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Abbreviations

ABR	Abbreviation
ADD	Advection-Diffusion-Decay
AQUAFIN	Flemish Wastewater Treatment Company
AZ	Algemeen Ziekenhuis/General Hospital
DHI	Danish Hydraulic Institute
ECO-Lab	Water quality model
FANC AFCN	Federaal Agentschap voor Nucleaire Controle Agence fédérale de contrôle nucléaire
HIC	Hydrologisch Informatiecentrum/ Flanders Hydraulics
IRSN	L'Institut de Radioprotection et de Sûreté Nucléaire
MIKE 11	River model
SCK CEN	StudieCentrum voor Kernenergie of Centre d'étude de l'énergie Nucléaire
UZ	Universitair Ziekenhuis/ University Hospital
VMM	Vlaamse Milieumaatschappij/ Flemish Environmental Agency
WWTP	Wastewater Treatment Plant



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1 Introduction

Radiopharmaceuticals are broadly used for diagnostic purposes, for treatment of cancer and other deceases. However, independently of their benefits, the radioactive compounds must still be handled with care to protect patients, hospital personnel and the environment. Before their release, the radioactive effluents are collected in special tanks until acceptable activity levels for rivers are reached. Then, they are conveyed through the sewer system to a wastewater treatment plant (WWTP) for further standard 24-48 hr treatment before they are released into the river system.

The hospital's storage tanks and the post waste treatment are efficient and reliable methods for lowering the activity levels of the effluents. In Belgium, the Belgian Federal Agency for Nuclear Control (FANC) undertook a continuous monitoring of the radioactivity levels (gamma spectrometry) during 2012 to 2014 with the aim of verifying if the levels of the radioactive effluents were in-line with the demands of the environmental legislation. FANC installed portable measuring stations at the inlet and outlet of the WTPs to detect traces of radioactivity discharged by medical centers (and from patients who are treated and then sent home) in connection with cancer treatment, diagnosis and research. These measurements not only confirmed the levels were lower than the discharge limits but they also provided information about the source term composition and the release schedule.

Besides all the measures taken to minimize the radiological risks, it is important to keep in mind that, once in freshwater, radiopharmaceuticals (thought low radioactive) are not contained nor isolated from the environment. They come in contact with most forms of life via ingestion or/and other exposure pathways as result of the use of water for consumption, house holding, industrial production, agricultural, recreational purposes and as main support of the ecosystems. Moreover, wastewater treatment plants can be subject to maintenance or simple exceptional malfunction that could require bypassing the wastewater directly to receiving rivers. Under circumstances of unusual radioactive levels, it is necessary to understand and predict how these elements spread in water systems and through the biosphere, to be able to evaluate and mitigate the impact of these discharges on the environment and the public.

In this study, we focus on the estimation of activity concentration in rivers after accidental release scenarios, that will be used in the future for radiological impact assessments. For this, we incorporate the most relevant characteristics and conditions that could affect the fate and transport of radiopharmaceuticals. We take into account their half-lives, their distribution between solid and liquid phases, the volumes and activity levels, the discharge periodicity of the radioactive effluents and the flow regime of the receiving rivers.

In order to reach our objectives, this report is divided in the following chapters:

This introduction, Chapter 1. In Chapter 2, where we present the study area and we comment the particularities of the environment, the river network and water use in Belgium.

In Chapter 3, we provide a short discussion about the source term derived from the measurements done by the Belgian Federal Agency for Nuclear Control (FANC). We also propose an additional accident scenario not related to the malfunction of the containers or WWTP but from direct release of radiopharmaceuticals in the sewer system. Additional aspects related to the receiving rivers and the selection of the representative year for the simulation are discussed.

In Chapter 4, the transport model used for the simulation of the fate and transport of the radioactive effluents is presented and its accuracy is discussed. Chapter 5 presents the results of the dose calculation for water ingestion and exposure due to submersion in water. Finally, Chapter 6 summarizes and discusses the main results of this study.



2 Study case: Belgium

2.1 Population and Water use

In Belgium, the reduction of the of groundwater recharge zones due to urbanization has led to a depletion of aquifers and a more strict control and limitations on the use of this water source. Nowadays, it can be seen from the available official data that surface water sources are being used to satisfy the increasing water demands. However, during August-September, the discharge of the inland rivers decreases considerably and the Belgian government applies contingency measures related to the water extraction to guaranty the minimum flow rate required to sustain the wild life. About 40% of Belgium's annual freshwater availability is satisfied by rivers, two-thirds by groundwater with net precipitation accounting for the rest. The major aquifers are located in Wallonia. Flanders and Brussel are highly dependent on water flows from Wallonia from which they satisfy 40% and the 98% of their water demands respectively.

The population density (in units of people per km²) in Belgium, according to the latest information published by Statbel (the Belgian statistical office) on 1st January 2021, is presented in Table 2.1. The population density in the Flemish Region is more than two times the density of the combined population of the Walloon and Brussels regions. More details about the spatial distribution of the population are presented in Figure 2.1. Despite housing only 37% of the population, Wallonia alone supplies 55% of the country's water.

Approximately 46% of the total surface of the Flemish Region is used as agricultural land while 43% in the Walloon Region is destined for this purpose. In Flanders, the groundwater is the main source of water for agriculture. However, during the last decennia, it is observed that part of its water needs are being satisfied by surface water. Regarding the water use in industry, the main source originates from surface water.

Place of residence	Population on 1st January 2022	Density Inhabitants/km ²
Belgium	11,584,008	375
Flemish Region	6,698,876	488
Walloon Region and Brussels	3,662,495	216

Table 2.1:Population and population density in Belgium



Figure 2.1:Saptial distribution of the Belgian population (Source: Statbel, https://statbel.fgov.be)





Figure 2.2: Source of water for agriculture (Source VMM, https://www.vmm.be/data/milieudata)



Figure 2.3: Source of water for industry (Source VMM, https://www.vmm.be/data/milieudata)

From the total amount of water used in agriculture, almost 56% is destined to animal husbandry. In Table 2.2, the information corresponding to 2010 is presented. At the moment of the elaboration of this project updated information is not available.

Table 2.2:Distribution of the water per agricultural sector in Flanders in 2010 (Source: Department Agriculture and Fishery of Flanders, https://lv.vlaanderen.be/)

Use	Percentage
Dairy cattle	13%
Beef cattle	4%
Pig breeding	15%
Other horticultural companies	14%
Other farms (including poultry)	24%

During the last years, periods of drought during summer (August) have been more frequent. The low dilution capacity of the surface waters used for ingestion and recreation during the dry periods increases the health risk due the enhanced concentration of both radioactive and non-radioactive pollutants. During this period, constant monitoring has been performed by the relevant Belgian environmental agencies (such as FANC, VMM) and WWTPs to see if releases occurred could cause peaks of pollutant concentrations.

2.2 Points sources of radioactive pollutants

The most relevant watercourses in Belgium receive effluents from hospital facilities. These effluents are not directly discharged into water bodies, but instead they are subject to treatment in waste treatment facilities. Effluents from hospitals are treated in two phases. In the first phase, the effluents



are stored in tanks inside the facility until the activity concentration decays to levels that do not represent any significant risk for people. Then, in the second phase, the much reduced radioactive effluents are discharged in the sewerage and conducted to the WWTP, where they are subject to standard treatment. As a result of this process, the data and reports provided by FANC show that the activity concentration in the effluents released to watercourses is in general below or near detection limits. Figure 2.4 presents the distribution of medical facilities that use radiopharmaceuticals for different purposes and the main rivers that receive their discharges after treatment.



Figure 2.4: Hospitals (Red dots) and river network

2.3 Radiological monitoring of hospital's effluents and data availability

From 2012 to 2014, automatic measuring stations were installed by the authorities in some of treatment plants that receive wastewater containing radiopharmaceuticals (FANC, 2015). FANC plans to extend its measuring network to all relevant point sources and receiving WWTP inlets during the coming years.

FANC initially focused on plants who receive and treat discharges from the largest nuclear medical facilities. They placed two probes in the wastewater treatment plant: one measuring station at the inlet, after the wastewater screen and a second measuring station at the outlet. The measuring campaign lasted for approximately three months.

During this period, the measuring stations were permanently submerged under at least 1 meter of water to shield the probe against external irradiation of cosmic origin or other sources. This surrounding water volume makes possible the correctly determine the radioactivity transported by the wastewater. Moreover, in order to closely capture realistic conditions, the probe was placed in such a way that it does not disrupt the normal water flow.

The on-site measurements were cross validated and complemented with accurate low radioactive measurements done in collected samples of sludge and water carried out by the FANC's-approved laboratories for radioactivity in the environment (suitably BELAC-accredited - ISO 17025).

2.3.1 Location of the measurement points

In the first stage towards the implementation of a full network for the monitoring of the radioactive effluents from medical facilities, the following nuclear medical centers listed in Table 2.3 and presented



in Figure 2.5 were selected as pilot projects. These sites as shown in Figure 2.5, are located in some of the biggest cities of the country¹.



Figure 2.5: Location of the sampling points.

¹ Source: Statbel, https://statbel.fgov.be



WWTP Collector	Latitude	Longitude	Medical facilities in the WTP collector	
W/WTD1 Decelies	NEO 426026°	F4 F64410°	Hôpital St. Joseph	
W W IPI Roselles	1050.420020	E4.304419	CHU de Charleroi, Site Châtelet	
			Grand Hôpital de Charleroi,	
			Site Notre Dame	
			Nucleris	
			Nuclear Med	
WWTP Montignies-sur-Sambre	N50.403346°	E4.456611°	CHU de Charleroi, Site Hôpital Civil	
			La Transfusion Du Sang	
			Grand Hôpital de Charleroi, Site Reine Fabiola	
			CHU de Charleroi,	
			Site Polyclinique de la Madelaine	
			Prins Leopold Instituut	
			C7A Ziekonhuis, Compus Sint Vincentius	
			GZA Ziekennuis, Campus Sint-Vincentius	
WWTP Antwerpen-Zuid	N51.196774°	E4.373094°	AZ Monica, Campus Eeuwfeestkliniek	
			ZNA, Campus Middelheim	
			ZNA, Site Stuivenberg	
			UZ Leuven, Site Sint-Rafael / Sint-Pieter	
WWTP Leuven	N50.901431°	E4.712953°	UZ Leuven – Site Gasthuisberg	
			Heilig Hart Ziekenhuis	
			AZ Sint-Lucas & Volkskliniek	
			CEDRI	
			Universiteit Gent,	
			Faculteit Farmaceutische Wetenschappen	
WWTP Gent	N51.055452°	E3.684634°	Universiteit Gent, Campus-UZ-Site	
			IBA Pharma	
			AZ Maria Middelares	
			CRI	

Table 2.3: Selected points where radioactive effluents from medical facilities are collected

2.3.2 Equipment used and radionuclides identified

Automatic gamma spectrometric measuring stations were equipped with a LaBr₃ detector (Saphymo) connected to a multichannel analysis system. The detection limit was approximately 1Bql⁻¹. The main radionuclides associated with hospital discharges identified are presented in Table 2.4. The measurements show that, in the influents at the inlet of the WWTPs, Tc-99m and I-131 are often detected while F-18, I-123, Ir-192 and Sm-153 were occasionally detected or not depending of the WWTP. Unusually high activity levels of I-131 in the influent were also observed, this situation was investigated by FANC (FANC, 2015).

The activity levels in the effluents are mostly below the detection limit. This can be explained by the combination of three aspects (1) the storage of the effluents in tanks before their release into the sewerage, (2) the short half-life of the radio isotopes and (3) possibly due to adsorption onto the sludge; though for the latter, the measurements report non detectable activity in the sludge (FANC, 2015).

According to the measurements, the influents of the WWTP in Leuven (designed for a Population Equivalent PE=108000) carries in general the highest activity values, For WWTP Antwerp-Zuid (PE=171000) the activity levels are lower. A possible reason for this difference can be a combination of the activities released by hospitals, the traveling time between the hospital and the WWTP and the wastewater volumes received by the WWTP. The report provided by FANC corresponding to WWTP



Montignies-sur-Sambre and WTP1 Roselies, suggests that the activity concentrations measured at both stations could not reflect the real situation because during the sampling period, the operation of the medical facilities was limited. With regard to the activity series of WWTP Gent (PE=207000), an unusual peak for I-131 was observed. This situation could hypothetically correspond to an accidental release. FANC carried out an investigation of this case; but, based on measurements only, it is not possible to define the exact origin because the monitoring point at the inlet of the WWTP account for the combined contribution of all medical facilities and also the domestic releases.

The inter-comparison of the activity concentration measured in influents is presented in Figure 2.6. The magnitude of the activity concentrations in the y-axis is not shown here, due to restriction in the publication of this information. Nevertheless, it is our objective to provide an indication of the difference between the registries of the monitoring points by using a common scale. It is important to mention that, according to the calculations of FANC (FANC, 2015), the current releases do not represent a risk for people or the environment; but hospital discharges should be the subject of monitoring and regular control (FANC, 2015).

Radionuclide	Half-Life ²	Uses ³		
		Frequently used radioisotope in positron emission tomography (PET)		
1 10	1.0 11	Radiopharmaceuticals in both clinical and preclinical research.		
I-123	13.22 hr	Radiopharmaceutical diagnostic agent used for the evaluation of		
		The thyroid function and/or morphology.		
I-131	8.02 d	Treatment of thyroid gland disorders and cancer.		
Sm-153	1.93 d	Treatment of metastatic bone pain and bone cancer.		
Tc-99m	6.01 hr	Used to image the skeleton and heart muscle in particular and other organs		
TI-201	3.04 d	Radiopharmaceutical agent used in the diagnosis of		
		coronary artery disease and parathyroid hyperactivity		
I-125	59.38 d	Treatment of prostate cancer and ocular cancer.		
Lu-177	6.65 d	Treatment of tumors.		
lr-192	73.83 d	Cancer treatment, including cancer of the lungs, head, neck,		
		Mouth, tongue and throat, and treatment of vascular constriction.		

Table 2.4: Radionuclides identified during the measuring camping.

² https://periodictable.com/

³ NRG, 2008: www.nrg-nl.com





Figure 2.6:Inter-comparison of the radioactive influents.



3 Scenario definition

3.1 Description of the current situation

In section 2, we discussed some aspects about the water use and radiological releases. There, it was also mentioned that the radioactive effluents from medical facilities are at or below detection limits when released into rivers. This relevant piece of information proves the efficiency of the wastewater treatment measures followed to minimize the radiological impact. The success of the treatment relies on a two-step system: (1) storage of radioactive effluent in decay tanks (2) standard treatment in WWTP. The failure of both is very unlikely. However, under extraordinary circumstances such as maintenance or expansion (especially in the WWTP) the radioactive effluents could be directly released into watercourses. Consequently, this situation needs to be included in the radiological impact assessment.

In this study, we focus on the simulation of the fate and transport of radioactive pollutants in surface water systems. The main objectives are: (1) to determine the activity levels that could be observed after a release and (2) to simulate the progressive movement of the pollutants along the river network. Activity levels can be measured, but the measurement points are limited to selected points and to specific times. Mathematical models give the possibility to estimate the activity concentrations at several locations in order to complement the monitoring point. Radionuclide transport models are useful tools when predictions and risk assessments are required. Here, we use one such model for the computation of the activity profiles corresponding to the isotopes mentioned before. The values obtained in this study will be used later on in the project for the estimation of radiological impact on human and biota.

The data required for transport modelling can be classified into three groups (1) hydrometric information, (2) radiometric data and (3) physical description of the rivers (i.e. bathymetry and plan form). In Europe, each member state must deploy real time monitoring networks for the collection of information related to the status of water bodies. For the specific case of Belgium, a quite dense hydrometric network exists with records that cover more than 30 years. Unfortunately, the network for the monitoring of radioactive liquid releases is less extended and restricted to the surroundings of the nuclear power installations. Thus, for the purposes of this study we are limited to use the information collected by FANC during its monitoring campaign at the WWTPs. Regarding the physical description of the rivers, the collection of bathymetric data is labour intensive, costly and mostly limited to the most relevant rivers. In the next section, we discuss further the data availability.

3.2 Radiological effluents of medical facilities

The effluents released by medical facilities are collected by different wastewater treatment plants before their discharge into rivers. Some of these WWTPs collect effluents of several hospitals. As discussed in section 2.2, detailed information about these releases is currently available just for some WWTP. FANC selected five pilot monitoring points (FANC, 2015). Here, it was decided not to restrict the scope of this study to these release points and their corresponding receiving rivers, but to try to include other point sources as well. Nevertheless, the inclusion of additional sources depends on the data available for the computation of the activity concentration, a situation that will be discussed in section 3.3.

The restriction on the availability of time series of radioactive effluents can be partially eased if a generic release time series is used. The use of a generic source term has several advantages. For example, for rivers where the data required for the transport simulation is available but the information about the effluents is unknown, the use of a generic source makes possible the computation of realistic activity concentration levels as well as the radiological impact assessment along this watercourse. Moreover, some rivers included in the pilot monitoring receive effluents at several locations. There, the use of a generic source makes it possible to consider the radiological



impact by the summation of most of the sources. This allows achieving completeness using the data available.

Information provided to us by FANC gives an overview of the range of the total expected activity concentrations and the release schedule of the medical facilities inside the selected zones. Hospital's specific data is not available; however, the information provided by the regulatory authorities is sufficient for the radiological impact assessment.

Since our objective is to evaluate the impact of a scenario where the radioactive effluents are released into rivers without the treatment at the WWTP, it was decided to use the radioactive influents at the WTP Leuven as an example case, given that UZ Leuven beside being the biggest hospital is also the most important center 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 the same levels as a result of the growing demand for medical treatments of this nature.

The activity concentrations at the inlet of WWTP Leuven are reported by the unit of volume of wastewater. These volumes are periodic and change over the day. Thus the measurements reflect the variability in the dilution of the activity concentration. The monitoring period covers approximately 4 months and took place during the summer (Figure 2.6). The highest activity in the rivers occurs during the periods where the river discharge is very low. This situation is very common during the month of August. In principle, it would be sufficient to just calculate the activity concentration in the river for the driest historical period, but we are 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 the full year.

In Figure 2.6, a peak in the activity concentration of I-131 at Monitoring site WWTP Gent is observed. This information can be used to model an occasional spike of radioactivity (although we are not in any way indicating that this particular event was an accidental situation). To evaluate this scenario conservatively, low dilution in the receiving river is considered, which means that, the accident happens when the rivers carry the lowest discharge.

3.3 Receiving rivers

The data required for the modelling of the fate and transport of radionuclides includes (besides the source term) hydrometric data and bathymetric information. In Belgium, the hydrometric data collection is managed by the regional environmental agencies: The Flemish Environmental Agency (VMM) and Flanders Hydraulics Research (HIC). In Wallonia the responsible organisation is the Direction générale opérationnelle de la Mobilité et des Voies hydrauliques. These agencies are also the main providers of bathymetric information. The information provided can be summarized as follows:

- Time series of water levels and discharge
- River cross sections or LIDAR surveys

Both data sets are used by the hydrodynamic river model to represent the flow and the river transport patterns, based on historical observations. The required information was available for the following rivers:

- 1. Meuse/Maas River (BE, NL)
- 2. Samber/Sambre River (BE)
- 3. Nete River
- 4. Dender River
- 5. Dijle River
- 6. Vunt River
- 7. Zenne River
- 8. Aa River



- 9. Grote Nete River
- 10. Kleine Nete River
- 11. Ruppel River
- 12. Albert Canal
- 13. Scheldt River

Data also exists for other rivers such as Demer and Leie, but the information could not be found in time for this report. The rivers listed above received effluents from medical facilities. The release points included in this assessment are presented in Figure 3.1. There, the red dots represent the outlets of the different WWTPs and the red crosses signify the hospitals connected to this plant.



Figure 3.1: Location of the hospitals (Red cross) and release points of the receiver WWTP (Red dots)

3.4 Selection of the representative year for the calculation of activity concentrations

FANC's monitoring campaign took place in 2012, 2013 and 2014. Since we decided to use a generic source term, we are not restricted to select one of these years for the simulation. Instead, we select the year based on standard international water quality indicators. That means, using the available hydrometric data to determine among all the years, which year fulfils the 7-day, 10-year low flow (Q7, 10) statistic. 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. The calculation shows that the summer of 2018 was one of the driest summers in the last decennia. The series used for the calculation are presented Figure 3.2. The frequency curve corresponding to the 7-d minimum flow is presented in Figure 3.3. The 7-d low flow corresponding to 10 years is identified by the dashed line and the discharge obtained is comparable to the 7-d low flow observed in 2018.





Figure 3.2: Time series of 7-d low flow per each considered year



Figure 3.3: Frequency-Discharge plot (dot line represent the T=10 y frequency)

From the statistical analysis presented in Figure 3.2, it can be seen that the number of low flows around the minimum flow observed in 2018 is higher in comparison to other years. This is another criterion that supports the selection of 2018 as the representative year for this study.



4 Modelling the fate and transport of radionuclides in river systems

The fate and transport of radionuclides and other pollutants is controlled by flow regime of the water system. Physically this is interpreted from variables related to the water flow such as velocity and water level. With regards to the process of transport, three components can be identified. The Advection Mechanism represents the transport of the pollutant with the water flow without including any change in its composition. The Dispersion Process considers the spreading of the pollutant due to molecular diffusion and turbulent mixing. Finally, the transformation of the pollutants, decay and distribution between phases is included also in the model. A vast corpus of literature devoted to the explanation of the state-of-the-art model for transport models (commonly known as water quality models) is available. More details about this can be found in Chapra (1997), Ji (2007), Fiengo Pérez (2016) and in the MIKE 11 model's description provided by the Danish Hydraulic Institute (DHI, 2017b).

4.1 Hydrodynamic modelling

Before modelling the transport, a correct simulation of the river hydrodynamics is required. More details about the state-of-the-art of this discipline can be found at Hamrick (2007) and Zheleznyak (2003). The simulation of water level and velocity demands the river bathymetry and measurements of water levels and velocity at the endpoints of the river model. It is common that water discharges are available instead of velocity. This situation does not represent a problem during modelling.

In Belgium, a significant number of registries from automatic measuring stations is available. Their location has been selected by the authority in such a way that they are placed at points that comprise most of the extension of the river network of the country. This allows modelling a complete river system in many cases. As example, Figure 4.1 illustrates how a river model is defined by selecting stations located at the end points of the area of interest (dots designated as Boundary Stations) and the use of other stations placed along the river for the verification of the simulations (green dots labelled as Validation Stations).



Figure 4.1: Illustration of the setup of the river model. (Blue line: river alignment)

The hydrometric information helps us (1) to keep the connection between the selected part of the river network and the rest of the river system not included in the simulation, and (2) to verify the calculations. The bathymetric data provides information about the shape of the river, the slope, contractions, expansions among other relevant characteristics that influence the water movement. An example of the type of information extracted from the bathymetric data is presented in Figure 4.2. The left panel presents details about the topography survey where the elevations across de river at regular distance intervals are collected. The middle and right panels show the processed information to be used during the modelling. In the right panel, we show an example of river cross section introduced into the model.





Figure 4.2: Bathymetric information example

The model used for the simulation is DHI MIKE 11. This model is widely used for several purposes such as flooding simulation, hydraulic structure design and water quality. This model solves the full, dynamic, 1-D shallow-water equations in unidirectional form, also called Saint Venant equations (Hervouet, 2007).

Once all the previously mentioned information is included in the model, validation of the result is required. Hydrodynamic river models include the friction between water and the rivers bed and walls. A widely used mathematical formulation for friction is the Manning-Strickler Formulation. This formula includes the influence of the water levels and the characteristics of the riverbed (e.g. presence of stones, vegetation). The last is represented by the Strickler Coefficient $M[m^{1/3}/s]$. Here this value was selected from past studies and adjusted based on inter-comparison information between measurements and simulations. The values adopted for the simulation are between 30 to 35 m^{1/3}/s (Arcement, 1989) (AMINAL, 2002). The methodology described here for the model set up was applied for each of the 13 watercourses mentioned in section 3.3. The results of the simulation and the inter-comparison with the observations are presented in Appendix A.

4.2 Conceptualization of the transport model

For the simulation of the fate and transport of radionuclides, besides Advection-Dispersion, Decay and Sorption-Desorption processes are also considered. Radionuclides are transported in dissolved and particulate forms. The last represents the radionuclides attached to the suspended sediment in the water column. The distribution of the total activity between forms is determined by the sorption-desorption process. In water quality modelling, this process is largely simplified and represented by the distribution coefficient K_d (defined as the ratio of equilibrium concentrations of a dissolved radionuclide and an aqueous phase) and the sorption-desorption rates. More information about these parameters can be found in IAEA (2010). The K_d is maybe the most important factor and the most meaningful during the model conceptualization, as the larger the distribution coefficient is, the more radionuclides will be adsorbed on the particulate form. The distribution between forms plays a significant role in the way of transport. While the dissolved form is governed simply by advection diffusion and decay, the transport of the particulate form is also governed by the sediment transport dynamics. This difference represents an increase in modelling complexity because sediment dynamics includes erosion and sedimentation mechanism.

The sorption-desorption process requires time before the equilibrium between phases is reached. The range of time required to achieve the equilibrium lies within a range of hours to days. Some of the radionuclides measured during the monitoring campaign (Table 2.4) have a very short half-life and in principle, it can be considered (1) that they remain in dissolved phase and (2) that the decay rate is so fast that their accumulation in the riverbed sediments, as result of sedimentation, can be safely ignored. The values of K_d obtained mainly from the database of the ERICA tool (Brown et al., 2008) for the radionuclides with half-life larger than a 6 hr are presented in Table 4.1. The K_d values show that the Sorption-Desorption process does not play a significant role in the transport. Therefore, the modelling approach can be limited to advection, diffusion and decay only. This situation brings



additional advantages such as less restrictive computational demands, increase in the extension of the river network and the long-term simulation, in our case one year.

Radionuclide	Speciation category (based on Kd)	Kd (L kg-1)	Kd (m³ kg⁻¹)
Tc-99m	Highly soluble	2.59E+01	2.59E-02
I-123	Moderately insoluble	1.43E+05	1.43E+02
I-131	Moderately insoluble	1.43E+05	1.43E+02
TI-201	Generally soluble. TI has multiple oxidation states, hence variable sorption affinity. TI(III) sorbs more strongly than TI(I).	7.14E+01	7.14E-02

Table 4.1: Partition coefficient for selected elements

4.2.1 Radionuclide transport modelling

The Advection-Diffusion-Decay equation (ADD) is implemented in tandem with the hydrodynamic model (Ji, 2007). The water quality model is implemented in the DHI ECO Lab framework (DHI, 2017a). It uses calculated discharge and water levels as input and simulates the transport in water. The ADD equation requires defining the diffusion coefficient in order to represent the spreading of the pollutant. However, this coefficient needs to be defined either by using tracer experiments or by model calibration. Unfortunately, in this study, neither site specific nor measurements of activity concentration were available for the selected rivers. Although FANC has some radiological monitoring points, this information is not sufficient for the determination of this parameter. Instead, we decided to base on ranges of magnitude reported in the literature and on knowledge gained during past and ongoing research in order to define appropriate values. The most common values adopted for the diffusion coefficient are 5 m²/s for streams and a range between 10 m²/s to 1000 m²/s for big rivers and estuaries (Chapra, 1997). Here, 5 m²/s was used for the inland rivers and 20 m²/s-500 m²/s were used for the Scheldt River.

The data availability restriction related to activity concentration does not only limit the possibility of the site-specific determination of the required model parameters but also the verification of our simulations. For that reason, though an important effort was done to guarantee a correct simulation of the river hydrodynamics and a (over) conservative release scenario was selected; the results are still theoretical values subject to verification. This exemplifies the unavoidable limitations of this type of study.

A site specific validation of the model results is not possible. However, from an ongoing research collaboration between SCK CEN (Belgium) and IRSN (France) on the Rhône river, it is clear that SCK CEN's model is capable to represent the transport in complex rivers systems. These results can be used as proof of the reliability of the modelling approach adopted in this study. More details of this evaluation can be found in Appendix B.

In this section, the results of the computation of the activity concentration for the selected rivers are presented and discussed. The time series for specific points are available but are not reported because the amount of data is extensive and does not contribute directly to the interpretation of the fate and transport of radiopharmaceuticals. In its place, envelopes of maximum instantaneous activity concentration are shown as illustration of the variation of the activity concentration along the rivers. In Appendix C the activity concentrations in the rivers at the corresponding release points are presented.

The instantaneous maximum envelope must not be confused with a release profile. An envelope presents the maximum activity at each calculation point, while a release profile shows how the pollutant is transported by the water flow. In this study, we simulated release pulses similar to those shown in Figure 2.6 and discussed in section 3.2. Therefore, the envelope represents just the concentration at a specific instant when the highest activity passed through each section of the river. For the sake of comparison, the envelopes of each radionuclide were normalized. This allows



visualizing how the maximum activity concentrations change with distance not only due to dilution and diffusion but also due to radionuclide decay.

The envelopes show the variation of peak activity concentrations along the river. The distance presented in the x-axis has as reference the most upstream point of the branch considered in the model. In some cases, the release point coincides with the starting point of the branch, while in others, the release point is located downstream. For this reason, the values of the envelopes do not start always at x = 0. The envelope map of activity concentrations for I-131 is also presented to provide a complete spatial overview of the possible releases.

The modelling of the fate and transport of radiopharmaceuticals is based on the following assumptions:

- 1. The generic source term is based on the time series of activity concentration measured at the inlet of the WWTP in Leuven.
- 2. The hospital effluents from the decay tanks are bypassed directly into the rivers.
- 3. The year 2018 was selected as reference year based on the Q7, 10 criteria.
- 4. A release of 1 MBq of I-131 directly into the sewer system is considered as a potential accident scenario.

4.2.1.1 Aa and Kleine Nete Rivers

The Aa and the Kleine Nete are important tributaries of the Nete River. Along these rivers, two hospitals exist where radiopharmaceuticals are used. Both are inland rivers with very limited tidal influence; however, this influence grows downstream of their confluence. Figure 4.3 presents the envelope map of activity concentrations for I-131 corresponding to routine releases for both rivers and the release points. The release points are represented by white dots.



Figure 4.3: Envelope of instantaneous activity of I-131 estimated for the Aa and Kleine Nete Rivers (White dots show the release points)

This map is just presented for the routine releases of I-131 to provide a spatial reference that helps with the identification of the watercourses, the release points and to provide an idea about the spreading of the pollutant in the river system. In Figure 4.4, the envelopes are presented. Each curve represents the maximum activity concentrations in the river network corresponding to each radioisotope considered in the generic source term corresponding to routine and accidental release of



I-131 discussed in section 3.2. The scaling factor for each curve is provided in the legend. The distance presented in the x axis is referred to the most upstream point of the branch considered in the model. In the Aa River, the highest activity concentrations are registered up to 4 km downstream the release point, whilst for the Kleine Nete, they extend less than 2 km from the release point.



Figure 4.4: Envelope of maximum activity concentrations estimated for the Aa (Top) and Kleine Nete (Bottom) Rivers

4.2.1.2 Nete River

The Nete River is a historically polluted river that has recovered gradually its good ecological status during the last decade. However, this river crosses several important cities and agricultural land. It receives discharges from several industries and is a source of water for agriculture and animal husbandry. During high flow events, flooding happens at different places along the rivers. Several hospitals are placed inside the river catchment; However, the two medical facilities selected for this river release their effluents in the tributaries of small and medium size. Figure 4.5 shows the distribution of activity concentrations corresponding to routine releases of I-131. The white dots represent the release points. The activity envelopes along the first 70 Km of the collector Molse Nete-Grote Nete are presented in Figure 4.6. In Figure 4.7, 25 Km of the activity profile along the collector Kleine Nete-Nete River is shown.





Figure 4.5: Envelope of instantaneous activity of I-131 estimated for the Nete River



Figure 4.6: Envelope of maximum activity concentrations estimated for the collector Molse Nete-Grote Nete River



Figure 4.7: Envelope of maximum activity concentrations estimated for the collector Kleine Nete-Nete River

Thought not presented in Figure 4.5 the activity concentrations originating from the medical facilities placed in the Aa and the upper Kleine Nete rivers (discussed in section 4.2.1.1) are also included with the aim of keep connection between the upper part and lower part of the of the Nete River. The envelopes of the Grote Nete show the dilution after the confluence of the Molse Nete with the Grote



Nete and after the confluence of the Grote Nete with the Grote Laak. Similarly, the envelopes of the Kleine Nete show the dilution after its confluence with the Grote Nete.

4.2.1.3 Dijle River

The Dijle River is an inland river that crosses the City of Leuven where the biggest medical facility of the country is located. Leuven is an university town, whereupon the wastewater effluents drastically diminish during the summer holidays, thus during this period less dilution of radioactive effluents is observed. Moreover, Leuven's university hospital Gasthuisberg is one of the most important medical centres for treatment of cancer in the country. In Figure 4.8 and Figure 4.9 the spatial distribution of the maximum activity concentrations after routine releases of I-131 and the envelopes for the other elements including the accidental I-131 release are presented respectively. The envelopes show the dilution after the confluence of the Dijle with the Vunt and the Demer rivers respectively.



Figure 4.8: Envelope of instantaneous activity of I-131 estimated for the Dijle River



Figure 4.9: Envelope of maximum activity concentrations estimated for the Dijle River

4.2.1.4 Scheldt River

The Scheldt River is the most important river of Belgium. It is a tidal river and the final collector of the rivers discussed before. It crosses the most populated zones of the country and in its plains several industries and the port of Antwerp are located. It has its origins in France and its main tributaries in Belgium are the Rupel, the Durme, Dender, and the Oude Scheldt. The Dender River is highly regulated



by locks placed along the river for navigation purposes. The Oude Scheldt connects the Scheldt and the channel system of the City of Gent. The flow between both is controlled by weirs located at the east of Gent. The channel systems of Gent is complex and the details required for its implementation are not reported as this goes beyond the objectives of this report.

Because its tributaries cross almost all-important cities of Flanders, the pollutants end in this river. The rivers discussed previously are part of the Scheldt River. Though possible, the complexities and the computational demands make it unpractical to simulate the complete system. In all cases, the activity concentrations rapidly decrease towards the outlet of the tributary and little mass is transfer to the Scheldt. Under this situation, it is possible to simulate this river independently. Nevertheless, in case of continuous releases, this situation does not hold true and the radioactive releases from the tributaries must be included.

There are several medical centres in the drainage area of this river; however, we restrict this study to those which release their effluents to the FANC's monitoring points (WWTP Antwerp and WWTP Gent) and to the General hospitals of Aalst and Geraardsbergen located at the Dender River. The release point of the WWTP Gent is located on the west site of the city. Its outlet does not discharge directly to the Scheldt but in one of the branches of the channel system of Gent. From that point onwards, the effluents are transported through the channel systems. As previously mentioned, the Channels of Gent are not part of the model; nonetheless, in order to include this source, it was assumed that the releases at WWTP Gent are identical downstream from the regulating weir. This is an overestimation of the activity concentration, thus the results at this point can be both merely indicative within a degree of conservatism, or considered as a possible range of values at the real release point. The activity distribution and the envelopes are presented in Figure 4.10, Figure 4.11, Figure 4.12 and Figure 4.13.

Three regions are identified along the Scheldt: (1) the upper Scheldt, (2) middle Scheldt and (3) the lower Scheldt. This division is based on the dimensions of the river section. As the river section width increases, the magnitude of transversal currents grows and it is not possible anymore to neglect them. Therefore, a 2-D modelling approach that considers the longitudinal and transversal currents is required. Here, the 1-D approach is valid for the upper and middle region where the assumption of homogeneous distribution of concentration across the river section is probable. However, the result for the lower part (near Antwerp) would not be valid due to well know variations in concentration across the river section observed for other variables such as salinity. Nonetheless, the assumptions adopted in terms of release scenario guarantee the conservativeness of our estimations.





Figure 4.10: Envelope of instantaneous activity of I-131 estimated for the Scheldt River



Figure 4.11: Envelope of maximum activity concentrations estimated for the Oude Scheldt River



Figure 4.12: Envelope of maximum activity concentrations estimated for the Dender River





Figure 4.13: Envelope of maximum activity concentrations estimated for the Scheldt River

The envelopes of the Oude Scheldt River show that the radiopharmaceuticals do not spread far away from the discharge point due to the tidal influence. In the case of the Dender, the envelopes indicate that the activity concentrations decrease with the distance as was observed in other rivers such as the Grote Nete, the Aa and the Dijle. Finally, the envelopes of the Scheldt show the influence of the tides on the transport. In the inland rivers, the order of the normalized envelopes (top Sm-153 and bottom F-18) is almost the same. This is not the case for the tidal rivers.

4.2.1.5 Sambre River

The Sambre River is one of the most important tributaries of the Meuse River; it starts in France and flows through the City of Charleroi in the Walloon region of Belgium. This is an inland river free of any tidal influence. It is regulated for navigation purposes by means of lock gates. The Sambre flows into the Meuse and the later feeds the Albert Canal through to The Netherlands. The estimation of the activity concentrations done in this study is not restricted to the Sambre, but instead it includes the Belgian Meuse, part of the Dutch Meuse (up to Maastricht in The Netherlands) the Albert Canal and the Flemish Channels. The selection of the model domain is justified by two facts, namely (1) the Meuse provides 40 % of the total drinking water for Flanders and (2) it is a transboundary river subject to international agreements related to water use. Two of the monitoring points of FANC are located in the Sambre. The map of activity concentrations of I-131 and the envelopes are presented in Figure 4.14 and Figure 4.15.





Figure 4.14: Envelope of instantaneous activity of I-131 estimated for the Sambre River



Figure 4.15: Envelope of maximum activity concentrations estimated for the Sambre River

The envelopes of the Sambre show a propagation of the activity concentrations in the downstream direction in a similar fashion as observed in the other inland rivers discussed before.



5 Preliminary dose calculation

To determine the impact of the discharges of radiopharmaceutical in rivers, the dose rate due to exposure and ingestion can be used to provide some context to the simulations of activity concentrations. For that, the standard procedure suggested by the ICRP (1991) and ICRP (2012) and EPA (Eckerman, 1993) is followed. The scenario for the dose computation is defined as follows:

The population groups considered are:

- Group 1: from 0-1 years
- Group 2: from 1-2 years
- Group 3: from 2-7 years
- Group 4: from 7-12 years
- Group 5: from 12-17 years
- Group 6: Adults

The dose due to exposure considers the dose received while the subject is submerged in water for a period of eight consecutive hours exactly at the discharge point. As suggested by the EPA just the 1% of the total dose to skin is considered (Eckerman, 1993). For the dose due to ingestion of contaminated water, we assume direct water ingestion (inadvertent ingestion during submersion or premeditated). The volumes assumed for each population group and are presented in Table 5.1.

Table 5.1: Assumption for ingestion and exposure scenario

Description	0-1y	1-2y	2-7y	7-12y	12-17y	Adult
Water ingestion [liters]	0.1	0.25	0.5	1	1	1
Time submersion [hours]	8	8	8	8	8	8

In Table 5.2, Table 5.3 and Table 5.4, we present the dose conversion factors used for the calculation of the total effective dose due to ingestion, submersion and the dose conversion factor for dose to skin respectively. The EPA's dose conversion factors only provide values for adults, but in Belgium, these factors are scaled to compute the dose to other population groups. The scaling factors are presented in Table 5.5.

Group	Ingestion					
Group	0-1y	1-2y	2-7у	7-12y	12-17y	Adult
F-18	7.2E-10	3E-10	1.5E-10	9.1E-11	6.2E-11	4.9E-11
I-123	5.2E-09	1.9E-09	1.1E-09	4.9E-10	3.3E-10	2.1E-10
I-131	4.8E-07	1.8E-07	1E-07	5.2E-08	3.4E-08	2.2E-08
Sm-153	3.4E-09	5.4E-09	2.7E-09	1.6E-09	9.2E-10	7.4E-10
Tc-99m	3E-10	1.3E-10	7.2E-11	4.3E-11	2.8E-11	2.2E-11
TI-201	5.4E-10	5.5E-10	2.9E-10	1.8E-10	1.2E-10	9.5E-11

Table 5.2: Dose conversion factors for ingestion (Sv Bq⁻¹)

Ground	Submersion						
Group	0-1y	1-2y	2-7y	7-12y	12-17y	Adult	
F-18	1.41E-16	1.41E-16	1.41E-16	1.22E-16	1.22E-16	1.07E-16	
I-123	2.13E-17	2.13E-17	2.13E-17	1.84E-17	1.84E-17	1.61E-17	
I-131	5.25E-17	5.25E-17	5.25E-17	4.54E-17	4.54E-17	3.98E-17	
Sm-153	6.8E-18	6.8E-18	6.8E-18	5.87E-18	5.87E-18	5.15E-18	
Tc-99m	1.73E-17	1.73E-17	1.73E-17	1.49E-17	1.49E-17	1.31E-17	
TI-201	1.12E-17	1.12E-17	1.12E-17	9.7E-18	9.7E-18	8.51E-18	

Table 5.3: Dose conversion factors for submersion dose (Sv $Bq^{-1}m^3$)

Table 5.4: Dose conversion factors for skin dose (Sv $Bq^{-1}m^3$)

Crown	Skin						
Group	0-1y	1-2y	2-7у	7-12y	12-17y	Adult	
F-18	1.81E-16	1.81E-16	1.81E-16	1.56E-16	1.56E-16	1.37E-16	
I-123	2.69E-17	2.69E-17	2.69E-17	2.33E-17	2.33E-17	2.04E-17	
I-131	7.3E-17	7.3E-17	7.3E-17	6.3E-17	6.3E-17	5.53E-17	
Sm-153	2.51E-17	2.51E-17	2.51E-17	2.17E-17	2.17E-17	1.9E-17	
Tc-99m	2.06E-17	2.06E-17	2.06E-17	1.78E-17	1.78E-17	1.56E-17	
TI-201	1.43E-17	1.43E-17	1.43E-17	1.23E-17	1.23E-17	1.08E-17	

Table 5.5: Scaling factors from adult to other groups

Scaling factors for external exposure and skin dose							
From/to	0-1y	1-2y	2-7у	7-12y	12-17y	Adult	
Adult	1.32	1.32	1.32	1.14	1.14	1	

Dose limits allowed in Belgium are based on European directives and on the recommendations of international organisations. As result, the effective dose limit is 1 mSv per year. This value excludes natural radiation or radiation used for medical purposes. The European directive of 1998 on drinking water stipulates that the total annual dose resulting from the ingestion of drinking water must not exceed 0.1 mSv per year (FANC, 2020). Additional information about the quality of water intended for human consumption can be found in the COUNCIL DIRECTIVE 98/83/EC of 3 November 1998⁴.

The range of doses estimated in this study for routine releases are between 3.17E-07 mSv/y and 2.46E-03 mSv/y; while for accidental release scenario are between 3.56E-10 mSv/y and 8.81E-06 mSv/y. The range of doses for each river is presented in Table 5.6 and Table 5.7. Based on the order of magnitude of the dose, three groups can be identified, (1) the Aa, Molse Nete and Kleine Nete, (2) the Grote Nete, Dender and Dijle and (3) The Sambre and the Scheldt. This is expected because the rivers in each group have similar flow rate at the release point of the WWTP, as presented in Figure 5.1, especially during the low flows period.

⁴ https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A31998L0083



Table 5.6: Range of doses corresponding to routine releases

Routine Release mSv/y						
River Name	Min	Max				
Aa	1.41E-03	2.46E-03				
Molse Nete	1.07E-03	1.97E-03				
Oude Schelde	8.98E-04	1.62E-03				
Kleine Nete	1.04E-03	1.56E-03				
Grote Nete	4.66E-04	8.87E-04				
Dender Geraardsbergen	4.12E-04	7.80E-04				
Dender Alst	3.69E-04	6.91E-04				
Dijle	1.63E-04	2.89E-04				
Sambre Roselies	4.85E-05	8.95E-05				
Sambre Montignies	4.18E-05	7.63E-05				
Scheldt	3.17E-07	5.68E-07				

Table 5.7: Range of doses corresponding to accidental releases

Accidental Release I-131 mSv/y						
River Name	Min	Max				
Aa	3.83E-06	8.81E-06				
Kleine Nete	3.66E-06	8.42E-06				
Molse Nete	2.20E-06	5.06E-06				
Oude Schelde	1.24E-06	2.85E-06				
Grote Nete	7.44E-07	1.71E-06				
Dender Geraardsbergen	6.64E-07	1.53E-06				
Dender Alst	6.05E-07	1.39E-06				
Dijle	2.68E-07	6.17E-07				
Sambre Roselies	1.32E-07	3.05E-07				
Sambre Montignies	8.32E-08	1.92E-07				
Scheldt	3.56E-10	8.21E-10				



Figure 5.1: Summary of river flow rates at the outlet of the WWTP

The magnitude of the doses calculated here are at least three orders of magnitude bellow the dose limit of 1 mSv y⁻¹. That means that the release of radiopharmaceuticals has no radiological significance whatsoever for the pathways considered. Moreover, the release and the exposure scenarios selected



are unlikely to occur, especially if the measurements done by FANC, where the activity levels at the outlet of the WWTP are around the detection limits, are taken as reference.

More details related to the calculation of the dose for the different groups can be in Appendix D. A more refined estimate of dose and the associated doses to non-human biota in various exposure scenarios (for internal exposure, the present estimation comprises only water ingestion) will be presented in more detailed radiological impact assessment that will follow the present deliverable.



6 Conclusions and recommendations

The activity concentration in rivers can be determined directly at the release point of the WWTP, but it is also important to know how far from the release point the impact of the radioactive release spreads. This makes it possible to evaluate the risk associated to the use of the water of the receiving river for consumption, agriculture and recreation at other locations in the system. The water quality models used for the simulation of the distribution of activity concentrations along rivers must be capable to also represent the temporal patterns of high and low flows also known as the river's flow regime. The relation between water flow and pollutant transport demands to simulate the first as accurately as possibly within the model limitations and the quality of the hydrometric variables in order to predict reliable concentrations of pollutants in rivers. Water quality models are site specific, that means, the parametrization and model setup reflect the characteristics observed on-site. However, as happens in Belgium, several types of rivers (i.e. inland rivers, navigable rivers and tidal rivers) receive radioactive releases from medical facilities and spread the pollutants in different ways. In order to include as many influencing factors as possible, several different rivers were included in this research. The quality of their prediction of the river's regime were evaluated and in the large majority of the cases, the results were quite satisfactory.

It was possible to estimate the activity concentration at and downstream the release points. According to the measurements done by FANC at its pilot monitoring sites, the activity levels in wastewater after treatment in the WWTP are rather low or bellow detection limits. This shows that the treatment of radioactive effluents is efficient. However, though unlikely, direct releases from hospitals to rivers need to be taken into account in order to prove that people and the environment are adequately protected at any time. For this aim, it was assumed a scenario where, due to maintenance in the WWTP, the hospital effluents from the decay tanks are bypassed directly into the rivers. The activity concentration time series measured at the inlet of the WWTP Leuven were used as generic source term and a year where the 7Q10 (i.e. the 7-d low flow that would be expected to occur every 10 years) was evidenced, was selected for the simulation of the river flows. Additionally, a spike release was considered as an illustration for an unplanned release. It considers the release of 1 MBq of I-131 directly into the sewer system.

To illustrate the spatial distribution of the predicted maximum activity concentration envelope, the map corresponding to the I-131 envelope was presented. The highest concentration happens at the release point of the WWTP into the river because for short-lived radiopharmaceuticals as I-131 (half-life in the range of hours), the activity concentrations decrease rapidly with distance. In the case of tidal rivers, the pollutant spreads both upstream and downstream the release point. For the case of the Lower Scheldt River near the city of Antwerp, the approach followed here may not be sufficient and a 2-D modelling approach would be required. In wide rivers and estuaries, longitudinal and transversal circulation patterns are observed and the assumption of uniform velocity and concentration across the river section is not valid. However, the magnitude of the releases and the location of the release points make it possible to still use the 1-D approach for the medium and upper part of the Scheldt River.

The effective dose due to ingestion and external exposure were calculated. The exposure scenario assumes that the person is submerged in water at the discharge point during the most penalizing consecutive eight hours and ingest water directly from the river. Based on the standard scenarios used in Belgium for radiological impact assessments, this represents a very conservative scenario with low probability of occurrence. The doses estimated show that for routine releases the maximum value is at least tree orders of magnitude lower than the maximum allowed limit. For accidental releases, the maximum activity is six orders of magnitude lower.

The results presented here will be used for a more detailed and realist radiological impact assessment for human and biota.



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Appendix A. Verification of the simulations of the hydrodynamic model

The hydrodynamic model requires defining the boundary conditions at the endpoints of the model. For this, time series of discharge and water levels are required; however, runoff flows into the river along the river surface and there is a constant exchange of water between the river and the groundwater. In addition to the natural hydrological cycle, water is extracted at different points along the rivers. These flow exchanges need to be accounted for in order to simulate properly the water levels and flow rates. Here, a mass balance analysis was performed by using records from the stations located at the start, the end and inside the model domain. For the Dijle River, the verification of water levels and flow was not possible because an intermediate station does not exist. Nevertheless, the proximity of the outlet of the WWTP Leuven at the start of the model domain minimized the influence of the water fluxes not taken into account. For the Scheldt River, only measurements of water level are available at several points. The results are presented in this section. In the majority of the cases, the model was capable to represent accurately both water levels and discharges.





Figure A- 1: Verification of the discharges and water levels for the Grote Nete River at Geel-Zammel Hulshout Station



Figure A- 2: Verification of the discharges and water levels for the Grote Nete River at Hulshout













Figure A- 5: Verification of the water levels for the Scheldt River at Kallo (August)



Figure A- 6: Verification of the water levels for the Scheldt River at Liefkenshoek (August)



Figure A- 7: Verification of the water levels for the Scheldt River at Melle (August)







Figure A-9: Verification of the water levels for the Scheldt River at Sint Amands (August)

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Figure A- 10: Verification of the water levels for the Scheldt River at Temse (August)



Figure A-11: Verification of the water levels for the Scheldt River at Wetteren (August)



Figure A-12: Verification of the water levels for the Scheldt River at Dendermonde (August)





Figure A- 13: Verification of discharges for the Sambre and the Meuse at Salzines, Huy and Amay Erembodegem Station



Figure A- 14: Verification of discharges for the Dender at Erembodegem



Appendix B. Verification of the simulations of the transport model

The validation of the transport model for the rivers selected in this study was not possible during the present study due to lack of information. In order to provide an idea of the performance of the SCK CEN's model implemented in the DHI MIKE 11-ECO Lab framework, the results obtained during an ongoing model evaluation in collaboration with the IRSN (France) for the Rhône River are presented. The Rhône River model has an total length of 330 Km. The river has several diversion channels, dams with movable gates that rises the water to allow the generation of electricity by small hydroelectric power plants and four nuclear power plants. The model simulations of the river's hydrodynamics were evaluated at three points. The results are presented in Figure B- 1. The transport model was verified four points and the results are presented in Figure B- 2.



Figure B-1: Verification of flow rates of the Rhône River





Figure B- 2: Verification of activity concentrations of tritium at several distances from the release point (Normalized values)

The comparison between measurements and simulations illustrates the complexity of the system. Thought the model is able to represent the main flow rate trends, the influence of the operation of the gates is noticeable. The Rhône river has several loop branches that extend for several kilometres in some cases. The water division is regulated by dams. This is an important influence in the water and pollutant transport. Unfortunately, the operation rules are not available and a fixed position was assumed for the different dam's dates during the simulation. Consequently, the determination of the flow paths is still in progress. Nevertheless, given the complexity of the river system, the length of the simulation domain (approx. 330 Km) and the data limitations, our predictions of flow rate and activity concentrations are close to the observations. The difference between simulations and observations for the case of activity concentration is attributable to the influence of the dams and flow paths. Nonetheless, the difference of the arrival time is the order of hours and the magnitude of the activity is in line with the observations, bringing confidence to the use of our transport model for the present project.



Appendix C. Activity concentrations in rivers

In this appendix the activity concentrations in rivers calculated at the release points are presented. As expected the highest concentrations happen during the driest months (July to September).



Figure C- 1: Activity concentration in the Aa River at the outlet of the WWTP Turnhout







Figure C- 2: Activity concentration in the Dender River at the outlet of the WWTP Aalst



Activities River Dender Geraardsbergen



Figure C- 3: Activity concentration in the Aa River at the outlet of the WWTP Geraardsbergen





Figure C- 4: Activity concentration in the Dijle River at the outlet of the WWTP Leuven



Activities River Grote Nete



Figure C- 5: Activity concentration in the Grote Nete River at the outlet of the WWTP Geel



Activities River Kleine Nete



Figure C- 6: Activity concentration in the Kleine Nete River at the outlet of the WWTP Herentals



Activities River Molse Nete



Figure C- 7: Activity concentration in the Molse Nete River at the outlet of the WWTP Mol







Figure C-8: Activity concentration in the Oude Scheldt River at the outlet of the WWTP Gent (Downstream lock)



Activities River Sambre Montignies



Figure C- 9: Activity concentration in the Sambre River at the outlet of the WWTP Motignies-sur-Sambre



Activities River Sambre Roselies



Figure C- 10: Activity concentration in the Sambre River at the outlet of the WWTP Roselies





Figure C- 11: Activity concentration in the Scheldt River at the outlet of the WWTP Antwerp-Zuid



Appendix D. Dose calculation

Here the results of the dose rates calculated for different groups is presented. This is a preliminary calculation. A more detailed and exhaustive evaluation of the dose to human and biota will be provided in a forthcoming report. The information presented here was used to provide a way to explain the meaning of the activity concentrations released to the selected rivers in terms of dose rate.

	River Name: Molse Nete						
		Rout	ine Release				
Dose	Ingestion [mSv y-1]	Dose Submersion [mSv y-1]	Dose Skin [mSv y-1]	Total [mSv y-1]	Observation(<=1mSv y ⁻¹)		
0-1y	1.24E-03	5.63E-04	7.12E-06	1.81E-03	Ok		
1-2y	1.26E-03	5.63E-04	7.12E-06	1.83E-03	Ok		
2-7y	1.39E-03	5.63E-04	7.12E-06	1.96E-03	Ok		
7-12y	1.47E-03	4.86E-04	6.15E-06	1.97E-03	Ok		
12-17y	9.65E-04	4.86E-04	6.15E-06	1.46E-03	Ok		
Adult	6.43E-04	4.26E-04	5.39E-06	1.07E-03	Ok		
		Accidenta	al Release I-131				
Dose	Ingestion [mSv y-1]	Dose Submersion [mSv y-1]	Dose Skin [mSv y-1]	Total [mSv y-1]	Observation(<=1mSv y ⁻¹)		
0-1y	4.55E-06	1.43E-07	1.99E-09	4.70E-06	Ok		
1-2y	4.27E-06	1.43E-07	1.99E-09	4.41E-06	Ok		
2-7y	4.74E-06	1.43E-07	1.99E-09	4.89E-06	Ok		
7-12y	4.93E-06	1.24E-07	1.72E-09	5.06E-06	Ok		
12-17y	3.22E-06	1.24E-07	1.72E-09	3.35E-06	Ok		
Adult	2.09E-06	1.09E-07	1.51E-09	2.20E-06	Ok		

Table D- 1: Dose rate i	n the Molse Nete	River at the outle	t of WWTP Mol
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Table D- 2: Dose rate in the Grote Nete River at the outlet of WWTP Geel

	River Name: Grote Nete						
		Rout	ine Release				
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y-1]	Dose Skin [mSv y⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)		
0-1y	6.05E-04	2.10E-04	2.66E-06	8.17E-04	Ok		
1-2y	6.02E-04	2.10E-04	2.66E-06	8.15E-04	Ok		
2-7y	6.66E-04	2.10E-04	2.66E-06	8.79E-04	Ok		
7-12y	7.04E-04	1.81E-04	2.30E-06	8.87E-04	Ok		
12-17y	4.61E-04	1.81E-04	2.30E-06	6.44E-04	Ok		
Adult	3.05E-04	1.59E-04	2.02E-06	4.66E-04	Ok		
		Accident	al Release I-131				
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y-1]	Dose Skin [mSv y-1]	Total [mSv y ⁻¹]	Observation(<=1mSv y-1)		
0-1y	1.54E-06	4.86E-08	6.75E-10	1.59E-06	Ok		
1-2y	1.45E-06	4.86E-08	6.75E-10	1.49E-06	Ok		
2-7y	1.61E-06	4.86E-08	6.75E-10	1.66E-06	Ok		
7-12y	1.67E-06	4.20E-08	5.83E-10	1.71E-06	Ok		
12-17y	1.09E-06	4.20E-08	5.83E-10	1.13E-06	Ok		
Adult	7.07E-07	3.68E-08	5.12E-10	7.44E-07	Ok		



	River Name: Sambre Roselies						
		Rout	ine Release				
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)		
0-1y	5.72E-05	2.42E-05	3.07E-07	8.17E-05	Ok		
1-2y	5.83E-05	2.42E-05	3.07E-07	8.28E-05	Ok		
2-7y	6.44E-05	2.42E-05	3.07E-07	8.89E-05	Ok		
7-12y	6.84E-05	2.09E-05	2.65E-07	8.95E-05	Ok		
12-17y	4.48E-05	2.09E-05	2.65E-07	6.60E-05	Ok		
Adult	2.99E-05	1.83E-05	2.33E-07	4.85E-05	Ok		
		Accidenta	al Release I-131				
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)		
0-1y	2.74E-07	8.65E-09	1.20E-10	2.83E-07	Ok		
1-2y	2.57E-07	8.65E-09	1.20E-10	2.66E-07	Ok		
2-7y	2.86E-07	8.65E-09	1.20E-10	2.95E-07	Ok		
7-12y	2.97E-07	7.47E-09	1.04E-10	3.05E-07	Ok		
12-17y	1.94E-07	7.47E-09	1.04E-10	2.02E-07	Ok		
Adult	1.26E-07	6.55E-09	9.10E-11	1.32E-07	Ok		

Table D- 3: Dose rate in the Sambre River at the outlet of WWTP Roselies

Table D-4: Dose rate in the Sambre River at the	e outlet of WWTP	Sambre Montignies-sur So	ambre
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River Name: Sambre Montignies					
		Rout	ine Release		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	4.75E-05	2.15E-05	2.73E-07	6.94E-05	Ok
1-2y	4.89E-05	2.15E-05	2.73E-07	7.07E-05	Ok
2-7y	5.40E-05	2.15E-05	2.73E-07	7.58E-05	Ok
7-12y	5.75E-05	1.86E-05	2.36E-07	7.63E-05	Ok
12-17y	3.77E-05	1.86E-05	2.36E-07	5.65E-05	Ok
Adult	2.53E-05	1.63E-05	2.07E-07	4.18E-05	Ok
		Accidenta	al Release I-131		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	1.72E-07	5.44E-09	7.55E-11	1.78E-07	Ok
1-2y	1.62E-07	5.44E-09	7.55E-11	1.67E-07	Ok
2-7y	1.80E-07	5.44E-09	7.55E-11	1.85E-07	Ok
7-12y	1.87E-07	4.69E-09	6.52E-11	1.92E-07	Ok
12-17y	1.22E-07	4.69E-09	6.52E-11	1.27E-07	Ok
Adult	7.90E-08	4.12E-09	5.72E-11	8.32E-08	Ok



River Name: Aa					
		Rout	ine Release		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	1.37E-03	8.63E-04	1.09E-05	2.25E-03	Ok
1-2y	1.44E-03	8.63E-04	1.09E-05	2.31E-03	Ok
2-7y	1.58E-03	8.63E-04	1.09E-05	2.46E-03	Ok
7-12y	1.69E-03	7.46E-04	9.41E-06	2.45E-03	Ok
12-17y	1.11E-03	7.46E-04	9.41E-06	1.86E-03	Ok
Adult	7.48E-04	6.54E-04	8.25E-06	1.41E-03	Ok
		Accidenta	al Release I-131		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	7.93E-06	2.50E-07	3.47E-09	8.19E-06	Ok
1-2y	7.44E-06	2.50E-07	3.47E-09	7.69E-06	Ok
2-7y	8.26E-06	2.50E-07	3.47E-09	8.52E-06	Ok
7-12y	8.59E-06	2.16E-07	3.00E-09	8.81E-06	Ok
12-17y	5.62E-06	2.16E-07	3.00E-09	5.84E-06	Ok
Adult	3.64E-06	1.89E-07	2.63E-09	3.83E-06	Ok

Table D- 5: Dose rate in the Aa River at the outlet of WWTP Turnhout

Table D- 6: Dose	e rate in the Klein	e Nete River at th	e outlet of WWTP	Herentals
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River Name: Kleine Nete					
		Rout	ine Release		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	4.28E-04	9.05E-04	1.15E-05	1.34E-03	Ok
1-2y	5.98E-04	9.05E-04	1.15E-05	1.51E-03	Ok
2-7y	6.46E-04	9.05E-04	1.15E-05	1.56E-03	Ok
7-12y	7.32E-04	7.81E-04	9.91E-06	1.52E-03	Ok
12-17y	4.78E-04	7.81E-04	9.91E-06	1.27E-03	Ok
Adult	3.48E-04	6.85E-04	8.70E-06	1.04E-03	Ok
		Accidenta	al Release I-131		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	7.58E-06	2.39E-07	3.32E-09	7.83E-06	Ok
1-2y	7.11E-06	2.39E-07	3.32E-09	7.35E-06	Ok
2-7y	7.90E-06	2.39E-07	3.32E-09	8.14E-06	Ok
7-12y	8.22E-06	2.06E-07	2.87E-09	8.42E-06	Ok
12-17y	5.37E-06	2.06E-07	2.87E-09	5.58E-06	Ok
Adult	3.48E-06	1.81E-07	2.52E-09	3.66E-06	Ok



River Name: Dijle					
		Rout	ine Release		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y-1]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	1.72E-04	9.35E-05	1.18E-06	2.66E-04	Ok
1-2y	1.76E-04	9.35E-05	1.18E-06	2.71E-04	Ok
2-7y	1.95E-04	9.35E-05	1.18E-06	2.89E-04	Ok
7-12y	2.07E-04	8.07E-05	1.02E-06	2.89E-04	Ok
12-17y	1.36E-04	8.07E-05	1.02E-06	2.17E-04	Ok
Adult	9.10E-05	7.08E-05	8.93E-07	1.63E-04	Ok
		Accidenta	al Release I-131		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y-1]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	5.56E-07	1.75E-08	2.43E-10	5.73E-07	Ok
1-2y	5.21E-07	1.75E-08	2.43E-10	5.39E-07	Ok
2-7y	5.79E-07	1.75E-08	2.43E-10	5.97E-07	Ok
7-12y	6.02E-07	1.51E-08	2.10E-10	6.17E-07	Ok
12-17y	3.94E-07	1.51E-08	2.10E-10	4.09E-07	Ok
Adult	2.55E-07	1.33E-08	1.84E-10	2.68E-07	Ok

Table D-7: Dose rate in the Dijle River at the outlet of WWTP Leuven

Table D- 8: Dose	rate in the	Scheldt at the	e outlet of	WWTP Antwerp-	Sud

River Name: Scheldt					
		Rout	ine Release		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	3.40E-07	1.74E-07	2.21E-09	5.16E-07	Ok
1-2y	3.53E-07	1.74E-07	2.21E-09	5.29E-07	Ok
2-7y	3.89E-07	1.74E-07	2.21E-09	5.66E-07	Ok
7-12y	4.16E-07	1.50E-07	1.91E-09	5.68E-07	Ok
12-17y	2.72E-07	1.50E-07	1.91E-09	4.25E-07	Ok
Adult	1.83E-07	1.32E-07	1.67E-09	3.17E-07	Ok
		Accidenta	al Release I-131		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y-1]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	7.39E-10	2.33E-11	3.24E-13	7.62E-10	Ok
1-2y	6.93E-10	2.33E-11	3.24E-13	7.16E-10	Ok
2-7y	7.70E-10	2.33E-11	3.24E-13	7.93E-10	Ok
7-12y	8.00E-10	2.01E-11	2.79E-13	8.21E-10	Ok
12-17y	5.23E-10	2.01E-11	2.79E-13	5.44E-10	Ok
Adult	3.39E-10	1.76E-11	2.45E-13	3.56E-10	Ok



River Name: Oude Schelde					
		Rout	ine Release		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y-1]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	9.84E-04	4.91E-04	6.18E-06	1.48E-03	Ok
1-2y	1.01E-03	4.91E-04	6.18E-06	1.51E-03	Ok
2-7y	1.11E-03	4.91E-04	6.18E-06	1.61E-03	Ok
7-12y	1.19E-03	4.24E-04	5.34E-06	1.62E-03	Ok
12-17y	7.78E-04	4.24E-04	5.34E-06	1.21E-03	Ok
Adult	5.22E-04	3.72E-04	4.68E-06	8.98E-04	Ok
		Accidenta	al Release I-131		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	2.56E-06	8.08E-08	1.12E-09	2.65E-06	Ok
1-2y	2.40E-06	8.08E-08	1.12E-09	2.49E-06	Ok
2-7y	2.67E-06	8.08E-08	1.12E-09	2.75E-06	Ok
7-12y	2.78E-06	6.98E-08	9.70E-10	2.85E-06	Ok
12-17y	1.82E-06	6.98E-08	9.70E-10	1.89E-06	Ok
Adult	1.18E-06	6.12E-08	8.51E-10	1.24E-06	Ok

Table D-9: Dose rate in the Scheldt River downstream the Lock of Gent (Proxy for the dose at the WWTP Gent)

Table D- 10: Dose rate in the Dender River at the	e outlet of WWTP Geraardsbe	ergen
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River Name: Dender Geraardsbergen					
		Rout	ine Release		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	5.24E-04	1.89E-04	2.40E-06	7.16E-04	Ok
1-2y	5.26E-04	1.89E-04	2.40E-06	7.17E-04	Ok
2-7y	5.82E-04	1.89E-04	2.40E-06	7.73E-04	Ok
7-12y	6.15E-04	1.63E-04	2.07E-06	7.80E-04	Ok
12-17y	4.03E-04	1.63E-04	2.07E-06	5.68E-04	Ok
Adult	2.68E-04	1.43E-04	1.82E-06	4.12E-04	Ok
		Accidenta	al Release I-131		
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)
0-1y	1.38E-06	4.34E-08	6.03E-10	1.42E-06	Ok
1-2y	1.29E-06	4.34E-08	6.03E-10	1.33E-06	Ok
2-7y	1.43E-06	4.34E-08	6.03E-10	1.48E-06	Ok
7-12y	1.49E-06	3.75E-08	5.21E-10	1.53E-06	Ok
12-17y	9.75E-07	3.75E-08	5.21E-10	1.01E-06	Ok
Adult	6.31E-07	3.29E-08	4.57E-10	6.64E-07	Ok



River Name: Dender Aalst							
	Routine Release						
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)		
0-1y	4.56E-04	1.76E-04	2.23E-06	6.34E-04	Ok		
1-2y	4.60E-04	1.76E-04	2.23E-06	6.37E-04	Ok		
2-7y	5.08E-04	1.76E-04	2.23E-06	6.86E-04	Ok		
7-12y	5.38E-04	1.52E-04	1.92E-06	6.91E-04	Ok		
12-17y	3.52E-04	1.52E-04	1.92E-06	5.06E-04	Ok		
Adult	2.34E-04	1.33E-04	1.69E-06	3.69E-04	Ok		
		Accidenta	al Release I-131				
Dose	Ingestion [mSv y ⁻¹]	Dose Submersion [mSv y ⁻¹]	Dose Skin [mSv y ⁻¹]	Total [mSv y ⁻¹]	Observation(<=1mSv y ⁻¹)		
0-1y	1.25E-06	3.96E-08	5.50E-10	1.29E-06	Ok		
1-2y	1.18E-06	3.96E-08	5.50E-10	1.22E-06	Ok		
2-7y	1.31E-06	3.96E-08	5.50E-10	1.35E-06	Ok		
7-12y	1.36E-06	3.42E-08	4.75E-10	1.39E-06	Ok		
12-17y	8.89E-07	3.42E-08	4.75E-10	9.23E-07	Ok		
Adult	5.75E-07	3.00E-08	4.16E-10	6.05E-07	Ok		

Table D- 11: Dose rate in the Dender River at the outlet of WWTP Aalst