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A dynamic model for assessing radiological consequences of routine releases in the Loire river: Parameterisation and uncertainty/sensitivity analysis

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Abstract

A dynamic model for assessing the transfer of several radionuclides (^{58}Co, ^{60}Co, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{54}Mn and ^{131}I) in a food-chain was applied on the Loire river, where 14 nuclear power plants situated on five different sites operate. The model considers the following potential exposure pathways: (i) transfer of radionuclides through the aquatic food chain and the subsequent internal exposure of humans due to ingestion of contaminated water and/or fish; (ii) use of river water for agricultural purposes (irrigation), transfer of radionuclides through the terrestrial food chain and the subsequent internal exposure of humans due to ingestion of contaminated foodstuffs; (iii) internal exposure due to inhalation of dust originating from resuspension of contaminated soil particles; (iv) external exposure from radionuclides present in the river or deposited on the river sediments or the soil. For each of the parameters introduced in this model, a probability density function, allowing further

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uncertainty and sensitivity analysis, was proposed. Uncertainty/sensitivity analysis were performed to: (i) compare calculations to empirical data; (ii) determine a confidence interval for the mean annual dose to critical groups; and (iii) identify the parameters responsible for the uncertainty and subsequent research priorities.

Keywords: Loire river; Nuclear power plants; Radionuclides; Dose assessment; Uncertainty/sensitivity

1. Introduction

Nuclear power plants which are situated along a river routinely release gamma emitters (essentially ^{58}Co, ^{60}Co, ^{110m}Ag, ^{131}I, ^{137}Cs, ^{134}Cs and ^{54}Mn) in freshwater. In order to evaluate the radiological impact of such releases on reference (or critical) groups, it is important to identify and simulate the potential exposure pathways that could lead to an increase of radiological doses:

1. Transfer of radionuclides through the aquatic food chain and the subsequent internal exposure of humans due to ingestion of contaminated water and/or fish.
2. Use of river water for agricultural purposes (irrigation), transfer of radionuclides through the terrestrial food chain and the subsequent internal exposure of humans due to ingestion of contaminated foodstuffs.
3. Internal exposure due to inhalation of dust originating from resuspension of contaminated soil particles.
4. External exposure from radionuclides present in the river or deposited on the river sediments or the soil.

Routine releases from nuclear power plants are not continuous because they are related to specific operations within the reactors. Consequently, it may be expected that transfers of radionuclides in the environment are not equilibrated. Therefore, time dependency of the transfer processes in conjunction with a seasonality of agricultural practices must be taken into account. This is the reason why a new model, called OURSON (French acronym for Tool for Environmental and Health Risk assessment), was developed to consider: (1) the many potential exposure pathways as verified through a review of the literature; (2) when possible, transfer processes were dynamically modelled; and (3) seasonality in growing cycles of crops and agricultural practices were considered.

However, parameters describing transfer of radionuclides in the aquatic and terrestrial environments are generally uncertain, and, as a consequence, it is important to know the uncertainty on the final result of the simulation due to the propagation of parametric uncertainties, and to identify which parameters are responsible for this uncertainty.

The aim of this paper is to synthesise studies conducted to determine the radiological consequences of routine releases in freshwater by NPPs present in the Loire watershed. The paper is then subdivided in three main parts:

1. Determination of best estimate values and probability density functions for each parameter of the model OURSON. When parameters were specifically calibrated for the Loire context, they were preferred to generic values. Otherwise, an exhaustive literature review was performed to propose generic PDFs.
2. Case study on the Loire river focused on uncertainty analysis. Because radioactive concentrations in routine releases are low and because subsequent concentrations in the environment are generally lower than detection limits, it is generally impossible to compare the model results with empirical data. In such a context, uncertainty analysis appears necessary because it allows study of all the potential parametric combinations. In such a way, it is expected to quantify confidence intervals for some endpoints (concentration in environmental compartments or mean annual dose for different exposure pathways) and to verify that the probability to exceed some limits is negligible.
3. Case study on the Loire river focused on sensitivity analysis. To define research priorities for the improvement of dose assessment in the context of routine releases in rivers, it is also necessary to identify sensitive parameters, i.e. parameters which are responsible for the global uncertainty.

2. Input data and methods

2.1. Study area and input data

The Loire river, chosen as a test case, is described in Fig. 1: four nuclear sites are present on the Loire river (Belleville, Dampierre, St Laurent and Chinon) and one on its tributary, the Vienne river (Civaux). The daily concentration of radionuclides (^{58}Co, ^{60}Co, ^{110m}Ag, ^{134}Cs, ^{137}Cs, ^{54}Mn and ^{131}I) in raw river water (dissolved phase + suspended matter) at different points of the river (Beaulieu, Gien, Ouzouer, Orléans, Beaugency, Nouan, Blois, Tours, La Chapelle, Bertignolles, Saumur, Angers, Montjean, i.e. over 350 km) was calculated by a specific model simulating hydrodynamic processes, called CALIDO, and described elsewhere (Luck, 2001; Siclet et al., 2002). For such calculations, actual radioactive releases from the nuclear power plants situated along the Loire river were considered, except for ^{90}Sr. For ^{90}Sr, which is not quantified in liquid radioactive releases, a hypothetic concentration of 1 Bq l^{-1} in the effluent was considered. Consequently, input data of the dose model OURSON were the activity in raw river water as a function of time and space. To show the spatial variability of radioactivity in raw water along the Loire river, mean annual concentrations in 1996 calculated at each station investigated are presented in Fig. 2. It may be observed that the activity of each radionuclide is significantly higher just downstream of a nuclear site, especially at Bertignolles, which is situated downstream of Chinon. On the contrary, for stations where releases are completely

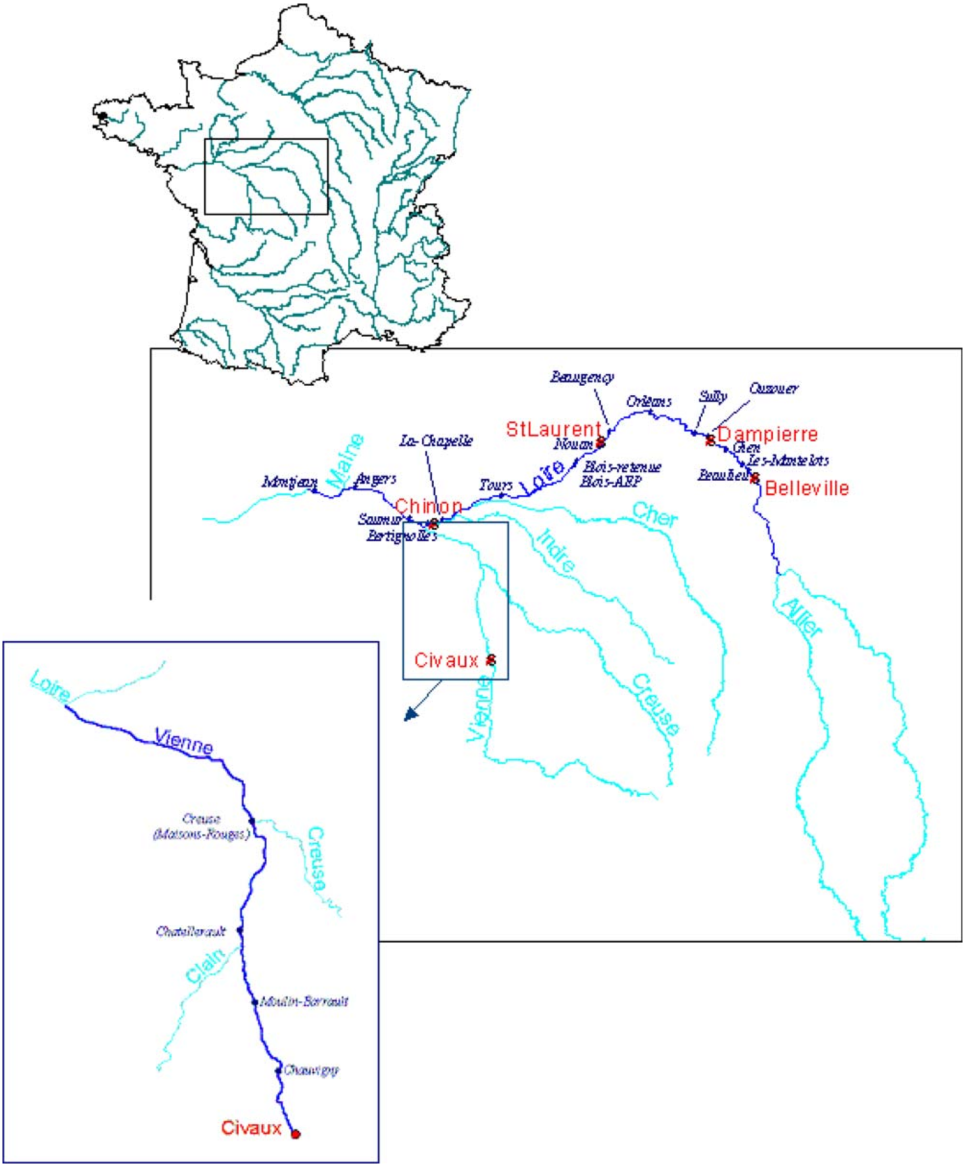

Fig. 1. Location map of the Loire river system and its tributaries (NPPs and major cities where radiological dose to man was calculated are indicated).

mixed with river water, no significant increase was observed along the Loire river. To show the temporal variability of radioactivity in raw water, monthly concentrations of each radionuclide at Montjean (last station investigated on the Loire river) are presented in Fig. 3. High temporal variations may be observed. In particular, releases are constrained by the flow rate of the river: when it is lower to a specific

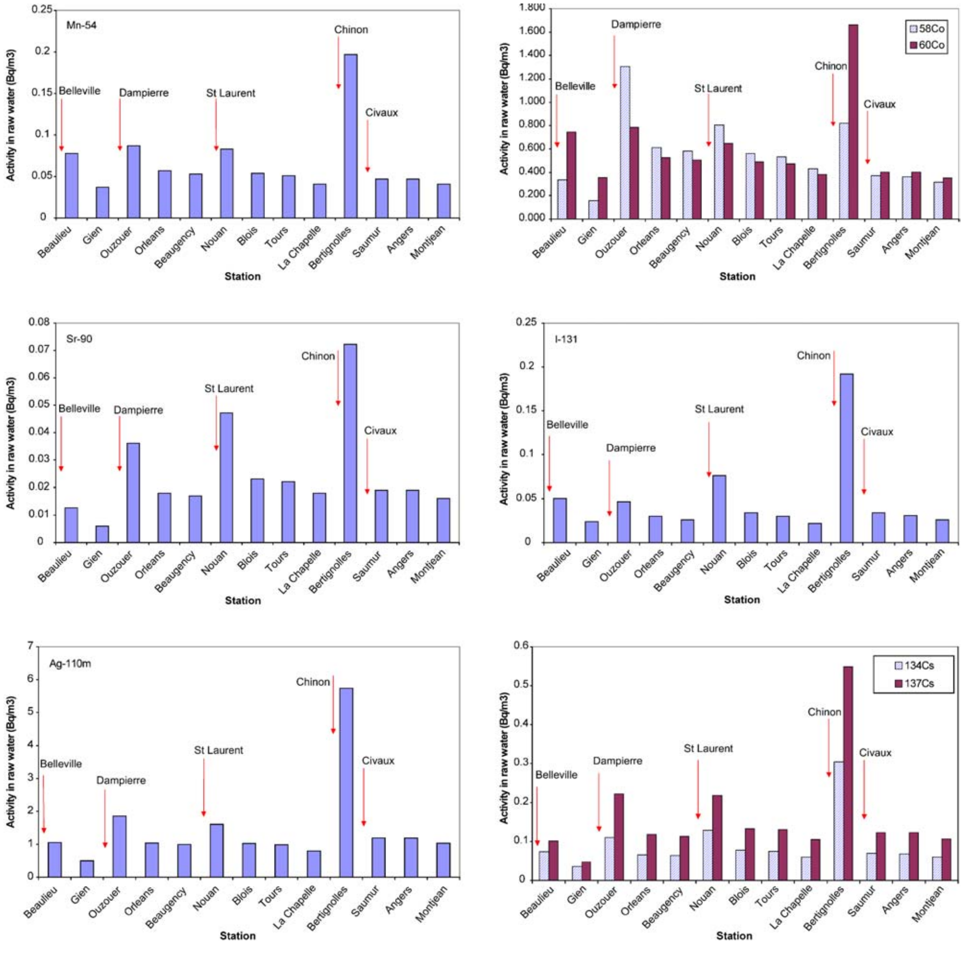

Fig. 2. Spatial variability of radioactivity along the Loire river – mean annual activity in raw river water in 1996.

value, releases are forbidden, because of the low dilution rate; when it is higher than another specific value, it is also forbidden to prevent flood consequences. Consequently, concentrations in river water may be low during some winter and/or summer periods. Furthermore, releases are also related to some particular operations on NPPs. Such temporal variations justify why it is necessary to represent transfer processes dynamically and to take into account seasonal variations in meteorological and agricultural conditions.

2.2. Uncertainty and sensitivity analysis

Uncertainty and sensitivity analysis were performed on some endpoints (concentration in environmental compartments or mean annual dose for different

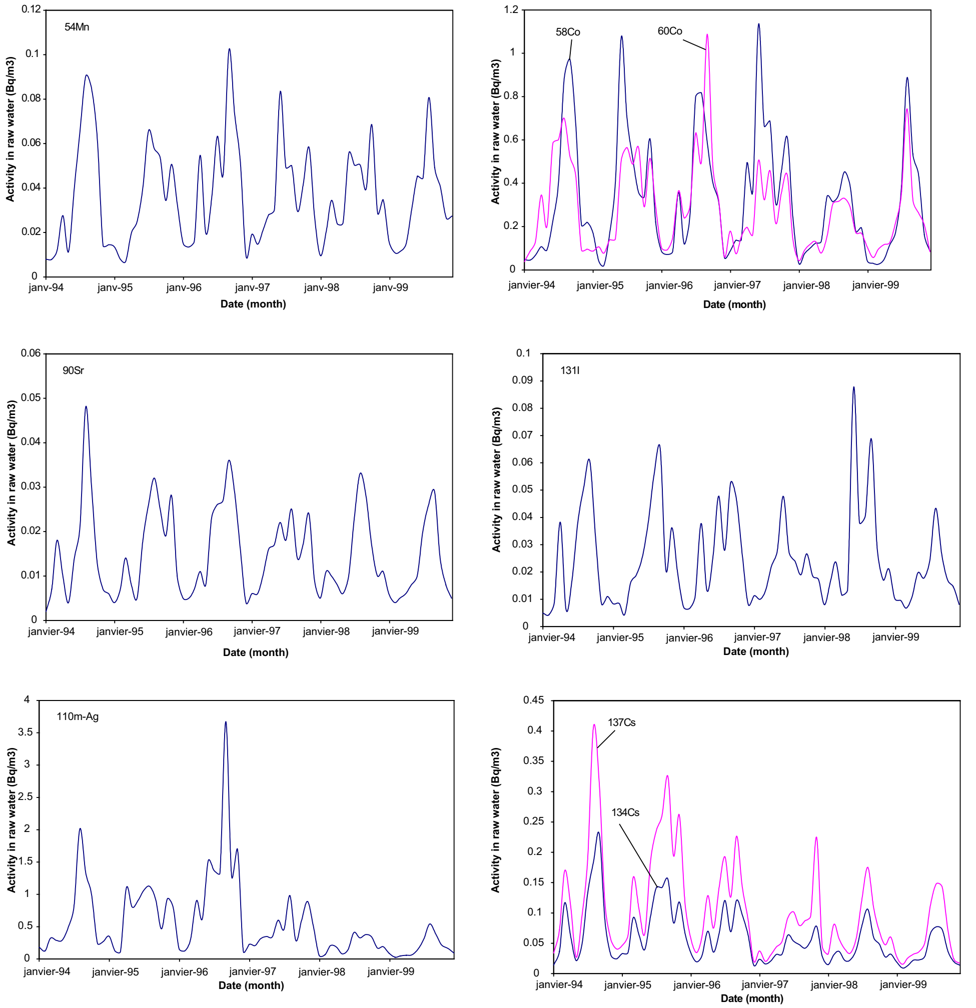

Fig. 3. Temporal variability of radioactivity in the Loire river – average monthly activity at Montjean in raw river water.

exposure pathways). For such analysis, each uncertain parameter is described by a probability density function and random samplings were performed on each of these PDFs by Monte Carlo (Latin Hypercube) procedures. When a correlation between two parameters was introduced, random samplings were rearranged according to the Iman-Conover's procedure (Iman and Conover, 1982) in order to obtain the desired correlation between the ranks of the correlated parameters.

The uncertainty analysis was based on the interpretation of the 5th and 95th percentiles and on the fit to typical distributions (log-normal, log-uniform or log-triangular).

The sensitivity analyses were based on the interpretation of regression calculations. In other words, a regression between the output of the calculation on one part and the input parameters on the other part is calculated as follows: $Y = a_0 + \sum_i a_i X_i + \varepsilon$. Several indicators were used to control the quality of the regression: (i) the determination coefficient r^2 of the regression; (ii) the skewness and kurtosis coefficients on the residues ε in order to verify that these residues are normally distributed; (iii) the distance between each residue and the axis of the regression (according to the Cook's distance) in order to identify possible outliers. When the quality of the regression is not good enough, some adaptations may be tested: (i) log transformations on the output and/or some input parameters (when they are distributed over several orders of magnitude); (ii) rank transformation on the input parameters (when the relations are not linear); (iii) introduction of interactions between input parameters in the regression analysis; in this case, the regression relationship is as follows: $Y = a_0 + \sum_i a_i X_i + \sum_{ij} a_{ij} X_i X_j + \varepsilon$. The hierarchy in the sensitivity of the investigated input parameters may be established according to the following indicators:

- "Multi-regression" analysis. Several regression calculations were performed; for each of them, one parameter was omitted. In other words, the first regression (for which the parameter X_1 was omitted) may be written as follows (when interactions are taken into account): $Y = a_0 + \sum_{i,i \neq 1} a_i X_i + \sum_{ij,i \neq 1,j \neq 1} a_{ij} X_i X_j + \varepsilon$, the same procedure being performed for all the parameters. A sensitivity index is defined as: $\mathrm{Sens}_{Xi} = r^2_{\mathrm{tot}} - r^2_{-X_i}$, where r^2_{tot} is the determination coefficient when all the parameters are taken into account and $r^2_{-X_i}$ is the determination coefficient when the parameter X_i is omitted. Consequently, the sensitivity index Sens_{Xi} measures the "loss" of correlation when the parameter X_i is ignored in the analysis.
- Student's t-values of the regression analysis, allowing to assess whether the coefficient of the regression associated to each term (parameter or interaction) is significantly different from zero. It may be considered that a high t-value (in absolute value) is associated to sensitive parameters or interactions.

In further applications, the maximum number of sensitive terms graphically described is limited to the 10 most sensitive terms.

3. Parameterisation

The objective of this section is to present how each parameter of the model was parameterised, i.e. how PDFs[1] were obtained for each of them. In this section,

[1] In the tables presented in this paper, uniform and log-uniform PDF are characterized by their minimum and maximum values; triangular and log-triangular PDF are characterized by their minimum value, their mode and their maximum values; normal and log-normal PDF are characterized by their 5th and 95th percentiles.

several sub-compartments of the environment were considered: (1) water, suspended matter and bed sediments; (2) fish; (3) soil layers; (4) atmosphere; (5) plant foliar surface; (6) consumable parts of plants; (7) animal products; (8) agricultural and human parameters. The equations of the OURSON model were not systematically detailed when they are not innovative versus existing models (Abbott and Rood, 1994; Boone et al., 1981; Koch and Tadmor, 1986; Müller and Pröhl, 1993; Simmons et al., 1995; Whicker and Kirchner, 1987; Zach and Sheppard, 1991; Zeevaert et al., 1995).

3.1. Water, suspended matter and bed sediments

3.1.1. Exchanges at the water–suspended particulate matter (SPM) interface

Two options are possible in the OURSON model to predict the distribution of radionuclides between water and suspended matter.

- The equilibrium option. The distribution of radionuclides between water and suspended matter is supposed to be equilibrated and may be described by a "Distribution coefficient", $K_{d,w}$, representing the ratio between the particulate activity (in Bq g^{-1}) and the dissolved activity (in Bq m^{-3}).
- The kinetic option. This approach describes kinetic exchanges between three sub-compartments: the dissolved phase, the particulate exchangeable sites and the particulate non-exchangeable sites. This approach is described in detail and parameterised for the Loire river in Ciffroy et al. (2001, 2003). For this approach, four kinetic parameters are needed.

In the applications presented in this paper (as a first approach), the "equilibrium" option was considered. When the sensitivity analysis showed that the $K_{d,w}$ parameter is sensitive, the more refined kinetic option can be used. Precisely, the objective of sensitive analysis is to create feed-backs to the models in order to improve the description of sensitive pathways, and on the contrary to maintain simple approaches when refinements are unnecessary (i.e. when the uncertain parameter was not identified as sensitive).

For the applications concerning the Loire river, specific in situ and laboratory experiments were performed in order to study the kinetic exchanges of radionuclides at the water–SPM interface and to measure $K_{d,w}$ values. The principle of in situ experiments is the following: the activity of several radionuclides was measured in the dissolved phase and in SPM (collected by centrifugation or cartridge filtration) downstream of nuclear power plants situated along the Loire river during scheduled discharges of low activity wastes; such experiments allowed the calculation of in situ $K_{d,w}$ values for Co, Cs, Mn and Ag (Ciffroy et al., 1995). Such in situ results were also compared to batch experiments: partitioning of several radionuclides between the dissolved phase and SPM were followed up over about 100 h after adding radioactive tracers to freshly collected Loire river water (Ciffroy et al., 2001, 2003). Results obtained by both in situ and batch experiments for Co, Cs, Mn and Ag, as well as results obtained by Thirion et al. (1983) who also performed batch experiments with Loire river water, allowed to propose PDFs of the $K_{d,w}$ parameter for the Loire river (Table 1). For ^{90}Sr, only results from batch experiments were available for the Loire river (Ciffroy, 2000); they were compared to other values available in the literature for determining a PDF (Table 2). The PDF for iodine was determined on the basis of literature information (see Table 2).

To calculate the activity in water and particles, it is also necessary to know the SPM concentration. As SPM concentration may highly depend on the hydrological conditions of the river, particularly during flood events, a relationship between the SPM concentration and the daily flow rate Q_n (in $m^3\ s^{-1}$) of the river is considered in the model: $SPM_n = aQ_n^b$. More sophisticated models, allowing to simulate hysteresis processes and succession of flood events, are also available (see Ciffroy et al., 2000). But, the comparison between calculated and empirical values over two years showed that the simple relationship tested in the present paper gave satisfactory results for the Loire river. Such results are showed in Fig. 4, where empirical data are compared to three kinetic calculations (with best estimates and extreme values of a and b). Consequently, to simulate the relationship between SPM and flow rate, two parameters, noted a and b, are needed. For calibrating these two parameters, a long-term monitoring of SPM in the Loire river was performed (daily measurements of SPM during the period July 1998–November 1999 and daily measurements of turbidity during the period February 1992–December 1995) (Luck, 2001). The uncertainty of the two parameters (considering only mineral SPM) was estimated by a bootstrap procedure and the PDFs presented in Table 1 were proposed.

3.1.2. Bottom sediments

Deposition and resuspension of particles (and associated particulate radionuclides) were simulated by the equations proposed by Krone and Partheniades, describing the flux of deposited or resuspended cohesive particles according to hydraulic conditions of the river (Krone, 1962; Partheniades, 1965; Ciffroy et al., 2000). According to this approach, deposition may occur only when the shear stress (related to the water velocity and the Manning's coefficient) is lower than a critical deposition shear stress τ_d and, on the contrary, resuspension may occur only when the shear stress is higher than a critical resuspension shear stress τ_r. When deposition or resuspension conditions are verified respectively, the particles flux to or from the sediments is linked to a settling velocity of particles W_c and an erosion rate e, respectively. Consequently, five parameters must be determined to calculate dynamics of radioactive contamination in bed sediments: the Manning's coefficient n_{Ma}, the critical deposition shear stress τ_d, the critical resuspension shear stress τ_r, the settling velocity of particles W_c, the erosion rate e. The PDFs of these parameters were determined as follows:

The deposition τ_d and W_c parameters were calibrated for the Loire river by estimating the mass of sediments stored upstream of the Blois dam at the end of a period of deposition (October 1999); this estimation was performed through in situ sampling of sediment cores and by using a hydraulic model (Luck, 2001; Siclet et al., 2002). The resuspension τ_r and e parameters were calibrated by measuring SPM concentrations during a resuspension event at the Blois dam (due to a seasonal operation on the dam). In order to estimate the uncertainty of these parameters, the values thus calibrated were compared with other published values determined by

Table 1
List of parameters in the model OURSON – 'Aquatic' parameters

Symbol	Unit	Definition	Value								References
			^{54}Mn	^{58}Co	^{60}Co	^{90}Sr	^{110m}Ag	^{131}I	^{134}Cs	^{137}Cs	
$K_{d,w}$	$m^3\ g^{-1}$	Distribution coefficient in the river	LN (0.02; 70)	LN (0.02; 20)		LN (10^{-5}; 10^{-2})	LN (0.05; 0.5)	LU (10^{-5}; 3×10^{-3})	LN (0.01; 0.2)		For Co, Mn, Ag and Cs: Ciffroy et al. (2001, 2003), Thirion et al. (1983) For Sr and I: see Table 2,
a	–	1st parameter of the relation SPM-flow rate	LN (2.05×10^{-3}; 4×10^{-3})								Luck (2001)
b	–	2nd parameter of the relation SPM-flow rate	N (1.35; 1.47)								Luck (2001)
W_c	$m\ d^{-1}$	Settling velocity of particles	LN (0.35; 35)								See Table 3
τ_d	Pa	Critical deposition shear stress	LT (0.05; 0.3; 0.4)								See Table 3
e	$g\ m^{-2}\ d^{-1}$	Erosion rate	LT (250; 10^5; 10^5) Correlation coefficient between e and τ_r: 0.9								See Table 3
τ_r	Pa	Critical resuspension shear stress	LT (0.1; 4; 4) Correlation coefficient between e and τ_r: 0.9								See Table 3
n_{Ma}	$s\ m^{-3}$	Manning's coefficient	N (0.02; 0.07)								Luck (2001), Siclet et al. (2002)
B_f	$m^3\ kg^{-1}$ FW	Water-to-fish transfer factor	LU (10^{-2}; 1)	LU (5×10^{-3}; 0.5)		LU (10^{-3}; 1)	LU (2×10^{-4}; 5×10^{-2})	LU (10^{-2}; 1)	LU (10^{-2}; 15)		See Table 4
$\lambda_{bio,f}$	d^{-1}	Biological elimination rate for fish	LU (5×10^{-4}; 10^{-2})	LU (5×10^{-3}; 5×10^{-2})		LU (5×10^{-5}; 10^{-3})	U (0; 2×10^{-3})	LU (5×10^{-4}; 10^{-2})	LU (5×10^{-4}; 10^{-2})		See Table 5

Table 2
$K_{d,w}$ for ^{90}Sr ($m^3\ g^{-1}$) – some values available in the literature

Radionuclide	Site	$K_{d,riv}$ ($m^3\ g^{-1}$)	Reference	Method of determination
^{90}Sr	Loire river	3.7×10^{-4} Range: 2.5×10^{-4}–4.6×10^{-4}	Thirion et al. (1983)	Batch experiment. Loire river water collected in winter and summer (9 values)
	Nete river	8×10^{-4}–1×10^{-3}	Zeevaert et al. (1985)	In situ sampling of SPM by centrifugation
	Ottawa river	5.5×10^{-4}	Joshi and McCrea (1992)	In situ sampling of SPM by centrifugation
	Snake river	1.1×10^{-5}–2.3×10^{-5}	Hemming et al. (1997)	Batch experiments (studied variables: sample agitation, SPM/water ratio, sample preparation)
	Chernobyl rivers	1.4×10^{-3}–8.5×10^{-3}	Matsunaga et al. (1998)	In situ monitoring (sampling by cartridge filtration)
	Loire river	10^{-4}	Ciffroy (2000)	Batch experiment
Radio-iodine	Pô river	2.6×10^{-3}	Battaglia et al. (1980)	In situ monitoring of scheduled discharges of radioactive wastes
	Winnipeg river	1.6×10^{-4}	Bird et al. (1995)	Batch experiments. The mean value obtained under oxic conditions was reported
	Winnipeg river + lake sediments	1.5×10^{-5}–10^{-4}	Bird and Schwartz (1996)	Batch experiments. Only results obtained under oxic conditions with peaty or silt sediments with a SPM/water ratio lower than 0.1 were considered in this study

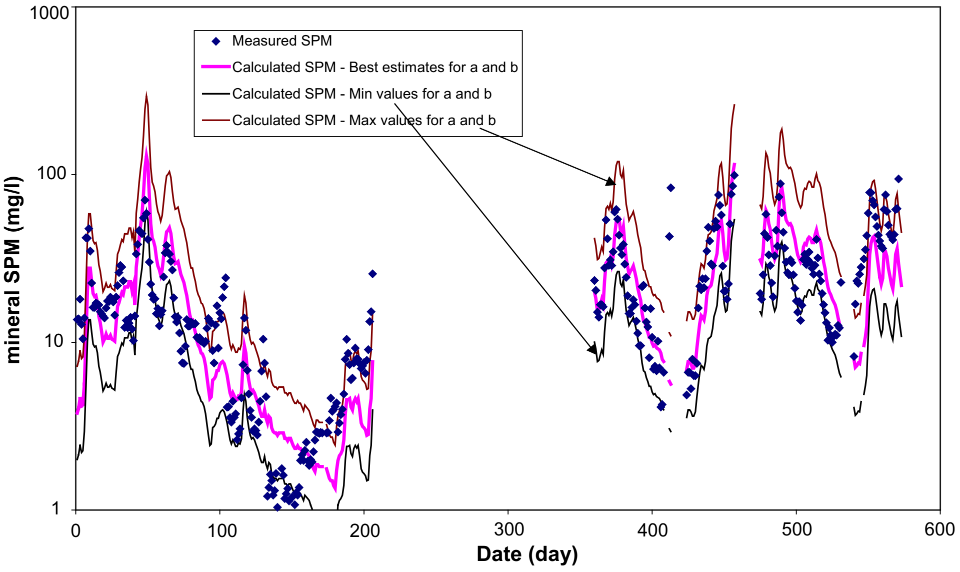

Fig. 4. SPM concentration in the Loire river over two years – model results and empirical data.

several methods: application of the Stokes' law (Liu et al., 2002), use of sediment traps (Kozerski, 2002), measurements of vertical gradients of SPM (Ciffroy et al., 2000), flume experiments (Blom and Aalderink, 1998; Krishnappan and Marsalek, 2002; Lau and Droppo, 2000), long-term monitoring of SPM concentrations in a river (Luck, 2001; Wu et al., 1998) (Table 3). The settling velocity of particles W_c may vary by one order of magnitude; consequently, a log-normal PDF was chosen to represent this parameter, the mode being the value specifically determined for the Loire river. The critical deposition shear stress τ_d and the critical resuspension shear stress τ_r vary by one and two orders of magnitude respectively. The values determined for the Loire river were among the highest values; consequently, the Loire river was chosen to represent these parameters by log-triangular PDFs. The erosion rate e shows a very high variability, the value determined by Luck (2001) for the Loire river being the highest one; a log-triangular PDF was therefore proposed. However, it has to be underlined that the calibration of the erosion rate is highly related to the value selected for the critical resuspension shear stress. Consequently, it was chosen to correlate the parameters e and τ_r as proposed by Blom and Aalderink (1998). It has also to be noted that the uncertainty chosen for each parameter in this study is higher than those proposed by Maurer et al. (1998) in their sensitivity analysis of deposition/resuspension models.

Specific tracing experiments were performed on the Loire river to calibrate the Manning's coefficient (Luck, 2001; Siclet et al., 2002). The mean value and the dispersion of the values determined for the Loire river were used to propose a PDF.

It must also be underlined that the direct exchanges at the interface water-sediments (i.e. exchanges between sediments particles and porewater, followed by diffusion) were not considered in this study. This assumption was based on

Table 3
Deposition/resuspension parameters – some values of the literature

Site	W_c (m j^{-1})	τ_d (Pa)	e (gm^{-2} d^{-1})	τ_r (Pa)	Reference	Method of determination
Ribble estuary		0.06–0.09		0.2–0.3	Burton et al. (1995)	SPM measurements in the Turbidity maximum and mathematical calibration
Lake Ketel	19 [(2)] range: 13–26		4800 [(1)]	0.14 [(1)]	Blom and Aalderink (1998)	[(1)] Flume experiment; [(2)] in situ monitoring of SPM concentrations over 1 week
Humber		0.07		0.15	Wu et al. (1998)	Long-term monitoring of SPM
Königshafen (D)				0.3–3.3	Tolhurst et al. (1999)	In situ measurement by a Cohesive strength meter
Seine river	26	0.13			Ciffroy et al. (2000)	Measurements of SPM vertical gradients
Flume experiment				Range: 0.03–0.24	Lau and Droppo (2000)	Flume experiment
Vienne river	1	0.4	430	1	Luck (2001)	Long-term monitoring of SPM
Loire river	3.5	0.3	104,000	4	Luck (2001)	Evaluation of the mass of sediments stored upstream of the Blois dam (cores); monitoring of a resuspension event
Flume experiment			1300–2600	0.02–0.04	Schaff et al. (2002)	Flume experiment
Spree river	4.3				Kozerski (2002)	Use of sediment traps
Tanshui river	22.5		260	0.1	Liu et al. (2002)	Application of the Stokes' law
Flume experiment		0.05		0.12	Krishnappan and Marsalek (2002)	Flume experiment

laboratory experiments aiming at quantifying the sorption/desorption kinetics at the water–particles interface (Ciffroy et al., 2001, 2003). According to these experiments, a great part of the particulate radioactivity is generally associated to non-exchangeable sites and only a low part is expected to desorb.

3.2. Fishes

The contamination level of fish is supposed to be controlled by a transfer rate between water and fish B_f and by a biological elimination constant $\lambda_{bio,f}$. In this global preliminary study, it was chosen to use such a simple approach, that could be refined when B_f and/or $\lambda_{bio,f}$ parameters are identified as sensitive parameters (see further sensitivity analysis in this paper). Indeed, the uncertainty on the B_f parameter could be reduced if cofactors are taken into account (as an example, for Cs: K concentration, fish size and distinction between predatory/non-predatory fishes (Smith et al., 2000, 2002)). However, before introducing such refinements in the OURSON model, it was necessary to identify whether the investigated parameters are sensitive or not in the final result and whether refinements can actually improve the dose assessment. Consequently, PDFs proposed for the two parameters representing transfer to fish include natural variability related to several environmental factors and include also indirect transfer from feeding.

Some values available in the literature for the water-to-fish transfer factor B_f are presented in Table 4. Log-uniform distributions were chosen because of the lack of information about the distribution of values over the range of experimentally observed transfer factors (Table 1). It may be observed that the modes of these PDFs are in the same order of magnitude as the values recommended by Poston and Klopfer (1986) on the basis of an exhaustive literature review.

Biological decorporation of radionuclides from fish were generally determined by in vitro experiments. It was generally observed that at least two biological half-lives may be distinguished, each of them representing low-retention and high-retention compartments respectively. In the study presented in this paper, only the high-retention compartment was considered, such a hypothesis being an overestimating hypothesis for dose calculations. Biological elimination rates determined by several authors were reviewed (see Table 5) and extreme values were used to define log-uniform PDF for Cs, Ag, Co and Mn (see Table 1). For iodine and strontium, the allometric approach proposed by Highley et al. (2003) was used to estimate analogies between these elements and Cs; thus, it was considered that biological elimination rates for iodine are in the same order of magnitude as for Cs, while they are one order of magnitude lower for Sr.

3.3. Soil layers

In the OURSON model, the soil compartment is divided in three sub-compartments: (i) the soil surface layer is defined as the soil sub-compartment which interacts with atmosphere via dust deposition and resuspension processes; (ii) the 'ploughing' zone of the soil is defined as the soil sub-compartment which is

Table 4
Water-to-fish transfer factor – some values of the literature

^{54}Mn	^{58}Co and ^{60}Co	^{90}Sr	^{110m}Ag	^{131}I	^{134}Cs and ^{137}Cs	Reference	Method of determination
	Range: 5×10^{-3}–0.65	7×10^{-4}–0.2 PDF: LN (1.7×10^{-4}; 4.4×10^{-3})		8×10^{-3}–1.1 PDF: LN (8×10^{-3}; 6×10^{-2})	Range: 0.3–14 PDF: LN (0.16; 2.5)	Blaylock (1982)	Literature review. Selection of data for proposing a PDF
Range: 4×10^{-2}–5 Recommended value: 0.4	Range: 1.8×10^{-2}–0.13 Recommended value: 3×10^{-2} for eutrophic water 0.3 for other water	8×10^{-4}–1.2 Recommended value: 5×10^{-2}		Range: 2×10^{-3}–0.8 Recommended value: 0.2	Range: 0.8–5 Recommended value: 3 for piscivorous Recommended value: 1 for non-piscivorous if $[K]=5$ mg l^{-1} (typical concentration for the Loire river)	Poston and Klopfer (1986)	Literature review (data for whole fish; data for muscles are also available)
Range: 6.3×10^{-2} –0.13	Range: 2×10^{-2}–8×10^{-2}				Range: 0.13–1.7	Lambrechts and Foulquier (1987)	In situ monitoring of the Rhône river between 1980 and 1984
Range: 5×10^{-2}–0.5	Range: 10^{-2}–0.3	Range: 10^{-3}–1	Range: 2×10^{-4}–10^{-2}	Range: 2×10^{-2}–0.6	Range: 3×10^{-2}–3	IAEA (1994)	Literature review
1.3×10^{-2} 0.1	9×10^{-3} 4×10^{-3}		5.1×10^{-2} 4×10^{-3}		2.7×10^{-2} 7×10^{-3}	Adam (1997), Adam et al. (1997)	Laboratory experiment (rainbow trout and carp)
8×10^{-3}						Garnier-Laplace et al. (2000)	Laboratory experiment (rainbow trout)
					Mean: 1.9 Range: 0.82–14	Smith et al. (2000)	In situ monitoring (28 data; 1992–1997) in non-predatory and predatory fishes in lakes after the Chernobyl accident

Table 5
Biological elimination rates for fish – some values of the literature

^{54}Mn	^{58}Co and ^{60}Co	^{90}Sr	^{110m}Ag	^{131}I	^{134}Cs and ^{137}Cs	Reference	Method of determination
6.2×10^{-4}	4.1×10^{-3}				7.4×10^{-4}	Baudin et al. (2000)	Juvenile trout fed with contaminated carps during the contamination phase
					Trout: 2.3×10^{-3}–4.8×10^{-3}	Smith et al. (2002)	More than 1000 measurements of ^{137}Cs in predatory fish in nine lakes
					Perch: 1.4×10^{-3}–4.4×10^{-3} Trout: 6.6×10^{-4}–1.9×10^{-3}		
					3×10^{-4}–4.3×10^{-4}	Peles et al. (2000)	^{137}Cs measurements in fish collected from Steel Creek in 1974, 1981 and 1998
		$\ln2/(107.(1000\times M)^{0.26})$		$\ln2/(6.8.(1000\times M)^{0.13})$	$\ln2/(3.5(1000\times M)^{0.21})$	Highley et al. (2003)	Allometric approach
Carp: 1.1×10^{-2}	Carp: 2×10^{-2}		Carp: 0		Carp: 7.5×10^{-3}	Adam (1997)	Fishes contaminated through addition of radionuclides in water
Trout: 8.1×10^{-3}	Trout: 3.6×10^{-2}		Trout: 0		Trout: 1.7×10^{-2}		
			Carp: 2.3×10^{-2}			Baudin et al. (1994)	Cited by Adam (1997)
	Carp: 2.5×10^{-2}					Fritsch and Baudin (1984)	Cited by Adam (1997)
					Carp: 6.5×10^{-3}	Garnier-Laplace et al. (1996)	Cited by Adam (1997)
			Trout: 5×10^{-3}			Garnier et al. (1992)	Cited by Adam (1997)
	2×10^{-2}					Reed (1971)	Cited by Adam (1997)
		1.2×10^{-3}				Kryshev (2003)	Fish contamination monitoring after the Kyshtym accident

mixed with the soil surface during ploughing operations. For some plants (vegetables, pasture), it may also correspond to the zone where root uptake processes occur; (iii) The 'cultivable' zone of the soil is defined as the deeper soil sub-compartment where root uptake processes may potentially occur for some plants (maize, cereals). Inputs and outputs of radionuclides to and from these soil layers are:

- *Irrigation of contaminated river water*. Practically, irrigation rates are dependent on meteorological conditions, at least for crops such as maize, cereals and pasture. This is the reason why in the present model, irrigation rates are calculated at each time step (during the period between germination and harvest of each crop) by taking into account precipitation and evapotranspiration, and the previous water budget in soil. To calculate needs in water of the soil, and associated irrigation rates, it is necessary to evaluate at each time step the water content in the soil. The water content in the soil is conditioned by two parameters: (i) the water content at wilting point and at field capacity, which represent the minimum and maximum water content respectively. When irrigation is needed (i.e. when the stock of water in the soil is insufficient), and according to current habits, a given quantity of water $Qirr_{vi,pass}$ is used for irrigation. In conclusion, three parameters are needed for calculating irrigation rates at each time step: the water content at wilting point; the water content at field capacity; the irrigation rate for each 'passage'.
- *Weathering from foliar surface of plants*. A fraction of the irrigated water is intercepted by leaves and, as a consequence, reaches soil after a retardation time, when weathering of intercepted radionuclides from leaves occurs. The parameterisation of this process is described in the section "Plants foliar surface".
- *Percolation of dissolved radionuclides into deeper zones of the soil (from the soil surface layer to the 'ploughing' zone, from the 'ploughing' zone to the 'cultivable' zone and from the 'cultivable' zone to the non-saturated zone)*. The velocity of percolation water is calculated taking into account several principles: (1) water content always ranges between wilting point and field capacity; (2) velocity of percolation water in soil depends on water budget in the soil, which is controlled by water inputs from rainfall and irrigation and outputs due to evapotranspiration; (3) loss of water may occur only if water budget in soil leads to a water content higher than water content at field capacity. Consequently, to calculate the velocity of percolation water in the soil, it is therefore necessary to parameterise the water content at wilting point and at field capacity. Furthermore, percolation of radionuclides depends also on their distribution between particles and porewater. Two parameters were used to describe this distribution: the distribution coefficient in the soil $K_{d,soil}$ and the bulk density of dry soil ρ_{soil}.
- *Erosion of particles from the soil surface*. This process depends on several factors such as soil composition, rainfall and plant canopy, ... In the OURSON model, it is parameterised by a global erosion loss from the soil.
- *Loss of radionuclides by runoff from the watershed.* The runoff velocity $v_{r,n}$ is determined according to the SCS runoff curve number method (Anonymous,

1986). According to this method, the runoff is related to rainfall and to a Curve number parameter depending on the type of soil and plant canopy.

The PDFs of each of the parameters previously mentioned were determined as follows:

- *Water content at wilting point and field capacity* may vary with regard to physico-chemical characteristics of soil (for example, fine particles content in the soil). Baes and Sharp (1983) reviewed more than 150 values of water content at wilting point and field capacity for soils in the US. Several types of soils were distinguished: silt loams, clay and clay loams, sandy loams, loams. To be as generic as possible, the PDFs for wilting point and field capacity were established considering the category 'all types of soils' (see Table 6). However, water content at field capacity cannot be determined independently of water content at wilting point as both parameters are correlated. Consequently, a correlation coefficient of 0.9 was applied.
- For a given radionuclide, the *distribution coefficient in the soil* $K_{\mathrm{d,soil}}$ may depend on many factors: (i) physico-chemical characteristics of the soil (pH, organic matter content, granulometric distribution, ...); (ii) speciation of radionuclides when introduced in the soil; (iii) experimental procedures undertaken for its determination; (iv) equilibrium or non-equilibrium conditions. Literature reviews allow one to estimate how far the distribution coefficient may vary versus various conditions (Baes and Sharp, 1983; Sheppard and Thibault, 1990). Sheppard and Thibault (1990) also distinguished different types of soils (sand, loam, clay, organic). The PDFs used in this paper were based on those mentioned in these reviews for loam and clay soils (sandy and organic soils were excluded from the analysis because they are not adapted to agricultural practices), with each PDF proposed by Baes and Sharp (1983) and Sheppard and Thibault (1990) being weighted by the number of data used for their determination (see Table 6). For iodine and cesium, respectively, the proposed PDFs are coherent with the range of data published more recently by Sheppard (2003) and Sanchez et al. (2002).
- *Bulk density of soil* depends on soil composition and porosity. Soltner (1994) proposed values of bulk density for several porosity ranges and several types of soil. For silty soils, bulk density may vary from 1000 to 1800 kg m^{-3}. This range is in accordance with best estimate values used in several dose models (Abbott and Rood, 1994; Müller and Pröhl, 1993; Simmons et al., 1995; Whicker and Kirchner, 1987; Zeevaert et al., 1995). Consequently, bulk density of soil was also described by a uniform PDF (see Table 4).
- Erosion of particles from the soil surface depends on several factors such as soil composition, rainfall and plant canopy, ... Soil loss from a watershed may be estimated by the solids load in surface water. On such a basis, Soltner reviewed erosion loss of several European rivers (Soltner, 1995): erosion loss ranged on several orders of magnitude, solids being eroded for plain rivers varying from 0.01 to 8.5 t ha^{-1} $year^{-1}$. Consequently, the soil erosion loss constant is described by a log-uniform distribution (see Table 6).

Table 6
List of parameters in the OURSON model – soil parameters

Symbol	Unit	Definition	Value								References
			^{54}Mn	^{58}Co	^{60}Co	^{90}Sr	^{110m}Ag	^{131}I	^{134}Cs	^{137}Cs	
θ_{wi}	–	Water content at wilting point	U (0.06; 0.27) Correlation coefficient between θ_{fc} and θ_{wi}: 0.9								Baes and Sharp (1983)
θ_{fc}	–	Water content at field capacity	U (0.22; 0.47) Correlation coefficient between θ_{fc} and θ_{wi}: 0.9								Baes and Sharp (1983)
$K_{d,soil}$	$m^3\ kg^{-1}$	Distribution coefficient in the soil	LN (3.2×10^{-3}; 23)	LN (3.7×10^{-3}; 7.2)		LN (1.1×10^{-3}; 8×10^{-1})	LN (1.4×10^{-2}; 1)	LN (1.4×10^{-4}; 10^{-1})	LN (7.2×10^{-2}; 37)		Baes and Sharp (1983), Sanchez et al. (2002), Sheppard (2003), Sheppard and Thibault (1990)
ρ_{soil}	$kg\ m^{-3}$	Bulk density of dry soil	U (1000; 1800)								Soltner (1994)
Q_{er}	$kg\ m^{-2}\ d^{-1}$	Average mass of eroded particles from soil	LU (3×10^{-6}; 2×10^{-3})								Soltner (1995)
CN	–	Curve number	U (60; 90)								Anonymous (1986)
K_{res}	m^{-1}	Resuspension factor	LN (1.7×10^{-9}; 4×10^{-7})								Garland et al. (1992)
h_{soil}	m	Height of the cultivable zone of the soil	U (0.6; 1.4)								Personal communication, 2001
h_{ss}	m	Height of the soil surface zone	U (10^{-3}; 10^{-2})								Abbott and Rood (1994), Müller and Pröhl (1993), Simmons et al. (1995), Whicker and Kirchner (1987)
h_{pl}	m	Height of ploughing zone of the soil	U (0.15; 0.3)								Soltner (1994)

- As mentioned in the NRCS document (Anonymous, 1986), the *curve number CN* may depend on several factors such as soil texture, cover type, soil treatment and antecedent runoff conditions. In this article, we considered the CN proposed in Anonymous (1986) for agricultural lands including bare soils, pasture and crops ('small grain' and 'legumes or rotation meadow') and for intermediate soil textures ('silt loam' and 'sandy clay loam'). Extreme values proposed in Anonymous (1986) for such conditions were used to determine a uniform PDF.

3.4. Dust in atmosphere

Resuspension and deposition of particles at the soil surface–atmosphere interface are supposed to be equilibrated. Consequently, activity of dust in near-ground air can be described by a resuspension factor K_{res}, representing the ratio between the activity in near-ground air (in Bq m^{-3}) and the concentration deposited in the superficial layer of the soil (in Bq m^{-2}).

The resuspension factor at the soil–air interface is highly variable because of the influence of many factors such as wind, soil characteristics, plant canopy and rainfall (Anspaugh et al., 1975; Garland et al., 1992; Nicholson, 1988; Sehmel, 1980; Slinn, 1978). The experimental estimation of this parameter is difficult as local resuspension and global transport of dust are mixed. Values reviewed by Nicholson (1988) and Sehmel (1980) concern essentially arid or desert regions and are not directly applicable to temperate climates. Consequently, it was preferred to statistically analyse data collected after the Chernobyl accident and reported by Garland et al. (1992). Resuspension factors determined experimentally ranged over three orders of magnitude and most of the values were in the range 10^{-7}–10^{-8} m^{-1}. Such an analysis led to a log-normal PDF (see Table 6).

3.5. Plants foliar surface

Calculation of the contamination of plants must distinguish between plants which are totally ingested (leafy vegetables, grass, maize silage) and those which are only partly consumed (cereals, root and fruit vegetables). However, for all plants, input of radionuclides on foliar surface is due to interception of river water used for irrigation, while output is due to weathering of the soil surface. For maize (grains), cereals, root and fruit vegetables, translocation of dissolved radionuclides to inner parts may also be considered as an output from the foliar surface. The interception by plants of a fraction of irrigated river water (dissolved phase and SPM, respectively) is simulated according to the Chamberlain's relationship (Chamberlain, 1970) and requires three parameters: the foliar uptake coefficients for water and particles, and the aerial biomass of plants at harvest. Weathering from leaves to soil is described by a loss constant from foliar surface λ_w. The PDFs of these parameters were determined as follows:

- *Foliar uptake coefficients*. Several authors have experimentally obtained values of foliar uptake coefficients (Chamberlain, 1970; Pinder and McLeod, 1988; Pinder et al., 1988; Pröhl and Hoffman, 1996; Vandecasteele et al., 2002). However, it is difficult to extrapolate their experimental conditions (gas or aerosols interception, short-term aspersion, coarse particles aspersion) to irrigation conditions. On the contrary, experiments undertaken by Hoffman et al. (1992, 1995) can be considered as similar to irrigation conditions: long-term aspersion, water containing dissolved radionuclides and fine particles, simulation of moderate- and high-intensity rainfall. Several parameters were studied leading to more than 80 experimental observations. For applications presented in this paper, only experiments undertaken under moderate-intensity conditions were statistically analysed because they are more similar to irrigation conditions. Experiments conducted by Hoffman et al. (1992, 1995) showed that several groups of radionuclides should be considered, the interception of cations being much more important than for anions (especially iodine). Such differences in retention values between radionuclides types were found to be more important than between plant types. Consequently, log-normal PDFs were determined for dissolved cations, dissolved anions and particles respectively with respect to work completed by Hoffman et al. (1992, 1995) (see Table 7).
- *Aerial biomass of plants at harvest*. Soltner (1990) reviewed extreme values of biomass at harvest for a great number of plants and such values were analysed for proposing PDFs in Anonymous (2002); these PDFs were considered in this article.
- *Loss constant from foliar surface λ_w*. Loss of activity from the foliar surface has several origins, such as wind removal, water removal and plant growth. All these losses are represented by a global loss constant for foliar surface λ_w. Miller and Hoffman (1983) reviewed 78 values determined under various experimental conditions. Because of its particular behaviour, iodine was analysed separately. For all the other elements, Miller and Hoffman (1983) proposed a PDF which was used for the study presented in this paper (see Table 7). It may be noted that values obtained from Chernobyl fallout are in good agreement with the pre-Chernobyl values (Kirchner, 1994; Pröhl and Hoffman, 1996).

3.6. Consumable parts of plants

Several pathways are responsible for the contamination of consumable parts of plants: (1) for grass and leafy vegetables: root uptake of radionuclides and interception on foliar surface; (2) for maize, cereals, root vegetables and fruit vegetables: root uptake of radionuclides, interception on foliar surface followed by translocation of dissolved radionuclides. In most dose models, it is considered that translocation is constant during the growth of the plant. However, Aarkrog (1969, 1971, 1975, 1983), Aarkog and Lippert (1971) and Pröhl et al. (1990) have shown that translocation depends on the physiological status of the plant. Consequently, in the OURSON model, time dependence of translocation is modelled according to the relationship proposed by Aarkrog and Pröhl (Aarkrog, 1969, 1975, 1983; Aarkrog

Table 7
List of parameters in the model OURSON – plant parameters

Symbol	Unit	Definition	Value								References
			^{54}Mn	^{58}Co	^{60}Co	^{90}Sr	^{110m}Ag	^{131}I	^{134}Cs	^{137}Cs	
$\mu_{v,p}$	m^2 soil kg^{-1} DW	Foliar uptake coefficient of particles	LN (0.9; 4.2)								Hoffman et al. (1992, 1995)
$\mu_{v,d}$	m^2 soil kg^{-1} DM	Foliar uptake coefficient of dissolved radionuclides	LN (0.95; 4.5)	LN (0.95; 4.5)		LN (0.95; 4.5)	LN (0.95; 4.5)	LN (0.13; 2.8)	LN (0.95; 4.5)		Hoffman et al. (1992, 1995)
$B_{vi,har}$	kg DW m^{-2} soil	Aerial biomass of the plant vi at harvest	Cereals: U (1; 1.4) – Maize: U (1.2; 1.6) – Grass: U (0.8; 1) – Root vegetables: U (0.8; 1) – Leaf vegetables: U (0.8; 1) – Fruit vegetables: U (0.75; 0.8)								Personal communication
λ_w	d^{-1}	Weathering constant for foliar surface	LN (1.9×10^{-2}; 8.9×10^{-2})	LN (1.9×10^{-2}; 8.9×10^{-2})		LN (1.9×10^{-2}; 8.9×10^{-2})	LN (1.9×10^{-2}; 8.9×10^{-2})	LN (0.03; 0.3)	LN (1.9×10^{-2}; 8.9×10^{-2})		Miller and Hoffman (1983)
$t_{tr,vi}^{max}$	d	Number of days before harvest when maximum translocation is reached	U (25; 50)	U (25; 50)		U (0; 10)	U (0; 50)	U (0; 50)	U (25; 50)		Aarkrog (1969, 1975, 1983), Aarkog and Lippert (1971), Pröhl et al. (1990)
b_{vi}	d^{-2}	Slope of the kinetic evolution of the translocation factor for the plant vi	U (7×10^{-4}; 2×10^{-3})								Aarkrog (1983), Pröhl et al. (1990)
$f_{tr,vi}^{max}$	–	Maximum translocation factor for the plant vi	N (0.02; 0.2)	N (0.045; 0.45)		N (1.5×10^{-2}; 0.15)	U (1.5×10^{-2}; 0.45)	U (1.5×10^{-2}; 0.45)	N (0.03; 0.3)		See Tables 8 and 9
$B_{v,vi}$ (cereals and maize)	kg$_{soil}$ kg^{-1} DW	Soil-to-plant transfer factor for plant vi	LN (3×10^{-2}; 3)	LN (1.7×10^{-4}; 8.2×10^{-2})		LN (1.1×10^{-2}; 1.2)	LN (4.7×10^{-5}; 8×10^{-3})	LU (5×10^{-4}; 3×10^{-1})	LN (7.5×10^{-4}; 0.18)		For Cs and Sr: Anonymous (1997); For Co, Mn, Ag: IUR (1989, 1992); For I: see Table 10

$B_{v,vi}$ (grass)	$kg_{soil}\ kg^{-1}$ DW	Soil-to-plant transfer factor for plant vi	LN (9.7×10^{-2}; 4.7)	LN (2.5×10^{-3}; 1.2)	LN (0.26; 6.5)	LN (4.7×10^{-5}; 8×10^{-3})	LN (3.4×10^{-4}; 3.4×10^{-2})	LN (4.4×10^{-3}; 2.8)	IUR (1989, 1992)
$B_{v,vi}$ (vegetables)	$kg_{soil}\ kg^{-1}$ DW	Soil-to-plant transfer factor for plant vi	LN (2.6×10^{-2}; 10)	LN (0.02; 2)	LN (0.23; 23)	LN (4.7×10^{-5}; 8×10^{-3})	LU (5×10^{-4}; 3×10^{-1})	LN (2.8×10^{-2}; 1.2)	IUR (1989, 1992); For I: see Table 10
$Y_{vi,har}$	kg DM m^{-2} soil	Biomass of the consumable part of the plant vi at harvest	Cereals: T (0.3; 0.75; 1.2) − Maize: T (0.4; 0.7; 1) − Grass: T (0.1; 0.4; 1) − Root vegetables: T (0.2; 0.6; 1.2) − Leaf vegetables: T (0.06; 0.3; 0.6) − Fruit vegetables: T (0.01; 0.06; 0.1)						Anonymous (2002), Soltner (1990)
τ_{vi}	–	Water content of the consumable part of the plant vi	Cereals: U (0.05; 0.2) − Maize: U (0.4; 0.6) − Grass: U (0.85; 0.95) − Root vegetables: U (0.7; 0.85) − Leaf vegetables: U (0.85; 0.95) − Fruit vegetables: U (0.7; 0.85)						IAEA (1994)

and Lippert, 1971; Pröhl et al., 1990), which requires three parameters: the maximum translocation factor $f_{\mathrm{tr,vi}}^{\max}$, the number of days before harvest when maximum translocation is reached $t_{\mathrm{tr,vi}}^{\max}$, and a kinetic parameter noted b. As far as the transfer from soil is concerned, it may be considered that root uptake of radionuclides is proportional to the mean activity in the root zone during the period separating germination and harvest of the plant. Consequently, this process requires the determination of a soil-to-plant transfer factor. The PDFs of these parameters were determined as follows:

- *Parameters describing foliar translocation*. Aarkrog (1983) studied the translocation of several radionuclides (Cs, Sr, Mn, Co, Zn, Sb, Fe) in barley and observed that time before harvest when maximum translocation is reached $(t_{\mathrm{tr}}^{\max})$ is relatively homogenous among radionuclides (from 25 days for Fe to 39 days for Zn), except for Sr $(t_{\mathrm{tr}}^{\max} = 2\ \text{days})$. These values were confirmed by Pröhl et al. (1990) for Cs in barley and wheat $(t_{\mathrm{tr}}^{\max}\ \text{from 35 to 40 days})$, but higher values were observed for potatoes (80 days). Experiments performed by Aarkrog (1969, 1975) with other radionuclides (Co, Sb, Ru, Sr, Cs, Mn, Fe) and by Macacini et al. (2002) with Cs showed that $t_{\mathrm{tr}}^{\max}$ is generally in the period (30–50 days) before harvest, except for Sr, Ru and in some cases Ce (less than 10 days). Such experimental data were used to propose uniform PDFs for the parameter $t_{\mathrm{tr}}^{\max}$. For iodine and silver, no experimental data was found. Consequently, a larger range of probable data was considered.
- Data published by Aarkrog (1983) and Pröhl et al. (1990) showed a relative homogeneity of the parameter b (slope of the kinetic evolution of the translocation factor) among radionuclides and plants (from 7×10^{-4} to 1.9×10^{-3}). Such data were used to propose a uniform PDF.
- A majority of data on maximum translocation factors $f_{\mathrm{tr}}^{\max}$ concerns Cs, and to a lesser extent Sr. Consequently, the methodology chosen for deriving a PDF applicable for each of the investigated radionuclides was the following: (1) derive analogy factors in order to apply Cs data to other radionuclides. Such analogies were determined on the basis of experiments described by Aarkrog (1969, 1975, 1983); ratios on translocation factors obtained for Cs and other radionuclides respectively are reported in Table 8. It was observed that Sr and Mn are translocated to a lesser extend than is Cs, and that an opposite trend was observed for Co; (2) collecting translocation factors for Cs for various plant types (cereals, vegetables, fruits). Data thus collected are reported in Table 9. It was observed that most of available data are in the range 0.1–0.2. On the basis of such values, a normal PDF was proposed. For iodine and silver, no data were found and PDF covering all the range of values observed for other radionuclides were proposed.
- *Soil-to-plant transfer factors*. Soil-to plant transfer factors depend on numerous factors such as speciation of radionuclides within the soil, soil characteristics, … Ng (1982) and Ng et al. (1982a) reviewed soil-to-plant transfer factors values, selecting the most suitable experimental conditions (outdoors lysimeters, absence of foliar contamination). The IUR (1989, 1992) completed this review,

Table 8
Ratio of maximal translocation factors obtained for Mn, Co, Sr on one part and Cs on the other part

Ratio of translocation factors measured for radionuclide i and Cs respectively			
^{54}Mn	^{58}Co and ^{60}Co	^{90}Sr	Reference
0.6	1.5	0.45	Aarkrog, 1983 (translocation in barley)
0.6	0.7	0.6	Aarkrog, 1983 (translocation in barley)
0.72	1.9	0.4	Translocation in wheat
0.5–0.6		0.25–0.3	Aarkrog, 1969 (translocation in rye)
0.6		0.6	Translocation in oats
0.6–0.8		0.6–0.8	Translocation in barley
0.3–0.8		0.35–0.8	Translocation in wheat

distinguishing several types of plants. The PDFs proposed by the IUR were used in the study presented in this paper. However, for Cs and Sr in cereals (and in maize), we used the 'aggregate expert distribution' determined in Anonymous (1997), which was based on judgement of seven experts in soil and plant processes. For iodine, data proposed in IUR (1989, 1992) was only for grass; for the other types of plants, a PDF was determined on the basis of a literature review (see Table 10).

3.7. Animal products

In the OURSON model, several animal products may be considered: cow milk, beef meat, lamb meat, poultry and eggs[2]. For each of these sub-compartments, inputs of radionuclides are: (1) ingestion of raw river water; (2) ingestion of contaminated feedstuffs (grass, cereals and/or silage maize); (3) for grazing animals, ingestion of soil; (4) inhalation of contaminated dust. For each animal product, output of radionuclides is: (1) biological elimination by metabolism. Consequently, the transfer from ingested water and/or plants and/or soil is described by a Transfer factor F_{ai} and by an elimination rate $\lambda_{bio,ai}$. The PDFs of these parameters were determined as follows (Table 11):

- *Transfer factors for animal products*. Several experts in animal processes were consulted within the framework of the EC Project, 'Probabilistic accident consequence uncertainty analysis' (Anonymous, 1997) and proposed PDFs for transfer factors of Cs, Sr and I to milk and beef meat (only milk for I). The 'aggregated expert distributions' obtained on this basis were considered in this article. For the other radionuclides, PDFs are based on those proposed by Hoffman et al. (1984), Ng (1982) and Ng et al. (1977, 1982b). For Co and Mn in milk, Ng (1982) considered the effect of chemical forms of the element on the transfer to animals, distinguishing 'radioisotope tracer data' and 'stable element concentration', respectively. More recent data (Van Bruwaene et al., 1984; Voigt

[2] In the applications presented hereafter, two animal products were considered: cow milk and beef meat.

Table 9
Translocation factors experimentally observed for Cs

Plant	Translocation factor	Reference
Barley	0.09	Pröhl et al. (1990)
Wheat	0.07	
Potatoes	0.45	
Rye	0.19–0.27	Aarkrog (1969)
Oats	0.16	
Barley	0.14–0.16	
Wheat	0.03–0.12	
Beans	0.16	Macacini et al. (2002)
Apple	0.17–0.42	References reviewed in Carini (2001)
Grapewine	0.018–0.096	
Pear	0.128	
Strawberry	0.112–0.2	

et al., 1988) showed that 'radioisotope tracer data' would better represent transfer of radiocobalt or radiomanganese to milk; consequently, we considered these data to achieve a uniform PDF. For Ag in milk, only a best estimate value was proposed by Ng (1982); such a value is in the order of magnitude as the value proposed for Cs. Consequently, the PDF obtained for Cs in milk was also applied to Ag. For iodine in meat, the recent review of Thorne (2003) was considered. Silver data published by Handl et al. (2000) showed high transfer factors for some organs in pigs and consequently the PDF proposed by Ng (1982) was extended to higher values to take into account the potential consumption of more contaminated organs. For Co and Mn in meat, the PDF proposed by Ng (1982) was considered.

- *Biological elimination rate for animals*. In the context of the previously mentioned EC Project, 'Probabilistic accident consequence uncertainty analysis' (Anonymous, 1997), experts in animal processes were also consulted on biological half-lives of Cs, Sr and I in meat; the 'aggregated range factors' were used to achieve uniform PDFs. The allometric approach proposed by Highley et al. (2003) and the data reported by Coughtrey et al. (1983) show that biological half-lives of Co

Table 10
Soil to plant transfer factors for iodine

Plant	Soil to plant transfer factor	Reference
Cereals	Mean value: 1.6×10^{-3} Range: 5×10^{-4}–2×10^{-2}	Shinonaga et al. (2001)
Fruits	Range: 4.1×10^{-4}–3.1×10^{-2}	References reviewed in Carini and Bengtsson, 2001
Chinese white cabbage	Mean value: 6.3×10^{-2} Range: 2.4×10^{-2}–1.9×10^{-1}	Yu et al. (2000)
Garden-like plot experiment	Range: 2.4×10^{-2}–1.9×10^{-1}	Sheppard et al. (1993)
Food crops and forage	^{129}I: Geometric mean: 3×10^{-2} Geometric Standard deviation: 4.9×10^{-2} ^{127}I: Geometric mean: 3.3×10^{-1} Geometric Standard deviation: 2.5×10^{-1}	Robens et al. (1988)

Table 11
List of parameters in the model OURSON – animal parameters

Symbol	Unit	Definition	Equations	Value: ^{54}Mn	^{58}Co	^{60}Co	^{90}Sr	^{110m}Ag	^{131}I	^{134}Cs	^{137}Cs	References
$X_{soil/past}$	–	Ingestion ratio soil-pasture for grazing animals	35	U (1.5×10^{-2}; 0.2)								Beresford and Howard (1991), Fries et al. (1982), Green and Dodd (1988), Healy (1968), Thornton and Abrahams (1983)
F_{ai}	j kg^{-1}	Transfer factor to milk	35	U (3×10^{-4}; 3.5×10^{-4})	U (8.7×10^{-5}; 1.1×10^{-4})		LN (4.3×10^{-4}; 4.8×10^{-3})	LN (10^{-3}; 2.4×10^{-2})	LN (5.3×10^{-4}; 3.7×10^{-2})	LN (10^{-3}; 2.4×10^{-2})		Ng et al., 1982a,b; Hoffman et al., 1984
F_{ai}	j kg^{-1}	Transfer factor to beef meat	35	LN (4.2×10^{-5}; 3.4×10^{-3})	LN (1.5×10^{-3}; 6.3×10^{-2})		LN (1.6×10^{-4}; 6.2×10^{-2})	T (3.3×10^{-4}; 2×10^{-3}; 0.1)	LU (8×10^{-4}; 2×10^{-2})	LN (3.1×10^{-3}; 9.1×10^{-2})		Ng et al., 1982a,b; Hoffman et al., 1984
$\lambda_{bio,ai}$	j^{-1}	Biological elimination rate for milk	35	LU (0.14; 1.4)	LU (0.14; 1.4)		LU (0.14; 1.4)	LU (0.14; 1.4)	LU (0.14; 1.4)	LU (0.14; 1.4)		See Table 12
$\lambda_{bio,ai}$	j^{-1}	Biological elimination rate for meat	35	U (2.3×10^{-2}; 6.9×10^{-2})	U (2.3×10^{-2}; 6.9×10^{-2})		U (8.7×10^{-4}; 1.4×10^{-3})	U (5×10^{-3}; 2×10^{-2})	U (1.4×10^{-3}; 3.45×10^{-3})	U (2.3×10^{-2}; 6.9×10^{-2})		See Table 12

and Mn in meat are close to those of Cs. Consequently, for these two radionuclides, the PDF obtained for Cs was considered. For Ag, data of Coughtrey et al. (1983) and Handl et al. (2000) were used to propose a uniform PDF. Biological half-lives data for milk are scarce and consequently, in order to compare values, information was also collected for elements not investigated in this study (Table 12). For some elements, two biological half-lives were obtained. However, it may be noted that the fast compartment is generally the compartment that contributes preponderantly to elimination from milk. If slow compartments are neglected, biological half-lives in milk are in the same order of magnitude for all the elements. Consequently, a common log-uniform PDF was selected for all the radionuclides investigated.

To simulate the ingestion of soil by animals, an 'Ingestion ratio soil-pasture for grazing animals' was considered. However, ingestion of soil by animals may depend on several factors such as site, season and species (Beresford and Howard, 1991; Thornton and Abrahams, 1983). Values reported in the literature were reviewed (Beresford and Howard, 1991; Fries et al., 1982; Green and Dodd, 1988; Healy, 1968; Thornton and Abrahams, 1983) and extreme values were used to define a uniform PDF.

3.8. Agricultural and human parameters – dose factors

3.8.1. Irrigation rates, schedule of cultures, alimentation of animals

PDFs for 'passage' irrigation rates, germination dates and harvest dates, and alimentation rates of animals were achieved considering local agricultural practices (inquiry in agricultural organisms of the Loire basin) and standard values published in the literature (Soltner, 1990). It has to be mentioned that grass and leafy vegetables were supposed to be harvested three times each year (standard agricultural practices).

3.8.2. Human habits

Specific human habits for people are reviewed in Anonymous (2002). For time budget near the river, local data and NCRP (1996) data were considered.

3.8.3. Dose factors

Dose factors for ingestion and inhalation are from the European Union Directive 96/29/EURATOM. For external exposure, dose factors proposed by US.EPA were considered.

4. Uncertainty/sensitivity analysis

A selection of some uncertainty/sensitivity analysis is presented in this paper to show how such analysis may help future model development. The objectives of such uncertainty and sensitivity analysis are remembered: (i) because empirical data are

Table 12
Biological half-lives in animal products

	Mn	Co	Sr	Ag	I	Cs	other	Reference
			500−800		200−500	10−30		Anonymous (1997)
		2.6 (1000 M)$^{0.24}$	107 (1000 M)$^{0.26}$		6.8 (1000 M)$^{0.13}$	3.5 (1000 M)$^{0.24}$		Highley et al. (2003)
Meat				pig: from 35 (liver) to 136 (brain)				Handl et al. (2000)
	41 (intermediate compartment)	6−60 (intermediate compartments)		58 (sc)		5.18 M$^{0.3}$		Coughtrey et al. (1983)
					0.5−1	1−2 (fc; 0.8) 10−20 (sc; 0.2)		Voigt et al. (1989)
Milk					1			Voigt et al. (1988)
					0.9−2.6			Vandecasteele et al. (2000)
	3.2 (fc;0.97); 51 (sc;0.03)	1.3 (fc;0.88); 23 (sc;0.12)					Cr: 1.7	Van Bruwaene et al. (1984)
							Fe: 2 (fc;0.95); 26 (sc;0.05) Tc: 0.14 (fc;0.67); 0.54 (sc;0.33) Mo: 2.5 Te: 1.9 (fc;0.85); 18 (sc;0.15) Ba: 2.1 (fc;0.77); 16 (sc;0.23) Zr: 1.6 Nb: 1.2	Johnson et al. (1988)

fc: fast compartment; sc: slow compartment; in parenthesis: contribution of the compartment to the elimination rate.

very scarce in the context of routine releases in freshwater, models constitute the only way to assess radiological doses to critical groups. However, all potential combinations of parameters included in the model must be tested in order to verify that the probability to exceed given target values is negligible. (ii) many pathways and associated parameters are introduced in such an approach. In order to define priorities for further research development (for future experimental works and/or refinement of models), it is necessary to identify which parameters are responsible for the output uncertainty.

In this paper, the uncertainty/sensitivity analysis focused exclusively on: (i) outputs for which validation data are available (bottom sediments and fish); (ii) the 'mean annual total dose' to critical groups. For this last output and for some radionuclides, the analysis was performed for two contrasted stations: Beaulieu, situated downstream of the first NPP of the Loire river and where no sedimentation is expected, and Montjean, the last investigated station on the Loire river, where sedimentation may occur. The results of the uncertainty analysis for the output 'mean annual total dose' are presented in Table 13 and those of the sensitivity analysis are presented in Table 14.

4.1. Confrontation to empirical data

The activity of γ emitters in several environmental compartments is extensively monitored once a year around each of the NPPs situated along the Loire river. However, because levels of radionuclides concentrations in routine releases are low, most of measurements are below the detection limits. Consequently, calculations can be compared to empirical data only for few compartments (i.e. bottom sediments

Table 13
Uncertainty analysis for several outputs

Output	5th percentile	Mean	95th percentile	Ratio 95th/5th percentiles	PDF
^{60}Co – Total dose – Mean value over 6 years – Montjean – Unit: Sv y^{-1}	2.1×10^{-9}	8.6×10^{-9}	6.7×10^{-8}	32	log-normal
^{60}Co – Total dose – Mean value over 6 years – Beaulieu – Unit: Sv y^{-1}	4.5×10^{-9}	1.6×10^{-8}	5.2×10^{-8}	12	log-normal
^{54}Mn – Total dose – Mean value over 6 years – Montjean – Unit: Sv y^{-1}	4.1×10^{-11}	2.1×10^{-10}	2.4×10^{-9}	59	log-triangular
^{54}Mn – Total dose – Mean value over 6 years – Beaulieu – Unit: Sv y^{-1}	5.5×10^{-11}	1.3×10^{-10}	5.3×10^{-10}	9.6	log-normal
^{137}Cs – Total dose – Mean value over 6 years –Montjean – Unit: Sv y^{-1}	4.9×10^{-9}	1.5×10^{-8}	5.2×10^{-8}	11	log-triangular
^{110m}Ag – Total dose – Mean value over 6 years – Montjean – Unit: Sv y^{-1}	6.5×10^{-9}	1.9×10^{-8}	9.7×10^{-8}	15	log-triangular
^{131}I – Total dose – Mean value over 6 years – Montjean – Unit: Sv y^{-1}	5.9×10^{-10}	1.9×10^{-9}	8×10^{-9}	13.6	log-uniform
^{90}Sr – Total dose – Mean value over 6 years – Montjean – Unit: Sv y^{-1}	1.2×10^{-9}	2.7×10^{-9}	6.4×10^{-9}	5.3	log-normal

Table 14
Sensitivity index of the most sensitive parameters for the following outputs

Symbol	Definition	(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)
R^2 tot	Total determination coefficient	0.87	0.89	0.89	0.88	0.89	0.89	0.95	0.91
$K_{d,w}$	Distribution coefficient in the river	0.18	0.45	0.07	0.29	0.05	0.026		
W_c	Settling velocity of particles	0.15		0.17			0.15		
τ_d	Critical deposition shear stress	0.20		0.24			0.15		
τ_r	Critical resuspension shear stress	0.02		0.03			0.017		
n_{Ma}	Manning's coefficient	0.31	0.014	0.34			0.25		
B_f	Water-to-fish transfer factor		0.016	0.03	0.05	0.45		0.72	0.08
$\lambda_{bio,f}$	Biological elimination rate for fish					0.012			0.016
ψ_{riv}	Percentage of time near the river	0.013		0.02					
$K_{d,soil}$	Distribution coefficient in the soil	0.01	0.01						
$\mu_{v,d}$	Foliar uptake coef. of dissolved radionuclides		0.022	0.007	0.04	0.02	0.014	0.05	0.04
$f_{tr,vi}^{max}$	Maximum translocation factor for fruit vegetables	0.03	0.9	0.017	0.04	0.09	0.15	0.07	0.27
$t_{tr,vi}^{max}$	Time before harvest when maximum translocation is reached (fruit)		0.01			0.018	0.013	0.05	
b_{vi}	Slope of the kinetic evolution of the translocation factor for fruit vegetables		0.01			0.01	0.01		0.011
$f_{tr,vi}^{max}$	Maximum translocation factor for root vegetables				0.025				
$Y_{vi,har}$	Biomass of the consumable part of fruit vegetables at harvest	0.03	0.1		0.04	0.08	0.06	0.03	0.26
$K_{d,w}$	Distribution coefficient in the soil				0.023				
$Q_{pi,m}$	Daily consumption of fruit vegetables by man	0.016	0.05	0.015	0.021	0.05	0.04	0.01	0.15
τ_{vi}	Water content of the consumable part of fruit vegetables	0.013	0.03			0.02	0.022		0.08
$Q_{pi,m}$	Daily consumption of fish by man					0.019		0.04	

(1): Mean annual dose – ^{60}Co – Montjean – With interactions – Rank transformation. (2): Mean annual dose – ^{60}Co – Beaulieu – With interactions. (3): Mean annual dose – ^{54}Mn – Montjean – With interactions – Rank transformation. (4): Mean annual dose – ^{54}Mn – Beaulieu – With interactions. (5): Mean annual dose – ^{137}Cs – Montjean – With interactions. (6): Mean annual dose – ^{110m}Ag – Montjean – With interactions – Rank transformation. (7): Mean annual dose – ^{131}I – Montjean – With interactions. (8): Mean annual dose – ^{90}Sr – Montjean – With interactions.

and fish). Furthermore, ^{137}Cs was not included in this analysis because routine releases from NPPs are not a major source in comparison to other past sources (e.g. Chernobyl). The results of the comparison between empirical data and calculation results are summarized in Table 15: the number of samples on which measurements were performed is indicated in the column 'Number of monitoring data'; among these samples, only some of them gave values higher than the detection limit and their number is indicated in the column, 'Number of monitoring data higher than the detection limit'. It was observed that only some data are actually higher than the detection limit and can be used for comparison with calculations. Consequently, data used for such a comparison are 'maximum' concentrations found over the investigated period and they must be compared to the PDF representing the distribution of maximum concentrations. Therefore, empirical data are situated on this PDF; in particular, in Table 15, it is indicated the percentile the empirical data correspond to. If the percentile is 0.5, that means that the empirical data corresponds to the most probable value calculated by the model (i.e. the mode of the distribution). If the percentile is in the range 0.1–0.9, that means that the empirical data is actually included in the 80th confidence interval. Results reported in Table 15 show that all available empirical data are actually included in the confidence level calculated by the model. Consequently, the orders of magnitude proposed by the model for these compartments can be considered satisfactory.

4.2. ^{60}Co

For ^{60}Co, the uncertainty/sensitivity analysis was also performed at Beaulieu and Montjean. Best estimate calculations showed that sedimentation processes are low at the first station as compared to the second one. The 90th confidence interval ranges from about 12 at Beaulieu to about 30 at Montjean. In Table 13, the most sensitive

Table 15
Comparison between empirical data and the PDF of maximum value on the investigated period

	Station	Radionuclide	Number of monitoring data	Number of monitoring data higher than the detection limit	Percentile of the empirical data on the PDF of maximum value over 6 years
Bottom sediments	Ouzouer	^{60}Co	14	6	0.25
	Nouan	^{60}Co	24	11	0.32
		^{110m}Ag	24	11	0.23
	Bertignolles	^{60}Co	16	10	0.54
		^{110m}Ag	16	7	0.45
Fish	Beaulieu	^{60}Co	19	1	0.74
		^{134}Cs	19	1	0.8
	Ouzouer	^{134}Cs	19	1	0.42
	Nouan	^{60}Co	23	1	0.72
	Bertignolles	^{60}Co	25	2	0.87
		^{58}Co	25	2	0.87
		^{134}Cs	25	2	0.49

parameters are listed according to their sensitivity index (measuring the loss of correlation between inputs and the investigated output – see the definition in the Section 'Input data and methods'). It was observed that the hierarchy of the most sensitive parameters depends on the station investigated. At Montjean, where best estimate calculations showed that sedimentation may occur in some conditions, parameters describing deposition processes in bed sediments are very sensitive (i.e. the Manning's coefficient, the critical deposition shear stress and the settling velocity of particles). It may be also observed that, in such a case, the distribution coefficient in the river plays a major role on the uncertainty, as shown by a significant sensitivity index (18%). Regression calculations accounting for interactions showed that the main effects on uncertainty are linked to interactions: for example, the distribution coefficient $K_{d,w}$ interacts with the Manning's coefficient and the critical deposition shear stress and the effect of the the Manning's coefficient is also essentially due to interactions. On the contrary, at Beaulieu, where no sedimentation is expected because of the configuration of the river, parameters describing translocation (sensitivity index of the Maximum translocation factor = 90%), as well as the distribution coefficient in the river (Sensitivity index = 45%), are the most sensitive parameters because they influence aerial transfer to vegetables. These observations explain also why the uncertainty at the Montjean station is higher than at the Beaulieu station: indeed, the uncertainty on deposition processes increases the global uncertainty on the total dose at Montjean. Consequently, to improve the dose assessment of ^{60}Co routinely released by NPPs, it appears essential to focus the research priorities on two axis: a good description of the sedimentation processes in the river, including a reduction of uncertainty on the $K_{d,w}$ value and a better investigation on translocation processes in consumable plants (fruit vegetables). Consequently, in further use of the OURSON model, refinements on the exchanges processes at the interface water-SPM (described in this paper by the sensitive parameter $K_{d,w}$) will be tested.

4.3. ^{54}Mn

^{54}Mn showed a similar behaviour as ^{60}Co: (i) a higher uncertainty at Montjean where the uncertainty on deposition processes is predominant; (ii) for the Montjean station, a high sensitivity of the parameters describing the deposition of sediments in the river (i.e. the Manning's coefficient, the critical deposition shear stress and the settling velocity of particles) and of the distribution coefficient in the river; (iii) at the Beaulieu station, the most sensitive parameter is the distribution coefficient in the river, followed by parameters describing the aerial transfer to plants. Consequently, research priorities are similar to those previously identified for ^{60}Co.

4.4. ^{137}Cs

For ^{137}Cs, the water-to-fish transfer factor is the most sensitive parameter. Consequently, it should be necessary to refine the "fish" model through the introduction of cofactors allowing to reduce the uncertainty on the water-to-fish transfer factor for example. It must also be underlined that some parameters describing translocation processes to fruit vegetables are also significantly sensitive (for example the maximum translocation factor, showing a Sensitivity index of about 10%) and that improvements on this pathway should also be investigated.

4.5. ^{131}I

The uncertainty of the total dose due to ^{131}I is almost exclusively linked to the water-to-fish transfer factor, as showed by its high Sensitivity index (72%). This result can partly be explained by the short radioactive half-life of ^{131}I and to the subsequent rapid loss in terrestrial compartments.

4.6. ^{90}Sr

^{90}Sr showed the lowest uncertainty among the investigated radionuclides, the 90th confidence interval being about 5. The aerial transfer to fruit vegetables is the most sensitive process and the most important parameters are the maximum translocation factor and the biomass at harvest. However, for ^{90}Sr, best estimate calculations showed that the relative importance of root uptake on one hand and interception-translocation are in the same order of magnitude and that in some cases, the soil-to-plant transfer factor is a sensitive parameter (results not shown in this paper). Consequently, to improve the dose assessment due to ^{90}Sr, all the parameters describing the transfer to plants (though interception–translocation pathway or soil–root pathway) should be better investigated.

4.7. ^{110m}Ag

For ^{110m}Ag, both parameters describing deposition processes in the river (Manning's coefficient, settling velocity of particles and critical deposition shear stress) and translocation to vegetables (maximum translocation factor) showed high Sensitivity index (between 15 and 25%). This result showed that the uncertainty is linked to both of these processes and that, as for Co, priorities should focus on these pathways. It may be also observed that, when performing the sensitivity analysis, a rank transformation was necessary. Consequently, the sensitivity of some parameters (those describing deposition of particles in the river) is not linear and the knowledge of "cut-off" values (i.e. when deposition begins) is important.

5. Conclusions

To assess radioecological doses to critical groups in the context of routine releases in rivers by NPPs, it is necessary to use models simulating the transfer of radionuclides in several environmental compartments. One of the difficulties in such approaches is that these models require a great number of parameters which are highly uncertain (because of the natural variability and/or the lack of knowledge).

Consequently, it is necessary to determine a confidence level for the evaluations and to identify major scientific gaps. Parametric uncertainty/sensitivity analysis is one of the ways to answer such questions. In this paper, probability density functions were defined for each of the parameters and a complete uncertainty/sensitivity analysis was performed. Such an analysis showed that research priorities are: (i) an improvement in the evaluation of the deposition of radionuclides in bed sediments and subsequent external exposure; (ii) an improvement of aerial contamination pathways of consumable vegetables.

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