}\ m^2] \times d[m] x\ \rho_{soil}^{bulk}[kg\ m^{-3}] \times y_h[\mathrm{y\ h^{-1}}] \times s_y[\mathrm{s\ y^{-1}}].$$

The ingestion dose rate arising from fish consumption is as follows:

$$H_{ing}^{fish}[\mathrm{Sv\ y^{-1}}] = \frac{CR_{fish}[kg\ kg^{-1}]}{\rho_w[\mathrm{kg\ m^{-3}}]} \times \frac{C_{river}^{avg}[Bq\ m^{-3}]}{1+S_{susp}K_d} \times I_r^{fish}[kg\ y^{-1}] \times DC_{ing}[\mathrm{Sv\ Bq^{-1}}]$$

In addition, for the ingestion of drinking water and unfiltered river water:

$$H_{ing}^{drwater}[\mathrm{Sv\ y^{-1}}] = \frac{C_{river}^{avg}[Bq\ m^{-3}]}{1+S_{susp}K_d} \times I_r^{drwater}[m^3\ y^{-1}] \times DC_{ing}[\mathrm{Sv\ Bq^{-1}}]$$

$$H_{ing}^{unfwater}[\mathrm{Sv\ y^{-1}}] = C_{river}^{avg}[Bq\ m^{-3}] \times I_r^{drwater}[m^3\ y^{-1}] \times DC_{ing}[\mathrm{Sv\ Bq^{-1}}]$$

2.6.4 Calculation of dose rates to the public arising from the agricultural use of sludge

As seen above, $C_{soil}[Bq\ kg^{-1}] = CPUD_{soil}[m^2\ s\ kg^{-1}] \times C_{sl}[\mathrm{Bq\ kg^{-1}}] \times \emptyset_{sl}[kg\ m^{-2}s^{-1}]$. This can be converted to external dose rate (internal exposure is negligible) by means of the following equation:

$$H_{ext} = DC_{ext}[\mathrm{Sv\ m^2 s^{-1} Bq^{-1}}] \times C_{soil}[\mathrm{Bq\ kg^{-1}}] \times \rho[\mathrm{kg\ m^{-3}}] \times d[m] \times f_{occ}^{farmland}[\mathrm{h\ y^{-1}}] \times y_h[\mathrm{y\ h^{-1}}] \times s_y[\mathrm{s\ y^{-1}}]$$

3. RESULTS AND DISCUSSION

3.1 Routine release scenario

3.1.1 Activity concentrations in water, sediment and the wildlife

Figure 2 gives the ctivity concentrations for the 6 radionuclides considered in the Molse Nete river at the outlet of the Mol WWTP as modelled by us (Fiengo Pérez et al., 2022), and the resulting activity concentration (calculated dynamically using the D-DAT model) in the upper layer of the riverbed sediment (assumed to be 5-cm). As stated previously, levels in river water assume extraordinary circumstances such as maintenance or expansion at the WWTP whereupon effluents are directly released into the river system under conditions of low flow.

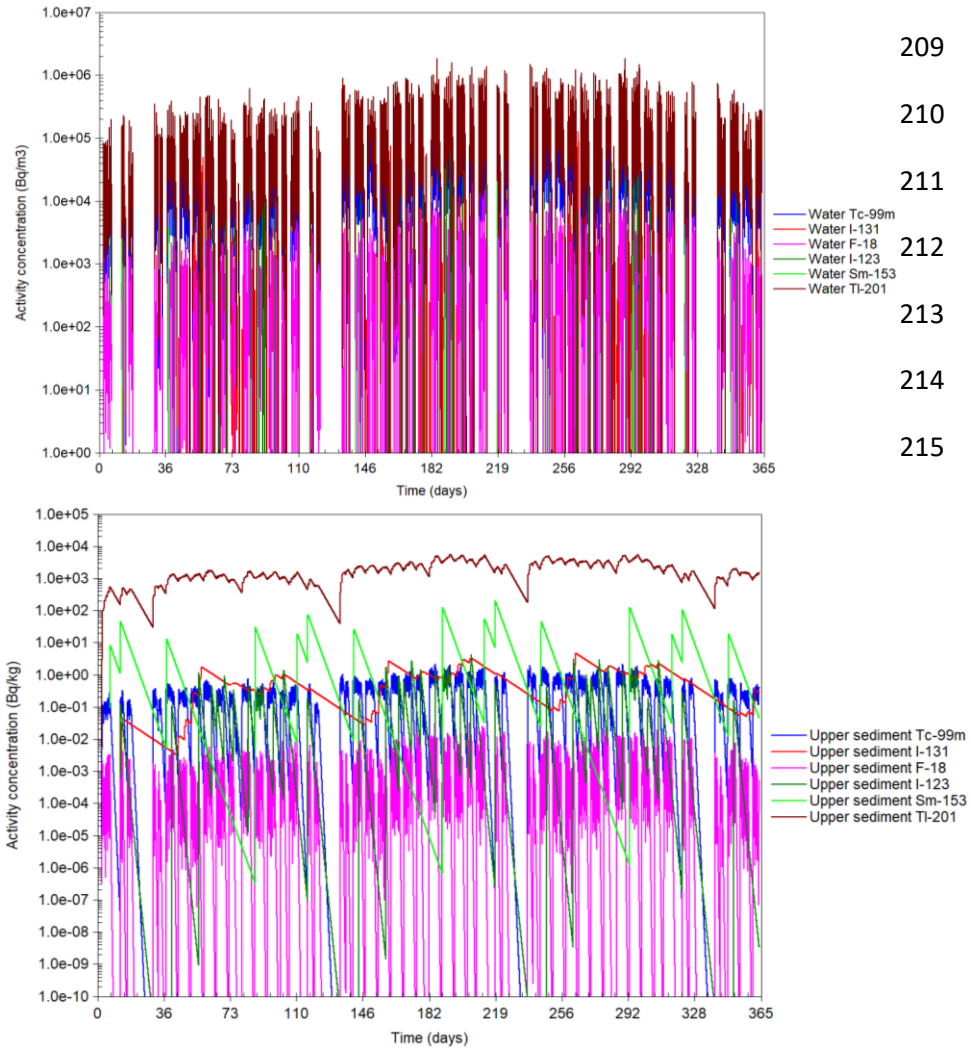

Figure 2: Activity concentrations in Molse Nete river water in 2018 used as source term for the modelling (above) and modelled concentrations in sediment (below)

The activity concentrations in water exhibit daily fluctuations directly related to the activities of the hospital discharges. The principal radionuclides in terms of activity concentration in water (by two orders of magnitude) are ^{201}Tl, followed by $^{123/131}$I and ^{99m}Tc, ^{18}F and the remainder. For sediment, the order is ^{201}Tl, ^{153}Sm, then ^{131}I and ^{99}Tc, ^{18}F and the remaining radionuclides. The time-integrating action of the sediment smoothens somewhat the oscillating water radionuclide levels, especially for ^{201}Tl and ^{153}Sm. Note that for the calculation of activity in upper sediment, we used an indicative reworking rate of 1.37×10^{-5} m d^{-1}, typical of shallow environments (Simmonds et al., 2004).

Figure 3 gives the dynamically modelled activity concentration of ^{18}F, ^{99m}Tc, ^{123}I, ^{131}I, ^{153}Sm, and ^{201}Tl in pelagic & benthic fish, crustaceans, mollusc, macro-algae, phytoplankton and zooplankton. From this figure, it can be seen that ^{18}F, ^{99}Tc, ^{153}Sm and ^{201}Tl concentrate principally in plankton, whereas for ^{123}I and ^{131}I, the radionuclide concentrates principally in fish. The activity concentrations display maxima in the order of magnitude 10^6 (^{201}Tl), 10^4 (^{18}F, ^{99}Tc and ^{153}Sm) and 10^3 ($^{123/131}$I) Bq kg^{-1}.

The main transfer parameters used to derive the above activity concentrations are detailed in Tables 1 and 2. We used data from the ERICA Tool, either primary data or applying the Tool's deductive method based on analogues. For some radionuclides (in our assessment, this concerns I and Tc) there is an additional source containing element dependent environmental input parameters for the Category A waste disposal project in Belgium (Sweeck, 2022), including some transfer parameters that cannot be found in IAEA TRS472 (IAEA, 2010). The reported K_d values for these radionuclides are 10^{-1} and 1.0×10^{-2} m^3 kg^{-1}, respectively (or 3.6 m^3 kg^{-1} if Mo and Tc are considered as analogues, as recommended here). Whilst drawing attention to this source, we retain the results obtained with the ERICA method because (a) the values used are either similar or more conservative, (b) there is consistency with the calculation approach for the other radionuclides for which there are no data in the Belgian source, and (c) an assessment for an European scenario would tend to use the ERICA values. A similar situation occurs for the concentration factors. The Belgian best-estimate CF for fish are 10^{-1} m^3 kg^{-1} for I and 1.5×10^{-2} for Tc, compared with the values in our database of 3.1×10^{-1} and 9.9×10^{-2} m^3 kg^{-1}, respectively, which again implies a higher degree of conservatism for the parameters in our database, as desired.

3.1.2 Dose rates to the wildlife

The dynamically modelled dose rates of ^{18}F, ^{99m}Tc, ^{123}I, ^{131}I, ^{153}Sm, and ^{201}Tl to pelagic & benthic fish, crustaceans, mollusc, macro-algae, phytoplankton and zooplankton, unweighted by radiation quality, are given in Fig. 4. This figure gives the total dose rate, summing of internal and external exposures (according to our simulations, internal exposure dominates over external by 2 – 3 orders of magnitude). The peaks in Figure 4 shows absolute maximum values of the order of magnitude 10^{-1} (^{99}Tc, $^{123/131}$I, ^{153}Sm), 10^0 (^{18}F) and 10^1 (^{201}Tl) µGy h^{-1} when using the generic and highly conservative source term calculated in this study.

In a dynamic situation like the one considered, peak maximum dose rates are not a meaningful quantity to measure the risk, because such dose rates are applied over a short time and lower dose rates prevail for most of the time. Rather, it is the integration of the dose rate received over a set of time divided by the time, i.e. the average dose, that should be used to compare with benchmark values of dose. The D-DAT model performs such a calculation for a time of 1 year, as shown in Fig. 5. For a given time T, the output is the average dose rate between t = zero and t = T, divided by T. In particular, we take the last point in the graph (T = 365 days) to give an average dose rate for the period.

The following conclusions are evident from Fig. 5. Firstly, external exposures (in the order macroalgae > mollusc > benthic fish > phytoplankton and zooplankton > pelagic fish) are several orders of magnitude below internal exposures, which are in the order mollusc > phytoplankton > crustacean and benthic fish > zooplankton and pelagic fish > macroalgae. Secondly, the highest annually averaged dose rate (internal dose rate for mollusc, arising mainly from ^{201}Tl), at 3.7 µGy h^{-1}, is below the 10 µGy h^{-1} incremental screening dose rate for risk characterisation from the ERICA methodology (Brown et al., 2016; Brown et al., 2008), with a risk quotient of 0.37.

The object of protection within the ERICA Integrated Approach is that generic ecosystems are protected from structure and function effects under chronic exposures. The ERICA methodology proposes the aforesaid 10 µGy h^{-1} screening dose rate based on examination of data on effects of ionising radiation in wildlife (Copplestone et al., 2008; FREDERICA, 2006). This is not a limit: exceeding it means simply that the site under analysis cannot be screened-out from further detailed assessment. In our case, therefore, it is therefore possible to state that the aquatic biota are not at risk, especially considering that the dose rates are well below the lower level of the ICRP derived consideration reference level (DCRL) bands for biota (ICRP, 2008).

3.2 Accidental ^{131}I release scenario

3.2.1 Activity concentrations in water, sediment and the wildlife

Fig. 6 gives the activity concentration in water and sediment per MBq ^{131}I release directly into the sewer system (iodine pill release scenario), reaching the river at the outlet of the WWTP during plant shutdown when the river carries the lowest flow. This scenario can be scaled-up if desired, as the doses are proportional to release. The resulting time-dependent and time-averaged dose rates are given in Fig. 6.

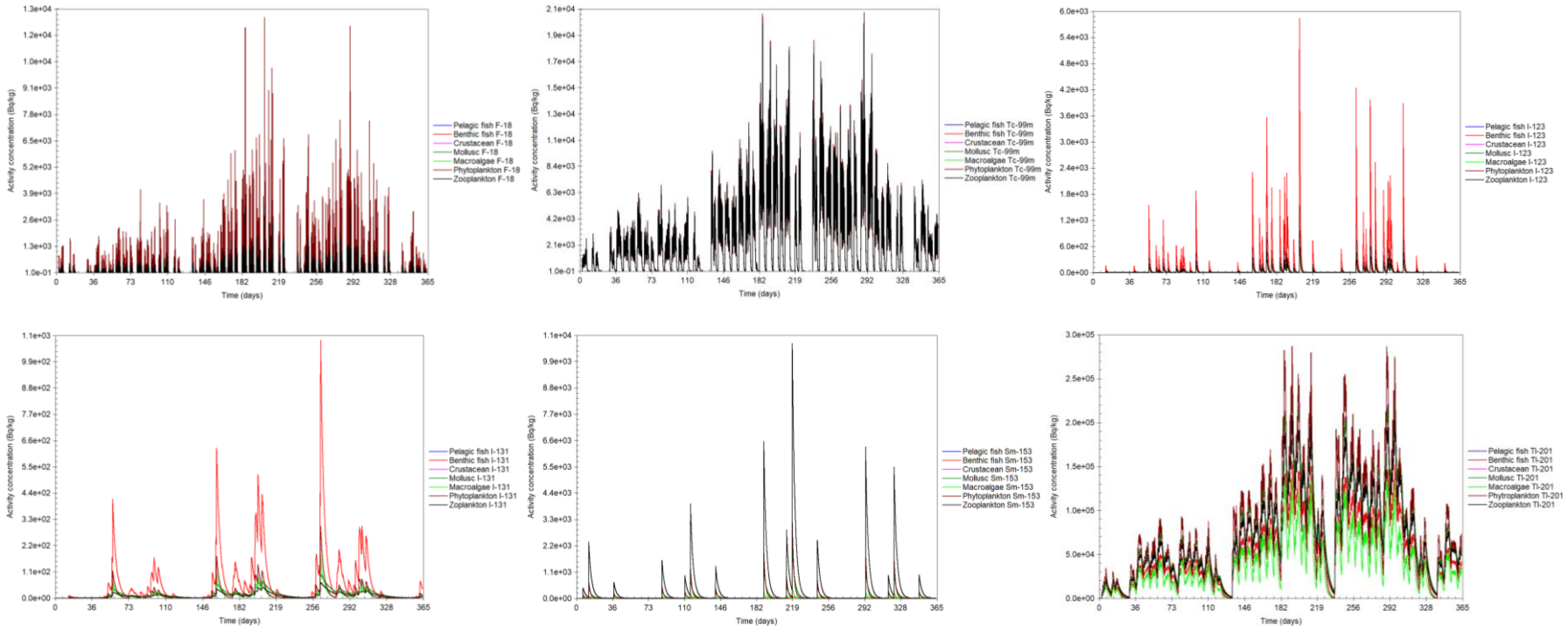

Figure 3: Dynamically modelled activity concentration of radionuclides in aquatic wildlife (Bq kg^{-1}) from the Molse Nete River

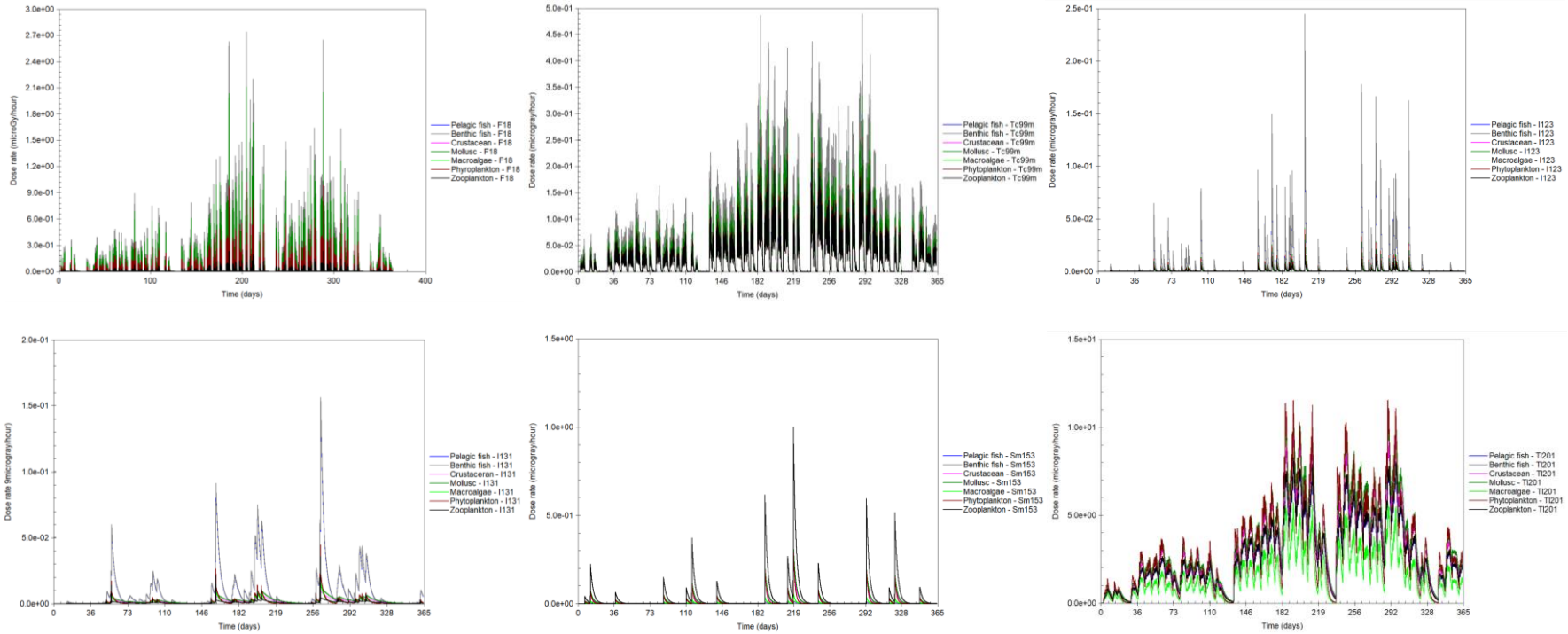

Figure 4: Dynamically modelled dose rates of radionuclides in aquatic wildlife (μGy h^{-1}) from the Molse Nete River

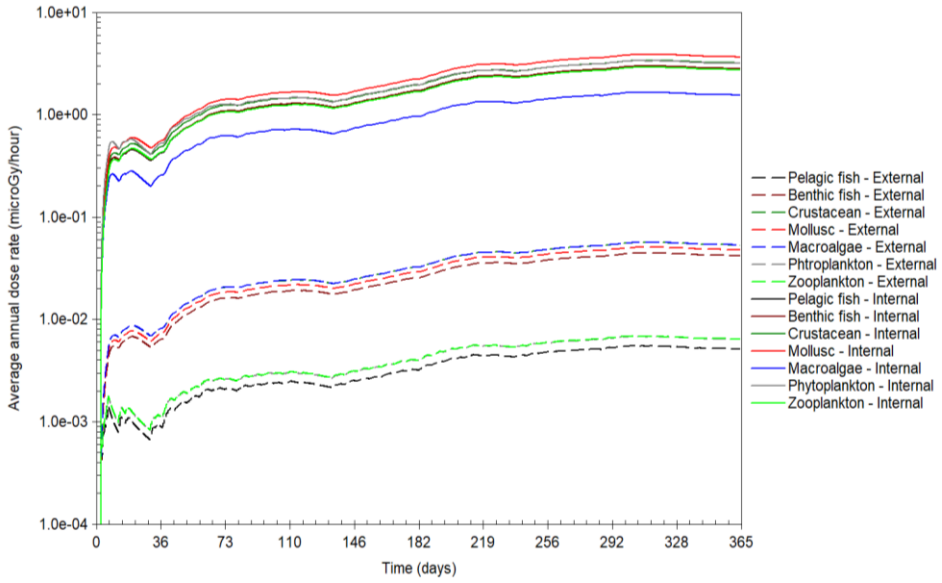

Figure 6: Time average of internal and external dose rates (sum of all radionuclides)

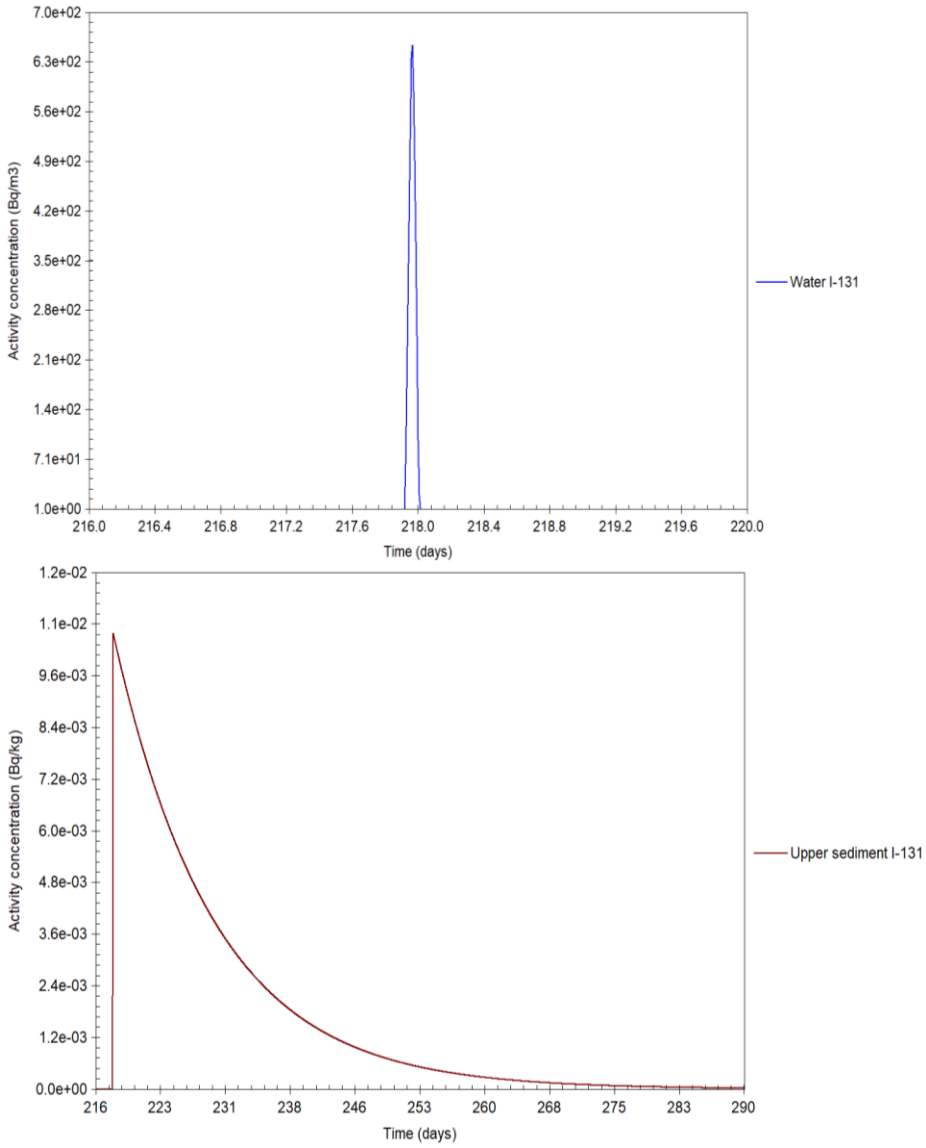

Figure 5: Modelled concentrations of ^{131}I in water (above) and sediment (below) for the accidental iodate pill release scenario

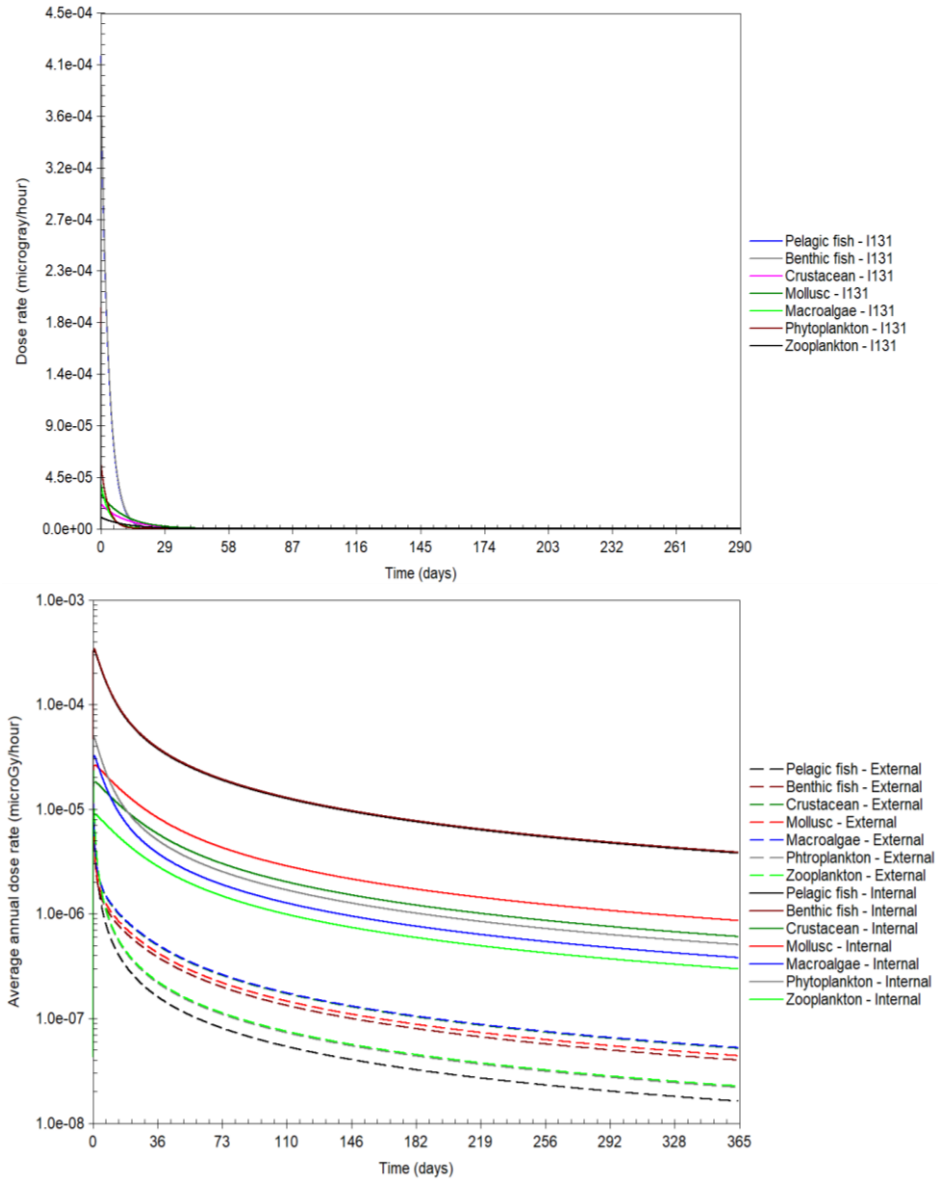

Figure 7: Modelled ^{131}I in dose rate in biota (above) and integration of internal and external dose rates over a one year period following release, divided by the time period considered for the accidental iodate pill release scenario (below)

In Fig. 7, a coordinate change is made to place the peak at T = zero in order to be able to follow-up the decaying dose profile over a 1-year period after release. The 1-year averaging cut-off time is arbitrary of course, but dose rates for different integration periods can be extrapolated if desired from the given figures.

The conclusion that the exposures to non-human biota have no radiological significance whatsoever can be determined by consultation of these figures. The single peak of water concentration, with a maximum of 654 Bq m^{-3} at the Molse Nete river at T = 218 days, and associated peak in sediment of a maximum of 0.011 Bq kg^{-1}, rapidly decay through the effects of delay and dilution. The maximal activity

concentration in biota is 218 Bq kg^{-1} for pelagic and benthic fish, with progressively lower concentrations of phytoplankton, macroalgae, crustacean and zooplankton (in that order).

3.2.2 Dose rates to the wildlife

The peak doses to the biota are of the order of 4.1×10^{-4} µGy h^{-1} for pelagic and benthic fish followed by phytoplankton > macroalgae > crustacean and mollusc > zooplankton (Fig. 8). The one-year time-averaged dose rates are very low. For internal exposure, they are 4×10^{-6} µGy h^{-1} for pelagic and benthic fish, decreasing to 9×10^{-7} µGy h^{-1} for mollusc, 6×10^{-7} µGy h^{-1} for crustacean, 5×10^{-7} µGy h^{-1} for phytoplankton, 4×10^{-7} µGy h^{-1} for macroalgae and 3×10^{-7} µGy h^{-1} for zooplankton. External dose rates are one order of magnitude lower with the most exposed group being macroalgae, with a very low dose rate of 5×10^{-8} µGy h^{-1}, and the least exposed group being pelagic fish at $< 2 \times 10^{-8}$ µGy h^{-1}. According to the methodology used here, such dose rates have no environmental significance. Hence, this type of accidental release scenario poses no significant risk to the environment.

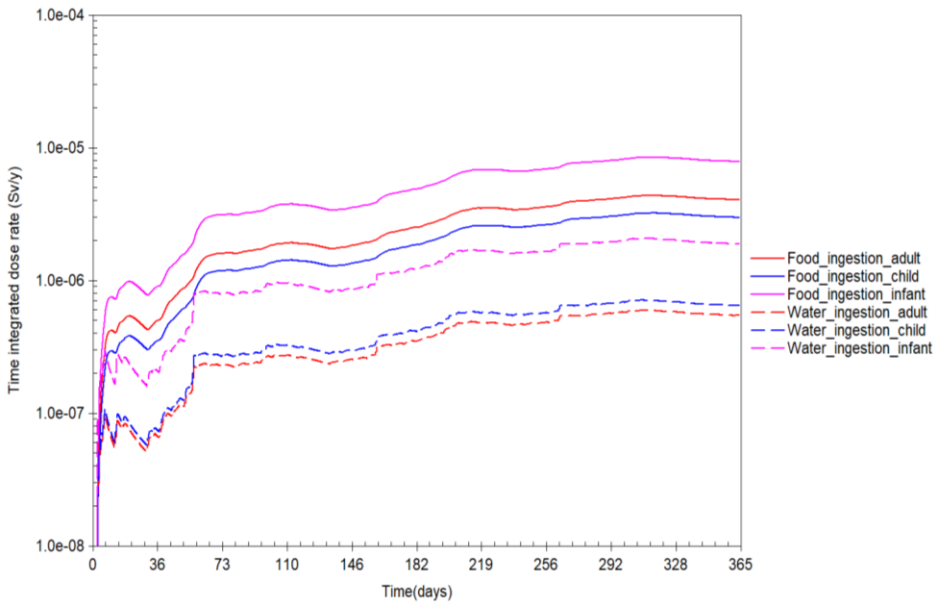

Figure 8: Time integrated ingestion dose rates for a routine discharge scenario, combining all radionuclides and food groups

3.3 Dose assessment for people

3.3.1 Doses to consumers and members of the public using the D-DAT dynamic model

The D-DAT model was used to calculate the dose rates to people of three age groups (adult, 10-year old and infant) arising from ingestion of water and of the biota, once consumption rates are set. The results are given in Fig. 9. D-DAT was also used to calculate the external dose rates from sediment (walking along the riverbank) and swimming exposure, as shown in Fig. 10.

The mean internal dose rates arising from ingestion of biota (mainly fish) at the Molse Nete river under the generic scenario considered over a 1-year period range between 2×10^{-3} mSv y^{-1} for child to 8×10^{-3} mSv y^{-1} for infant, the differences being caused by the different dose factors (reflecting the age-

dependent radiation sensitivity)and Belgian consumption rates for the different age groups. The mean internal dose rates from water ingestion are lower, ranging from 5×10^{-4} mSv y^{-1} for adult to 1×10^{-3} mSv y^{-1} for the infant.

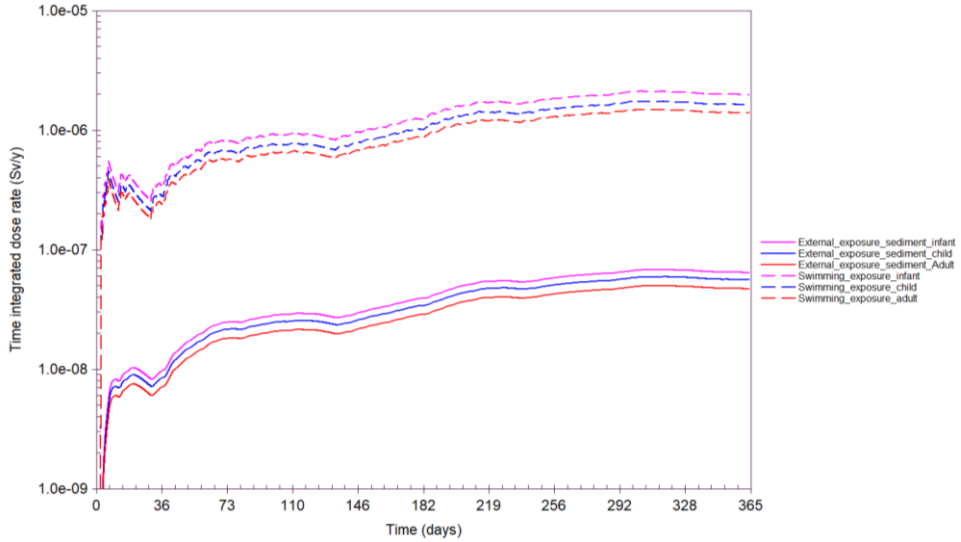

Figure 9: Time-integrated external exposure dose rates for a routine discharge scenario

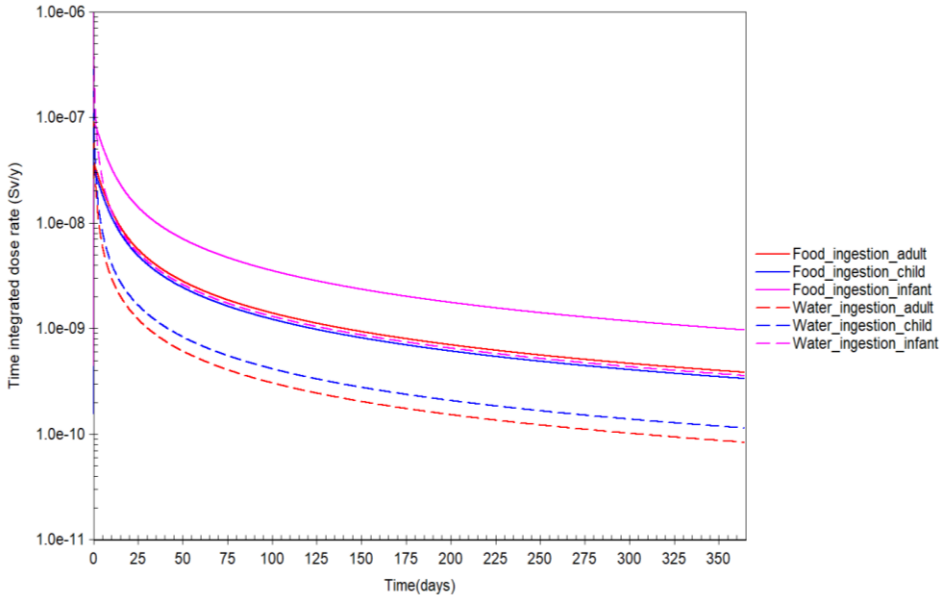

Figure 10: Time integrated ingestion dose rates for an accidental release of ^{131}I, combining all radionuclides and food groups

The above dose rates are much lower than the worldwide average annual radiation dose rate from exposure due to naturally occurring radiation sources, including radon, of 2.4 mSv (UNSCEAR, 2000) and very close to 10 µSv y^{-1} which is considered a trivial dose in terms of risk (IAEA, 2014). These exposures are of no radiological significance whatsoever.

External exposures have the same implications. Exposure to sediment walking along the riverbank ranges between 1.5 × 10^{-3} mSv y^{-1} for adult and 2 × 10^{-3} mSv y^{-1} for the infant, due to age-related differences in dose factors. The dose rates for external exposure due to swimming are much lower, between 5 × 10^{-5} and 6 × 10^{-5} mSv y^{-1}. These dose rates are essentially trivial.

The ^{131}I accident exposure scenario simulation gives significantly lower exposures compared with the routine scenario, as seen in Fig. 11. As we did in the assessment for biota, we placed the discharge at T = 0 to perform integration over a complete year (the averaging cut-off time is arbitrary but doses for different integration periods can be obtained from the Figures). Modelled average dose rates after 1 year are several orders of magnitude below the trivial dose rate of 10 µSv y^{-1} indicating no radiological significance whatsoever for this situation.

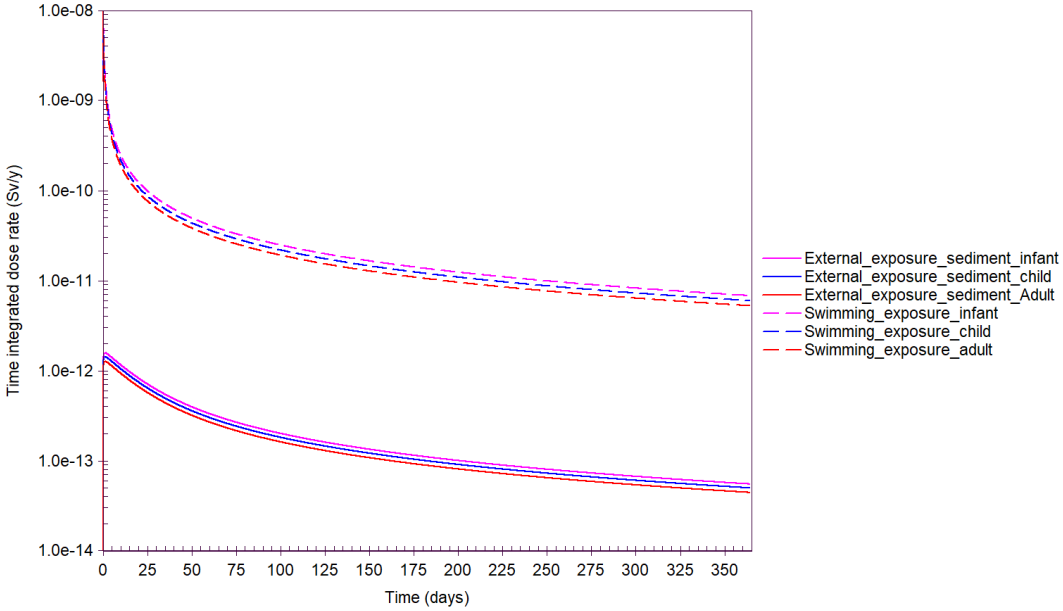

Figure 11: Time-integrated external exposure dose rates for an accidental release of ^{131}I

3.3.2 Doses to WTP workers and the public

3.3.2.1 Fractionation of radionuclides

We made calculations of the doses to workers and the public at a Mol-sited water treatment plant which, unlike the Leuven plant, does not make the pessimistic assumption that radionuclides are diverted to the watercourse, but rather assume that the plant is operational, drawing water from the river. The main source of uncertainty in the dose assessment is the plant removal efficiency, that is, fraction of activity concentration entering the plant that is retained and thus separated from the effluent. Hence, the first step was to arrive at an estimate for these efficiencies.

The Belgian radiation protection regulator FANC performed automatic gamma spectrometric measuring stations at both inlet and outlet of several WWTPs in Belgium. This would, in theory, enable calculation of the efficiencies. However, in practice, it is not straightforward to do so because the detection limit

was approximately 10^3 Bq m^{-3}, sufficient in principle to detect the main radionuclides associated with hospital discharges (^{99m}Tc, ^{131}I and ^{201}Tl), but not others like ^{18}F, ^{123}I, ^{153}Sm (Fiengo Pérez et al., 2022). The problem is compounded by not knowing the transit time of the different effluent fractions – for indicative purposes, a transition time of 15 hours is assumed for liquid effluent and 17 days are assumed for conditioned sewage sludge, which are treated for long enough to make them suitable for application to land (EA, 2022). All these factors can distort the calculation of the removal efficiencies, given the short half-life of the radionuclides involved.

We decided that it was preferable to source the (radionuclide-dependent) WWTP fractionation parameters, for which a reference was found in the literature (McDonnell, 2004). The first parameter required is the removal efficiency of the sewage processing, that is, the fraction not exiting the installation (in our study, we assume for simplicity that there is no loss due to decay during the few hours that the radioactivity is in the WTP except for ^{18}F, ^{99m}Tc and ^{123}I). The second parameter is the fraction of the initial effluent that ends-up in the produced sludge (incorporating estimated decay in all cases, meaning that the fraction of ^{18}F, ^{99m}Tc and ^{123}I is virtually zero). The data are given in Table 3. For ^{18}F and ^{123}I for which the reference gives no data, a 90% efficiency of removal for sewage is assumed by expert judgement, similar to ^{99m}Tc due to considerations of chemistry and fast decay. For ^{153}Sm, the same efficiency as for ^{201}Tl is assumed, based on similar considerations.

Table 3: Fractionation of radionuclides in sewage treatment plants

Radionuclide	Fraction in sewage	Fraction in sludge
^{18}F	0.9	0
^{99m}Tc	0.9	0
^{123}I	0.9	0
^{131}I	0.2	0.05
^{153}Sm	0.8	0.01
^{201}Tl	0.8	0.01

The assessment was performed using average concentrations because it is too complex now to do otherwise and the data are too limited to perform a dynamic modelling as was done for the non-human biota. Based on an assumption of 1% of the activity captured in the blocked sewer scenario and the input data, we arrived at average activity concentrations in the different plant fractions, as shown in Table 4.

Table 4: Average activity concentrations in the different plant fractions

Radionuclide	Average activity concentration (Bq m^{-3})			
	Blocked Sewer	Liquid phase	Sludge	River discharge
^{18}F	4.2E+04	3.8E+04	0.0E+00	4.3E+01
^{99m}Tc	8.3E+06	2.3E+06	0.0E+00	2.6E+03
^{123}I	2.9E+05	3.6E+04	0.0E+00	4.1E+01
^{131}I	2.0E+06	1.7E+04	3.0E+04	1.5E+02
^{153}Sm	1.1E+05	3.7E+03	1.4E+03	8.4E+00
^{201}Tl	3.1E+08	6.9E+06	2.5E+06	1.6E+04

3.3.2.2 Radiological exposures for the routine scenario

The radiation dose rates to general plant workers and sewer maintenance workers (repairing a blockage), are given in Table 5. It can be seen that the order of exposure is external gamma > ingestion > inhalation, and that dose rates due to maintenance work are an order of magnitude lower than for general work, mainly due to the lower occupancy rate. It can be seen also that the order of contribution by radionuclide to total exposure is ^{201}Tl > ^{99m}Tc > ^{18}F > ^{131}I > ^{123}I > ^{153}Sm. Given that ^{201}Tl is not a strong gamma emitter, with main gamma emissions of 70.8 keV (46.5%), 68.9 keV (27.4%) and 80.3 keV (20.5%), much of the resulting dose is likely reduced by shielding provided by walls, tanks and containers, leading to very low dose rates for workers. Nevertheless, the highest dose rate (plant worker, sum of all radionuclides) is 58 µSv y^{-1}, a small fraction of the dose from naturally occurring radiation sources, including radon, of 2.4 mSv (UNSCEAR, 2000). All maintenance worker doses, and the majority of doses to WWTP workers except the external gamma exposure to ^{99m}Tc and ^{201}Tl , are below the trivial dose level of 10 µSv y^{-1}.

Table 5: Dose rates to workers at the WWTP including general work and sewer maintenance

Radionuclide	Dose rate (mSv y^{-1})			
	Ingestion	Inhalation	Ext. Gamma	Totals
WWTP workers				
^{18}F	1.90E-08	2.50E-09	2.20E-03	2.20E-03
^{99m}Tc	5.00E-07	3.00E-08	1.40E-02	1.40E-02
^{123}I	7.60E-08	2.90E-09	2.80E-04	2.80E-04
^{131}I	1.00E-05	3.80E-07	1.00E-03	1.00E-03
^{153}Sm	3.80E-08	3.50E-09	1.80E-05	1.80E-05
^{201}Tl	8.90E-06	4.60E-07	4.00E-02	4.00E-02
Total	2.00E-05	8.80E-07	5.80E-02	5.80E-02
Maintenance workers				
^{18}F	8.30E-11	1.90E-12	9.90E-06	9.90E-06
^{99m}Tc	7.30E-09	7.80E-11	2.10E-04	2.10E-04
^{123}I	2.40E-09	1.70E-11	9.10E-06	9.10E-06
^{131}I	1.70E-06	1.10E-08	1.70E-04	1.70E-04
^{153}Sm	3.10E-09	5.20E-11	1.50E-06	1.50E-06
^{201}Tl	1.20E-06	1.10E-08	5.20E-03	5.20E-03
Total	2.90E-06	2.20E-08	5.60E-03	5.60E-03

Dose rates for the freshwater pathways are given in Table 6. The dose rates from drinking water, either WTP treated water or unfiltered river water at the release point, are of no radiological significance whatsoever. The fish ingestion and external gamma radiation doses in Table 7 provide a point of comparison with the doses for the same pathways as dynamically calculated by D-DAT. The main difference between the two is that the human dose calculator used here considers removal of a significant part of the radioactivity at the treatment plant entering at monthly averages, whereas D-DAT uses a dynamic calculation. The D-DAT dose rates are more conservative and should be used as primary results given that the model calculates radionuclide transfer dynamically.

Table 6: Dose rates for the freshwater pathways

Radionuclide	Dose rate (mSv y^{-1})			
	External gamma at the riverbank	Fish ingestion	Drinking WWTP treated water	Drinking unfiltered river water
^{18}F	2.60E-06	1.40E-08	9.20E-07	1.50E-09
^{99m}Tc	4.40E-04	3.60E-08	2.50E-05	4.10E-08
^{123}I	3.80E-04	1.60E-08	3.60E-06	6.20E-09
^{131}I	3.90E-03	6.40E-06	1.40E-03	2.40E-06
^{153}Sm	8.90E-03	2.00E-10	2.20E-07	4.60E-09
^{201}Tl	1.20E-01	3.60E-05	6.10E-04	1.10E-06
Total	1.30E-01	4.20E-05	2.00E-03	3.60E-06

The predicted concentrations for application of water in irrigation and resulting dose rates are shown in Table 7. Dose rates arising from use of WTP sludge as a ground fertiliser are shown in Table 8. Dose rates from use of sludge as fertiliser predominate over dose rates from irrigation, but in both cases, the total dose rates (7.1×10^{-5} and 2.9×10^{-6} mSv y^{-1}, respectively) are insignificant, being near the 10 µSv y^{-1} trivial dose level.

Table 7: Predicted concentrations and resulting dose rates for the irrigation pathway

Radionuclide	Concentrations (Bq kg^{-1})		Dose rates (mSv y^{-1})		
	Green Vegetables	Root Vegetables	Green Vegetables	Root Vegetables	Total
^{18}F	1.3E-04	6.2E-05	3.00E-10	3.50E-10	6.50E-10
^{99m}Tc	2.2E-01	5.7E-02	2.20E-07	1.50E-07	3.70E-07
^{123}I	1.3E-05	1.3E-05	1.20E-10	3.20E-10	4.50E-10
^{131}I	7.1E-04	7.1E-04	7.00E-07	1.80E-06	2.50E-06
^{153}Sm	1.9E-07	1.9E-07	6.40E-12	1.70E-11	2.30E-11
^{201}Tl	7.4E-03	1.4E-03	3.20E-08	1.50E-08	4.70E-08
Total	2.3E-01	5.9E-02	9.60E-07	2.00E-06	2.90E-06

Table 8: Activity concentration in sludge and dose rates arising from use of WWTP sludge as a ground fertiliser

Radionuclide	Concentration (Bq kg^{-1})	Dose rate (mSv y^{-1})
^{18}F	0.0E+00	0.0E+00
^{99m}Tc	0.0E+00	0.0E+00
^{123}I	0.0E+00	0.0E+00
^{131}I	3.0E+01	1.0E-05
^{153}Sm	1.4E+00	1.8E-08
^{201}Tl	2.5E+03	6.1E-05
Total	2.5E+03	7.1E-05

3.3.2.3 Radiological exposures for the accidental scenario

For the accidental scenario, this method of calculating dose, based on a time average, is not naturally suited for a assessing a single isolated pulsed discharge. The integration period is by its nature arbitrary. In order to derive the average dose rate over the integration period, there is a need to use the average

activity concentration in water for that period. The time integrated dose rate is calculated and divided by the number of days of the integration period. For consistency with what was done in the routine scenario, we select this period to be one year.

The calculated ^{131}I dose rates to regular and maintenance workers at the WWTP for a 1 year integration period are 4.8×10^{-7} and 7.9×10^{-8} mSv y^{-1}, respectively. For exposure to the public through the freshwater pathways, the dose rates from external gamma (riverbank occupancy), fish ingestion, drinking WWTP-processed water and drinking unfiltered river water are 1.8×10^{-6}, 2.9×10^{-9}, 6.3×10^{-7} and 1.1×10^{-9} mSv y^{-1}, respectively. Dose rates from ingesting green and root vegetables upon irrigation of farmland are 3.2×10^{-10} and 8.3×10^{-10} mSv y^{-1}, respectively. Finally, the dose rate arising from the use of WWTP sludge in agriculture is 4.6×10^{-9} mSv y^{-1}. All these dose rates are negligible, being significantly below the trivial dose level of 10 µSv y^{-1}.

4. CONCLUSIONS AND RECOMMENDATIONS

In this project, we have made a practical demonstration of an approach for the environmental impact assessment of radiopharmaceuticals released from medical facilities, considering simultaneously both humans and the non-human biota, and able to dynamically calculate dose rates to non-human freshwater biota for short-lived hospital-sourced radionuclides, based on the biokinetic model D-DAT. We have also developed a method to calculate doses to water treatment plant workers and from agricultural practices in equilibrium conditions over a 1-year integration period.

We generated a radiological source term for a conservative scenario of radionuclides at the Belgian Molse Nete River during the low-flow year 2018, to symbolise typical environmental conditions likely to reach a WWTP from a hospital. The radionuclides covered (^{18}F, ^{123}I, ^{131}I, ^{153}Sm, ^{99m}Tc and ^{201}Tl) are the only ones for which environmental monitoring data were available. Additionally, a spike release of one MBq of ^{131}I was used as a case for unplanned release, to simulate the accidental disposal of an iodate pill into the sewer system. Having trialled the approach with a generic source term, our method is now ready for use in realistic case studies.

All dose rates calculated in our maximising release scenario are low, even for the highly conservative scenario considered. In the case of biota, they do not exceed the ERICA predicted no effects dose rate of 10 µGy h^{-1}, meaning that no effects are expected at the population level for the fauna and flora in the Molse Nete River (and, by inference, in any other Belgian cases where the generalised concentrations of hospital-released radionuclides tend to be lower). For humans, the dose rates computed for the different exposure pathways are substantially below the 2.4 mSv y^{-1} public dose rate for all natural sources. In most cases, they are also below what is considered a trivial dose (10 µSv y^{-1}).

Nevertheless, it is not possible to state ”case closed”. It is necessary to continue to perform such assessments, since they are still infrequent due to the primary focus being on exposure to patients. In

addition, environmental exposures to medical radionuclides (and so discharges of radiopharmaceuticals in rivers) may increase with new nuclear therapies in the future. Moreover, occasional accidental discharges in European hospitals where higher concentrations are involved are not unheard of. There is a wider range of radionuclides to consider (e.g. ^{89}Zr, ^{90}Y, ^{99}Mo, ^{131}m, ^{133}Xe, $^{177, 177m}$Lu, $^{223, 226}$Ra, ^{225}Ac, and ^{227}Th), and there is a need to bring down uncertainty in model parameters. Along the way, there is a need to improve and standarise modelling methods, in order to be able to explicitly demonstrate to regulators, the public and the relevant stakeholders that people and the environment are adequately protected.

In line with the above, we make the following specific recommendations so that the screening approach used here can be improved. Firstly, we believe that significant radiopharmaceutical industries and hospitals should conduct and publish annually their own environmental radioactivity monitoring, just as the nuclear industry does. There is a knowledge gap here, and significantly we had to resort to build our source term upon one of the few WWTPs where monitoring data is available to make our assessment, and environmental release data from actual hospitals could generally not be found.

Secondly, we recommend to extend the assessment approach to other radionuclides, which necessitates biokinetic research to establish the transfer parameters of the relevant radionuclides in their relevant physico-chemical chemical form (speciation) for biota (Vives i Batlle et al., 2022), which in the present study had to be deduced for some radionuclides, based on a chemical analogue methodology and other proven extrapolation methods. This research could involve aquatic tank experiments with freshwater biota, aiming at establishing transfer parameters (concentration factors and biological half-lives of elimination) for the radionuclides for which the data are not available and had to be deduced by applying extrapolation methods, especially for ^{201}Tl.

Thirdly, there needs to be better knowledge of the *modus operandi* of water treatment plants to help better define the assessment scenario. We had to make certain (reasonably conservative) assumptions and simplifications to cover for a certain lack of generalisable plant process information. In order to reduce conservatism and minimise model conceptual uncertainties, there is need for actual knowledge of the retention/separation efficiencies of the different waste streams (water and sludge), as well as the basic working pattern (occupancy fractions) at the treatment plant in terms of worker hours per year spent between plant operation and plant maintenance. Other improvements needed include establishing the transit times of the different effluents to calculate accurately the relevant radionuclide decay factors, and also establishing what are the realistic shielding conditions for external beta and gamma exposure, which is especially important for ^{201}Tl as this radionuclide appears to dominate external exposure to workers.

Finally, there should be a move towards a unified European approach for dose assessment from medical radionuclides, possibly by further developing the modelling methodology that we have developed in the

present project, so that different member states can be in a position to perform and compare assessments of the impact of radiopharmaceuticals on people and the environment using a consistent methodology. We particularly recommend that the environmental impact assessment approach should be part of the development process of radionuclide treatments.

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