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SCK•CEN-BLG-1039

RADIOECOLOGICAL SURVEY OF THE BELGIAN UPPER PART OF THE MEUSE RIVER DURING THE TRIENNIAL LOW WATER.

PERIOD 28 SEPTEMBER – 30 SEPTEMBER 2004
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January, 2007

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1. INTRODUCTION

The Belgian radiological surveillance programme around the nuclear power plant sites started in 1965, before the commissioning (18/10/66) of the French-Belgian nuclear plant (SENA - 300MW_e) at Chooz (France), near the border between the two countries. Under the joint effort of the former Ministry of Public Health and the Belgian Nuclear Research Centre (SCK•CEN), an environmental sampling programme has been set up to control the radioactive contamination levels in different compartments and bio-indicators of the two most important vectors of radioactive releases, air and water. In the framework of this programme, water, sediments and biological samples were routinely collected from the Meuse river.
Every three years, when the dams on the upper Meuse river are opened to allow heavy maintenance works on the sluices and banks, specific and more extensive sampling campaigns are organised, taking advantage of the low water. This particular situation facilitates sampling and, hence, provides the opportunity to enlarge the sample set and the sampled quantities. These triennial sampling campaigns have been performed regularly since 1968 and the data collected during these periods have provided directly useful information to guide the choice of the most relevant bio-indicators used in the routine surveillance programme. A synopsis of results has been published by [Kirchmann et al., 1997].

The triennial campaigns in September 1992 [Vangelder *et al*., 1994] and September 1995 [Vangelder *et al*., 1997] offered a good opportunity to evaluate the response of the Meuse ecosystem to the shut-down of the operation of the reactor of Chooz-A on 30/10/1991, and before the commissioning of two new 1455 MW_e nuclear reactors, Chooz-B1 and B2. The first unit of Chooz-B reached its first criticality on 25/07/96 and was connected to the electricity grid on 30/08/96; the second unit was made critical on 10/03/97 and connected to the grid on 09/04/97. The 1998 sampling campaign allowed to evaluate the impact of the new reactors on the Meuse ecosystem [Hardeman *et al*, 2001].

This triennial campaign, carried out in September 2004, allowed to confirm the impact of the commissioning of the new units on the Meuse. Upon demand of the Federal Agency for Nuclear Control (FANC), the programme has been modified considerably, as the activity levels observed in previous years (1998 - 2001) proved to be very low in most of the compartments, often even below limits of detection. Upon demand of FANC, ^{7}Be has been added to the library used in gamma-spectrometry. ^{7}Be is produced globally in the upper atmosphere; its production rate depends on the latitude and solar activity; its availability on the earth is also influenced by climate and shows seasonal changes; an important influencing parameter is precipitation [Gonzalez-Gomez et al., 2006]. It is sometimes used for migration and soil erosion studies as it is highly particle reactive [Doering *et al*., 2006].

2. SAMPLING AND SAMPLE PROCESSING

2.A. General

The implementation of the sampling programme needed a close collaboration between the two laboratories directly involved in the field work, namely:

- the Institute Environment Health and Safety of the SCK•CEN (Mol),
- the Freshwater Ecology Unit of the Faculty Notre-Dame de la Paix (Namur).

After identification, the samples are labelled and transported to the Mol laboratory for preparation in view of the radioactivity measurements.

The period of sampling extended from 28 September to 30 September 2004 and covered a section of about 40 km of the river Meuse (see fig. 1), from the dam of Ham-sur-Meuse, upstream of the Chooz nuclear power plant to Hun.

The choice of the plant and animal species as well as the locations of the sampling places were based on the previous experience and availability of the species. France did not participate to the lowering of the Meuse water level and has maintained high waters in the French part of the Meuse. As a consequence, the water level at the sites of Ham-^{s}/Meuse and Givet remained high, preventing sampling of bottom sediments in Givet.

2.B. Sampling of bottom sediments

Bulk bottom sediments were collected at Ham-sur-Meuse, Heer, Waulsort, Dinant and Hun. Fine sediments (~ 7000 g) were sampled by hand between pebbles from the top sedimentation layer (up to 3 cm depth) in accessible zone of the river's bed abandoned by the low water stream.

At Heer and Hastière, sediments were collected per 10 cm of depth to get information on the depth profile. Deeper sampling than 60 cm (Heer) or 80 cm (Hastière) was impossible due to the fact that soil was saturated with water at that level.

2.C. Sampling of aquatic organisms

Plant and animal samples were put in polyethylene bags, labelled and transported to the laboratory for analyses.

2.C.1. Plant species:

One aquatic moss (*Cinclidotus*) and one higher plant (*Carex*) were hand picked at the following places: Ham-sur-Meuse (France, upstream NPP), Givet (France, downstream NPP), Waulsort, Dinant, Yvoir and Hun. There was no Cinclidotus available in Dinant and no Carex in Yvoir.

2.C.2. Animal species:

Fresh water molluscs (*Anodonta anatina* and *Dreissena polymorpha*) were hand picked at 5 locations according to availability. Fish samples were collected at Heer, Hastière, Waulsort, Dinant and Hun with the help of the Faculty Notre Dame de Namur people, using electric catch.

The list of animal bio indicators is provided in table 1.

Scientific name	French name	Dutch name	English name
Anodonta anatina	anodonte	vijvermossel	pond mussel, duck mussel
Dreissena polymorpha	dreissène, moule zébrée	driehoeksmossel	zebra mussel
Alburnus alburnus	ablette	alver	bleak
Leuciscus cephalus	chevaine	kopvoorn	chub
Rutilus rutilus	gardon	blankvoorn	roach
Perca fluviatilis	perche commune	baars	perch

Table 1: Animal species used as bio indicators of the radioactive contamination of the Meuse ecosystem.

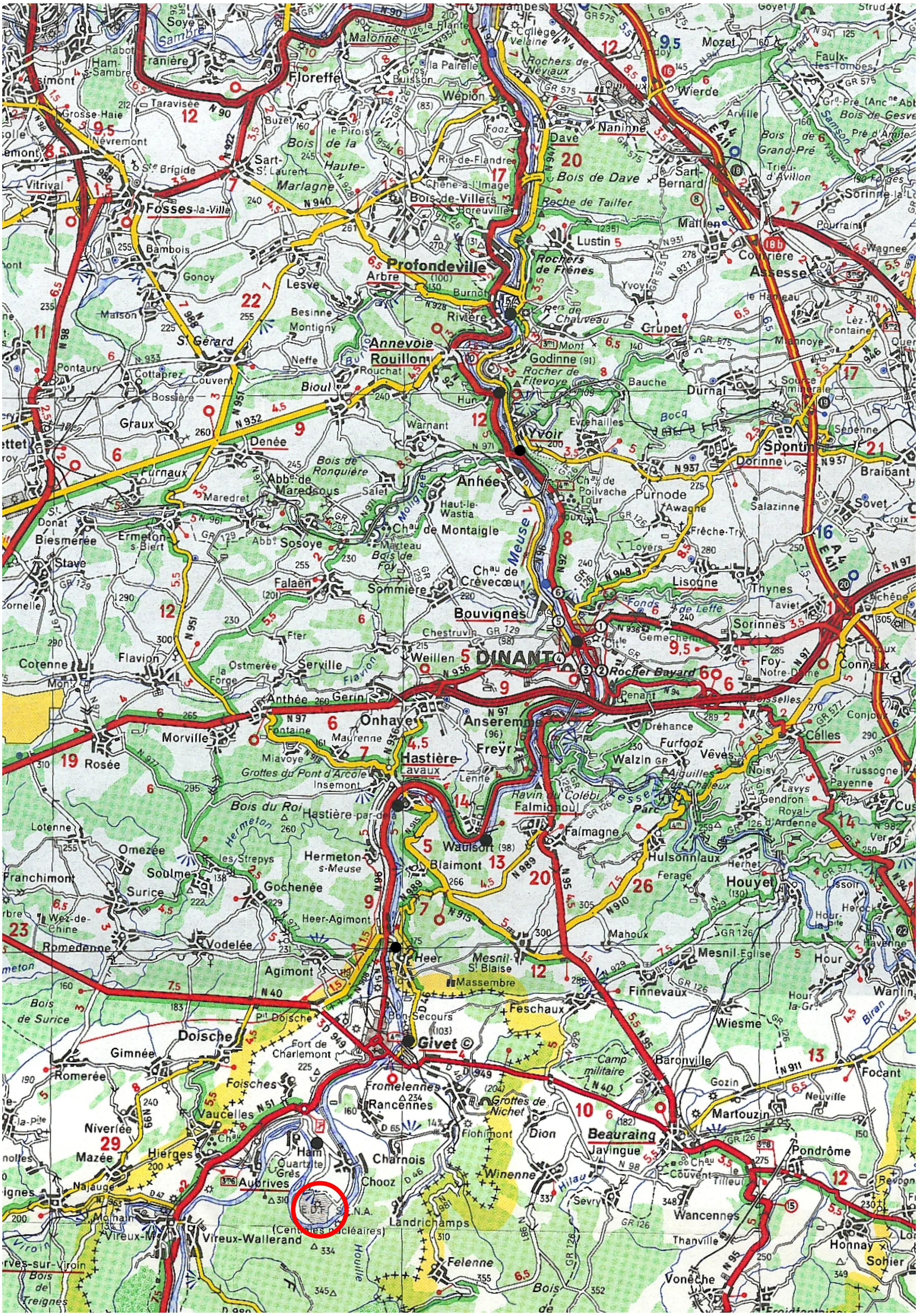

Fig. 1: The Meuse river between Chooz and Namur. The black dots indicate the sampling sites.

2.D. Sample processing

Sediment samples were strained out and the sample was dried in an oven at 100°C. The dried samples were crushed to fine powder for radiochemical analysis and γ-spectrometry.

Plant and animal samples were washed thoroughly to free them from attached sand and silt. The mussels (Anodonta) and fish were carefully dissected to separate the soft tissues/muscle from their shells/bones. Wet weights of the samples were recorded. Afterwards the samples were dried in an oven at 100°C to obtain dry weights. The dry samples were ground for further radiochemical analysis and γ-spectrometry.

3. METHODS OF MEASUREMENTS

Twenty-one radionuclides (table 2) were analysed in the different samples collected from the upper Meuse river. They include natural radioelements, artificial radionuclides potentially originating from the nuclear fuel cycle, as well as radionuclides (namely ^{137}Cs and ^{90}Sr) that were widely dispersed in the past as a consequence of atmospheric bomb testing (global fallout) and, more recently, of the Chernobyl accident.

Nuclide	Origin	Detection method
^{3}H (OBT)	cosmogenic & fission product	liquid scintillation counting on combustion water
^{7}Be	cosmogenic	γ-spectrometry
^{40}K	primordial radionuclide	γ-spectrometry
^{54}Mn	activation product	γ-spectrometry
^{57}Co	activation product	γ-spectrometry
^{58}Co	activation product	γ-spectrometry
^{60}Co	activation product	γ-spectrometry
^{65}Zn	activation product	γ-spectrometry
^{90}Sr	fission product	gas proportional counter after radiochemical separation
^{110m}Ag	activation product	γ-spectrometry
^{134}Cs	fission product	γ-spectrometry
^{137}Cs	fission product	γ-spectrometry
^{226}Ra	primordial radionuclide	γ-spectrometry
^{228}Th	primordial radionuclide	γ-spectrometry
^{232}Th	primordial radionuclide	γ-spectrometry
^{234}U	primordial radionuclide	radiochemistry, α-spectrometry
^{235}U	primordial radionuclide	radiochemistry, α-spectrometry
^{238}U	primordial radionuclide	radiochemistry, α-spectrometry
^{241}Am	activation product	radiochemistry, α-spectrometry
^{238}Pu	activation product	radiochemistry, α-spectrometry
^{239}Pu	activation product	radiochemistry, α-spectrometry

Table 2: List of measured radionuclides with reference to their origin and detection mode.

3.A. Radiochemical analysis

3.A.1. Tritium

Organically bound tritium (OBT) from plant and animal samples was measured after combustion of dried samples by determination of tritium in the water produced by oxidation of the organic matter.

The tritium free water in plant and animal tissues (TFWT) was not separated, nor measured, as the result would not be really of significance due to the extreme mobility of water in the biosphere.

3.A.2. Strontium-90

^{90}Sr is determined by low-level beta measurements after separation on Sr spec column from EIChroM®.

Strontium is separated from the bulk environmental sample using Sr spec column from EIChroM®, using a method adapted from the procedure by IAEA [2000]. The ^{90}Sr eluted from the column is then kept for two weeks to let its daughter nuclide ^{90}Y grow in. ^{90}Y is then precipitated and measured in a low background gas proportional counter.

3.A.3. Uranium, Americium and Plutonium

^{238}Pu and ^{239}Pu, ^{241}Am and the various radioisotopes of uranium (^{234}U, ^{235}U and ^{238}U) are determined by α-spectrometry subsequent to a radiochemical separation and purification on anion exchange resin: BioRad® AG-1X2 for the Pu's and TRU.spec column from EIChroM® for the other isotopes.

3.B. Gamma-spectrometry

Gamma spectrometry allows to determine the presence and to quantify the activity of a vast number of radionuclides for a relatively limited effort and cost. However, it is worth mentioning that gamma spectrometry also shows some disadvantages in assessing some of the naturally occurring radionuclides. The following radionuclides were looked for using high-resolution γ-spectrometry with a germanium detector: ^{7}Be, ^{40}K, ^{54}Mn, ^{57}Co, ^{58}Co, ^{60}Co, ^{65}Zn, ^{134}Cs, ^{110m}Ag, ^{137}Cs, ^{226}Ra, ^{228}Th, ^{232}Th.

The direct γ-emission of ^{226}Ra is very limited; even the only suitable gamma-ray at about 186 keV has a low intensity, and it interferes with the predominant gamma-ray of ^{235}U. Therefore, one is very often obliged to use radiation emitted by daughter nuclides such as ^{214}Pb (several γ-emissions, e.g. 352 keV) and ^{214}Bi (multiple γ-emissions, including 609 keV and 1764 keV). The main problem here is the risk of ^{222}Rn exhalation, which may perturb the equilibrium between mother- and daughter element activities. Adding activated coal and using tight beakers for the analysis is the best solution, but even then some uncertainty remains.

In the decay of ^{232}Th, no suitable γ are emitted. Therefore, one has to rely on γ-rays emitted by ^{228}Ac, daughter of ^{228}Ra, which itself is a daughter nuclide of ^{232}Th. However, due to the long lifetime (5.75 years) and to a higher solubility of ^{228}Ra, one can not be absolutely sure that the natural equilibrium one expects is always present in the collected sample. In samples from a natural origin, one does not expect very huge differences, but clearly in samples originating from chemical factories, the results for ^{228}Ra are not always representative for ^{232}Th.

^{228}Th can be measured directly by γ-spectrometry, but the main peak at 84 keV is not always suitable due to a relatively high background in that part of the spectrum; furthermore, analysis may be more difficult due to the presence of X-rays of heavy elements, and at this energy, inhomogeneity may also influence the accuracy. Therefore, one usually relies on daughter nuclides for the analysis. This shows a similar problem, as ^{220}Rn might also perturb the analysis. However, the lifetime of ^{220}Rn is much shorter as compared to ^{222}Rn, which strongly limits the inaccuracy that may be induced. In natural samples, one expects generally equilibrium between ^{232}Th, ^{228}Ra and ^{228}Th and their daughter products. The comparison between the ^{228}Th and ^{228}Ra activities may serve as a good indicator to check whether equilibrium indeed is present.

3.C. Presentation of the data

The radioactivity data presented in the tables represent the radioactivity concentrations measured in the samples or a representative fraction of the sample collected at a single sampling site. Detection limits in gamma-spectrometry are computed according to the Currie formalism [Currie, 1968] with α and $\beta = 0.05$. The variance of the background is determined in a ROI (Region of interest) of 6 FWHM wide centred around the considered peak centroids. When the radioactivity content in a sample is not significant, the detection limit is provided. Detection limits for a given radioisotope are not constant but vary from one sample to another, namely, according to the quantity of material used for the measurement, the counter efficiency, the counting time, and interferences with other radionuclides present in the sample. Only the uncertainty due to counting statistics expressed at the 1σ confidence level are reported.
In the analysis and discussion of the results, averages were calculated for specific radioisotopes and type of sample. To calculate these mean values, only the significant data were used, since statistics can not handle detection limits.
The distance (in km) given in tables refers to the distance between the sampling site and the Chooz nuclear power plant.

4. RADIOACTIVITY LEVELS MEASURED

4.A. Natural radioactivity

4.A.1. Sediments

The concentration levels measured for natural radionuclides in bulk bottom sediments from the Meuse river at different distances from the Chooz-B nuclear plant are reported in table 3. Table 4 shows the results of the deeper layers. All measurements in sediments, except Be-7 in layer samples, are well above detection limits.

- ^{40}K radioactivity levels in bottom sediments are fairly homogeneous. The radioactivity level corresponds fairly well with the average level in soils (about 410 Bq kg^{-1}, within a range from 70 to 900 Bq kg^{-1} [UNSCEAR, 2000]). The total K^+ concentration was estimated to 1.33 ± 0.30 % of the sediment dry weight.
- ^{238}U and its daughter ^{234}U show, as expected, similar activities. The ratio between these two U-isotopes amounts to 1.01 ± 0.05, corresponding to the expected ratio (1 at equilibrium) in natural uranium. The average ^{238}U-radioactivity in sediments is about 20.9 ± 5.7 Bq kg^{-1} dw well in the range of values reported for most soils (16 to 110 Bq kg^{-1} [UNSCEAR, 2000]). The mean ^{234}U is 21.0 ± 5.3 Bq kg^{-1} dw.
- ^{226}Ra is found in radioactivity concentrations close to that of ^{238}U, its parent, at 34.4 ± 7.0 Bq kg^{-1} dw. ^{235}U is present in the Meuse sediment at an average activity concentration of 1.2 ± 0.4 Bq kg^{-1}. Its mean ratio to the activity concentration of ^{238}U (0.06 ± 0.02) is higher than the expected ratio in natural U (0.047), but this calculated ratio is associated to such a high uncertainty that it is not significantly different from the expected ratio in natural ores.
- ^{232}Th radioactivity levels are higher than those measured for ^{238}U. This agrees with the average values reported for igneous, sedimentary and limestone rocks [UNSCEAR, 2000]. The ^{228}Th value is of the same order of magnitude as those for ^{232}Th, in agreement with the expected 1:1 ratio in most soils and waters [Eisenbud, 1987].

Location	Dist. (km)	^{7}Be Bq kg^{-1}	^{40}K Bq kg^{-1}	^{238}U Bq kg^{-1}	^{234}U Bq kg^{-1}	^{226}Ra Bq kg^{-1}	^{235}U Bq kg^{-1}	^{232}Th Bq kg^{-1}	^{228}Th Bq kg^{-1}	^{234}U/^{238}U	^{235}U/^{238}U
Ham s/Meuse	-3.0	< 40	256±25	12.4±1.0	13.5±1.1	22±2	0.9±0.3	28±3	30±3	1.09	0.07
Heer	12.0	37±7	392±39	26.0±2.0	25.0±2.0	37±3	1.2±0.4	43±4	48±4	0.96	0.05
Waulsort	19.5	41±7	446±43	17.9±1.4	17.5±1.0	39±4	1.0±0.3	44±4	44±4	0.98	0.06
Dinant	31.0	80±11	484±47	25.0±2.0	26.0±2.0	38±3	2.0±0.4	47±4	47±4	1.04	0.08
Hun	39.5	47±10	480±47	23.0±2.0	23.0±2.0	36±3	1.1±0.4	42±4	44±4	1.00	0.05
All sites mean ± sd		51±20	411±94	20.9±5.7	21.0±5.3	34.4±7.0	1.2±0.4	40.8±7.4	42.6±7.3	1.01±0.05	0.06±0.02

Table 3: Activity concentration of natural radionuclides in bottom sediments from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

- Be-7 activity was similar at all locations and below detection limit for the samples at different depth due to smaller amount of material for measurement, and due to decay if older sediments are at stake.
- The ^{226}Ra, ^{232}Th and ^{228}Th concentration is of the same order of magnitude in every layer.
- Total K content is comparable with bulk samples, 1.1± 0.1 Bq kg^{-1} dw for Heer and 1.1 ± 0.2 Bq kg^{-1} dw for Hastière.
- U was not measured in samples at different depth.

Location	Depth (cm)	^{7}Be Bq kg^{-1}	^{40}K Bq kg^{-1}	^{226}Ra Bq kg^{-1}	^{232}Th Bq kg^{-1}	^{228}Th Bq kg^{-1}
Heer	0-10	< 200	324±35	31±4	35±3	38±3
	10-20	< 260	330±36	33±4	37±4	42±4
	20-30	< 120	360±36	36±4	41±4	49±4
	30-40	< 80	346±34	34±4	39±4	45±4
	40-50	< 80	417±41	43±5	46±4	54±4
	50-60	< 100	360±35	35±4	39±4	46±4
mean ± sd all depths			356±33	35±4	40±4	46±5
Hastière	0-10	< 80	415±41	27±3	33±4	37±4
	10-20	< 80	413±49	28±4	35±4	40±4
	20-30	< 120	397±47	28±3	33±4	38±4
	30-40	< 80	294±75	20±5	23±6	26±4
	40-50	< 190	285±75	17±5	23±7	23±4
	50-60	< 160	308±81	20±5	24±7	28±4
	60-70	< 150	300±50	22±4	27±5	32±4
	70-80	< 200	300±55	21±4	29±5	31±4
mean ± sd all depths			339±58	21±4	26±4	30±5

Table 4: Activity concentration of natural radionuclides in bottom sediments per 10 cm of depth from the Meuse river at Heer and Hastière (in Bq kg^{-1} dw ± 1 counting σ).

4.A.2. Immersed Aquatic plants

Measurements of ^{40}K, ^{226}Ra and $^{228,232}Th$ were carried out on aquatic mosses collected from the upper Meuse river (table 5). The activity concentration for ^{40}K corresponds to a total K concentration of 1.59 ± 0.87 % in dry *Cinclidotus* sp material. The moss from Waulsort contains 2 to 3 times more ^{40}K.

The average ^{232}Th and ^{228}Th are respectively 31±12 and 28±17 Bq kg^{-1} dw.
U-isotopes were not measured in these samples.

species	Location	Dist. (km)	DM %	^{7}Be Bq kg^{-1}	^{40}K Bq kg^{-1}	^{226}Ra Bq kg^{-1}	^{232}Th Bq kg^{-1}	^{228}Th Bq kg^{-1}
Cinclidotus	Ham s/Meuse	-3.0	13.8	170±60	350±120	44±14	53±18	13±5
Cinclidotus	Givet	9.5	16.0	220±80	330±110	24±9	29±11	13±5
Cinclidotus	Waulsort	19.5	11.6	< 130	970±260	< 17	26±10	< 13
Cinclidotus	Yvoir	39.0	14.7	180±55	400±120	23±7	25±8	43±13
Cinclidotus	Hun	39.5	14.1	160±60	410±140	20±7	24±8	42±14

Table 5: Activity levels of natural radionuclides in aquatic moss from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

4.A.3. Vascular Aquatic plants

Natural radionuclides have been identified and quantified in the aerial part of Carex, partly immersed in the river Meuse (table 6).

- ^{40}K activity concentrations range from 530 to 850 Bq kg^{-1} dw, with the highest value in the Waulsort sample (as in moss). Based on these activity concentrations, the total K concentration estimated amounts to 2.2 ± 0.4% of the dry weight.
- Only three values regarding the ^{226}Ra radioactivity concentrations in aerial parts of vascular plants are significant. They are obtained in samples collected at Ham-s/Meuse Givet and Dinant and amount from 3 to 12 Bq kg^{-1} dw.
- Two plant samples (at Ham-s/Meuse and Givet) exhibit ^{232}Th and ^{228}Th concentrations in their aerial parts above detection limits. The ^{232}Th content is about 50 % higher than ^{228}Th, but given the high uncertainties their confidence intervals show some overlap.
- U-isotopes were not measured in these samples

Species	Location	Dist. (km)	DM %	^{7}Be Bq kg^{-1}	^{40}K Bq kg^{-1}	^{226}Ra Bq kg^{-1}	^{232}Th Bq kg^{-1}	^{228}Th Bq kg^{-1}
Carex	Ham s/Meuse	-3.0	38.2	180±50	580±160	5.8±1.7	6.9±2.2	3.8±1.2
Carex	Givet	9.5	39.4	160±40	530±150	3.1±1.0	3.3±1.1	2.3±0.8
Carex	Waulsort	19.5	26.1	< 300	850±350	< 30	< 50	< 18
Carex	Yvoir	39.0	45.5	160±70	780±280	12±5	< 30	< 9
Carex	Hun	39.5	34.0	130±60	690±260	< 19	< 40	< 16

Table 6: Activity levels of natural radionuclides in aerial part of aquatic plants from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

4.A.4. Molluscs

Natural radionuclide content in two common mollusc species used as bio indicators of the ecosystem contamination are given in table 7.

species	organ	Location	Dist. (km)	DM %	^{7}Be Bq kg^{-1}	^{40}K Bq kg^{-1}	^{226}Ra Bq kg^{-1}	^{232}Th Bq kg^{-1}	^{228}Th Bq kg^{-1}
Anodonta	flesh	Heer	12.0	10.4	< 30	110±20	15.0±2.5	17±3	7.9±1.4
	flesh	Hastière	15.0	9.7	< 40	100±20	12.0±1.5	19±2	6.4±1.0
	flesh	Waulsort	19.5	10.9	< 30	130±20	14.1±1.5	16±2	6.5±0.8
	flesh	Dinant	31.0	11.4	< 40	110±20	13.0±2.5	16±2	5.3±1.2
	flesh	Hun	39.5	9.3	< 40	100±20	22.0±3.5	23±4	11±2
Dreissena	whole	Ham s/Meuse	-3.0	45.5	< 30	29±15	3.6±1.0	< 8	1.9±0.7
	whole	Heer	12.0	45.6	< 30	< 40	3.9±1.5	< 9	< 4
	whole	Waulsort	19.5	46.9	< 30	< 30	7.2±2.0	11.0±3.0	1.9±0.8
	whole	Dinant	31.0	47.3	< 30	< 30	3.2±1.0	7.1±2.0	2.7±1.0
	whole	Hun	39.5	48.4	< 30	< 20	5.3±1.5	6.4±2.0	2.4±0.8

Table 7: Activity concentration of natural radionuclides in mussels (Anodonta, Dreissena) from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

- ^{40}K activity concentration was measured in the entire animal for Dreissena; only one sample was above detection limit (Ham) it amounted to 29 ± 15 Bq kg^{-1} dw. For Anodonta, the shell and flesh were separated. The average ^{40}K activity content in flesh amounted to 110 ± 12 Bq kg^{-1} dw. The shell was not measured.
- ^{226}Ra content in the entire animal amounted to 4.6 ± 1.6 Bq kg^{-1} dw in *Dreissena*. Its content in *Anodonta* flesh was 15.2 ± 4.0 Bq kg^{-1} dw.
- ^{228}Th concentrations in the entire shellfish were a factor 2 lower than ^{226}Ra values and amounted to 2.2 ± 0.4 Bq kg^{-1} dw in *Dreissena*. The ^{228}Th content in soft tissue of *Anodonta* was 7.4 ± 2.2 Bq kg^{-1} dw.
- ^{232}Th concentrations in the entire shellfish were of the same order of magnitude as ^{226}Ra values and amounted to 8.2 ± 2.5 Bq kg^{-1} dw in *Dreissena*. The ^{232}Th content in soft tissue of *Anodonta* was 18.2 ± 2.9 Bq kg^{-1} dw.
- U content was not measured.

4.A.5. Fish

Five mixed fish samples of different species (species per sample indicated in table 8) were collected from the upper Meuse river. Their content in natural radioisotopes is presented in table 8. Among the natural radionuclides, only ^{40}K and ^{226}Ra (in two samples) were above detection limit. The ^{40}K level was fairly homogeneous in fish flesh and amounted to 416 ± 25 Bq kg^{-1} dw. This value corresponds to a stable K content of 1.34 ± 0.08 % of dw.

^{226}Ra concentrations samples from Heer and Hastière amounted to 1.6 and 6.8 Bq kg^{-1} dw respectively.

U isotopes were not measured in fish.

species	organ	Location	Dist. (km)	DM %	^{7}Be Bq kg^{-1}	^{40}K Bq kg^{-1}	^{226}Ra Bq kg^{-1}	^{232} Bq kg^{-1}	^{228} Bq kg^{-1}
Alburnus Perca Leuciscus	flesh	Heer	12.0	27.1	< 130	380±60	1.6±0.5	< 6	< 3
Alburnus Perca Leuciscus Rutilus	flesh	Hastière	15.0	25.3	< 300	440±80	6.8±2.5	< 11	< 5
Alburnus Perca Rutilus	flesh	Waulsort	19.5	25.4	< 180	410±70	< 4	< 8	< 4
Alburnus Perca Leuciscus Rutilus	flesh	Dinant	31.0	26.5	< 140	420±90	< 3	< 6	< 3
Perca Leuciscus	flesh	Hun	39.5	25.9	< 300	440±80	< 6	< 13	< 6

Table 8: Activity concentration of natural radionuclides in fish from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

4.B. Artificial radioactivity

4.B.1. Sediments

The artificial radioactivity content in sediments from the upper Meuse river are given in tables 9, 10 and 11. ^{54}Mn, ^{57}Co, ^{58}Co ^{60}Co, ^{65}Zn, ^{90}Sr, ^{110m}Ag, ^{134}Cs and transuranic elements are systematically below detection limits.

^{137}Cs was detected in all sediment samples downstream NPP with an average activity of 8.5 ± 2.3 Bq kg^{-1} dw (within a range from 6 to 11 Bq kg^{-1} dw), more than a factor 2 lower than the samples taken in 1998. The values in 2001 (unpublished) were: 8.0 ± 3.1 Bq kg^{-1} dw (within a range from 4 to 14 Bq kg^{-1} dw)

^{60}Co was under detection limit. This value is lower than the contamination measured in 1998 because 7 to 8 years after the commissioning of the two new units the corrosion processes are stabilized; this value is also less than the 1995 and 1992 values, because of the improvement in the quality of metals used in this reactor generation.

The same finding is valid for ^{110m}Ag which was detected in sediment at only one location, and in much lower concentration than in 1998.

^{54}Mn measurements were statistically significant in several samples, while they did not exceed detection limits in 1998 (detection limit ranging from 0.4 to 1.9 Bq kg^{-1}). The concentration of ^{54}Mn remains however very low and the values are not incompatible with the 1998 data. The fact that significant values were obtained during this campaign can be attributed to improvements in measuring sensitivity.

Location	Dist. (km)	^{54}Mn Bq kg^{-1}	^{57}Co Bq kg^{-1}	^{58}Co Bq kg^{-1}	^{60}Co Bq kg^{-1}	^{65}Zn Bq kg^{-1}	^{110m}Ag Bq kg^{-1}	^{134}Cs Bq kg^{-1}	^{137}Cs Bq kg^{-1}
Ham s/Meuse	9.5	0.37±0.10	< 0.8	< 3	< 0.6	< 3	< 1	< 1	1.8±0.2
Heer	12.0	0.89±0.15	< 1	< 4	< 0.7	< 4	< 2	< 2	6.0±0.6
Waulsort	19.5	0.74±0.14	< 1	< 3	< 0.7	< 4	< 2	< 2	7.2±0.7
Dinant	31.0	0.66±0.14	< 1	< 4	< 0.8	< 4	< 2	< 2	11±1
Hun	39.5	0.74±0.18	< 2	< 5	< 1	< 5	1.1 ± 0.2	< 2	9.7±1.0
All sites downstream NPP mean ± sd		0.76±0.10							8.5±2.3

Table 9: Activity concentration of artificial radionuclides in sediments from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

Despite of high K_d values, none of the sediment samples exhibited measurable amounts of the transuranic elements. ^{238}Pu and ^{239}Pu levels do not exceed 1.1 Bq kg^{-1} dw and ^{241}Am values are less than 1.2 Bq kg^{-1} dw.

Location	Dist. (km)	^{241}Am Bq kg^{-1}	^{238}Pu Bq kg^{-1}	^{239}Pu Bq kg^{-1}
Ham s/Meuse	9.5	< 1.2	< 0.8	< 0.7
Heer	12.0	< 0.9	< 0.5	< 0.5
Waulsort	19.5	< 1.0	< 0.8	< 0.7
Dinant	31.0	< 0.8	< 1.0	< 0.9
Hun	39.5	< 0.9	< 1.1	< 1.1

Table 10: Activity concentration of transuranic radionuclides in sediments from the Meuse river (Bq kg^{-1} dw ± 1 counting σ).

Most of the results of the samples at different depth are below detection limit, except for ^{137}Cs. In Heer ^{137}Cs content is higher than in Hastière and shows an increasing concentration in deeper layers. In Hastière, the difference between layers is not statistically significant

Location	Depth (cm)	^{54}Mn Bq kg^{-1}	^{57}Co Bq kg^{-1}	^{58}Co Bq kg^{-1}	^{60}Co Bq kg^{-1}	^{65}Zn Bq kg^{-1}	^{110m}Ag Bq kg^{-1}	^{134}Cs Bq kg^{-1}	^{137}Cs Bq kg^{-1}
Heer	0-10	< 4	< 3	< 14	< 3	< 10	< 5	< 3	9.6±1.1
	10-20	< 4	< 3	< 17	< 3	< 13	< 6	< 4	9.7±1.1
	20-30	0.95±0.22	< 2	< 7	< 1	< 5	< 3	< 2	10.0±1.0
	30-40	0.86±016	< 1	< 5	0.43±0.08	< 4	< 2	< 1	13.0±1.5
	40-50	1.2±0.2	< 1	< 5	< 1	< 4	< 2	< 2	14.0±1.5
	50-60	< 2	< 2	< 6	< 1	< 5	< 2	< 2	14.0±1.5
Hastière	0-10	< 2	< 2	< 6	< 1	< 5	< 2	< 2	2.9±0.3
	10-20	0.59±0.18	< 2	< 5	< 1	< 5	< 2	< 2	3.8±0.5
	20-30	< 2	< 2	< 6	< 1	< 5	< 2	< 2	4.3±0.5
	30-40	< 2	< 2	< 8	< 2	< 6	< 3	< 3	1.9±0.6
	40-50	< 3	< 2	< 13	< 2	< 9	< 4	< 3	1.5±0.5
	50-60	< 3	< 2	< 10	< 2	< 8	< 3	< 3	2.2±0.7
	60-70	< 3	< 2	< 10	< 2	< 7	< 3	< 3	3.5±0.7
	70-80	< 3	< 2	< 14	< 3	< 9	< 4	< 3	2.4±0.6

Table 11: Activity concentration of artificial radionuclides in sediments per 10 cm of depth from the Meuse river
(in Bq kg^{-1} dw ± 1 counting σ).

4.B.2. Immersed Aquatic plants

The artificial radioactivity content in mosses from the upper Meuse river are given in table 12. All but four measurements lie below detection limits. The two radiocaesium isotopes, which were still detectable in mosses in 1995, could not be significantly detected in the samples collected in 1998 and remains hardly detectable in 2004.

species	Location	Dist. (km)	DM %	^{3}H Bq kg^{-1}	^{54}Mn Bq kg^{-1}	^{57}Co Bq kg^{-1}	^{58}Co Bq kg^{-1}	^{60}Co Bq kg^{-1}	^{65}Zn Bq kg^{-1}	^{110m}Ag Bq kg^{-1}	^{134}Cs Bq kg^{-1}	^{137}Cs Bq kg^{-1}
Cinclidotus	Ham s/Meuse	-3.0	13.8	< 10	< 6	< 4	< 9	< 6	< 16	< 6	< 5	< 6
Cinclidotus	Givet	9.5	16.0	< 20	< 5	< 3	< 8	< 5	< 13	< 6	< 5	4.0±2.0
Cinclidotus	Waulsort	19.5	11.6	< 11	< 9	< 5	< 14	< 10	< 30	< 9	< 8	< 9
Cinclidotus	Yvoir	39.0	14.7	< 10	3.8±1.3	< 3	< 5	< 3	< 11	< 4	< 6	3.5±1.3
Cinclidotus	Hun	39.5	14.1	< 11	< 5	< 5	< 12	< 8	< 30	< 8	3.8±1.7	< 9

Table 12: Activity concentration of artificial radionuclides in immersed aquatic plants from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

4.B.3. Vascular Aquatic plants

The artificial radioactivity concentrations in vascular aquatic plants from the upper Meuse river are given in table 13. ^{57}Co, ^{58}Co ^{60}Co, ^{65}Zn, ^{90}Sr, ^{110m}Ag and ^{134}Cs were looked up but were systematically below detection limits. One significant value has been obtained for ^{54}Mn in *Carex* leaves collected at Ham-sur-Meuse.
^{137}Cs was measurable in two samples at a concentration level of 1.2 ± 0.2 Bq kg^{-1} dw.

^{3}H (OBT) was detectable in 1 sample (Waulsort)

^{241}Am, ^{238}Pu and ^{239}Pu were not measured.

Species	Location	Dist. (km)	DM %	^{3}H Bq kg^{-1}	^{54}Mn Bq kg^{-1}	^{57}Co Bq kg^{-1}	^{58}Co Bq kg^{-1}	^{60}Co Bq kg^{-1}	^{65}Zn Bq kg^{-1}	^{110m}Ag Bq kg^{-1}	^{134}Cs Bq kg^{-1}	^{137}Cs Bq kg^{-1}
Carex	Ham s/Meuse	-3.0	38.2	< 9	0.58±0.28	< 1.1	< 3	< 1.5	< 5	< 1.5	< 1.5	1.3±0.5
Carex	Givet	9.5	39.4	< 8	< 1.3	< 0.9	< 3	< 1.3	< 4	<1.4	< 1.1	1.0±0.4
Carex	Waulsort	19.5	26.1	19±5	< 12	< 8	< 30	< 11	< 30	< 12	< 11	< 10
Carex	Yvoir	39.0	45.5	< 12	< 7	< 5	< 11	< 7	< 17	< 7	< 6	< 7
Carex	Hun	39.5	34.0	< 11	< 11	< 7	< 17	< 10	< 30	< 10	< 5	< 10

Table 13: Activity concentration of artificial radionuclides in vascular aquatic plants from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

4.B.4. Molluscs

In mollusc samples collected from the upper Meuse river, ^{57}Co, ^{58}Co ^{60}Co, ^{65}Zn, ^{90}Sr, ^{110m}Ag and ^{134}Cs were systematically below their detection limits (Table 14).

species	organ	Location	Dist. (km)	DM %	^{3}H Bq kg^{-1}	^{54}Mn Bq kg^{-1}	^{57}Co Bq kg^{-1}	^{58}Co Bq kg^{-1}	^{60}Co Bq kg^{-1}	^{65}Zn Bq kg^{-1}	^{110m}Ag Bq kg^{-1}	^{134}Cs Bq kg^{-1}	^{137}Cs Bq kg^{-1}
Anodonta	flesh	Heer	12.0	10.4	< 18	< 3	< 1.6	< 3	< 3	< 6	< 3	< 3	< 3
	flesh	Hastière	15.0	9.7	< 16	< 3	< 1.6	< 4	< 3	< 7	< 3	< 3	< 3
	flesh	Waulsort	19.5	10.9	< 28	< 2	< 1.2	< 3	< 3	< 6	< 3	< 1.9	< 3
	flesh	Dinant	31.0	11.4	12±2	< 3	< 1.8	< 4	< 3	< 7	< 3	< 4	< 3
	flesh	Hun	39.5	9.3	20±3	< 3	< 1.8	< 4	< 3	< 8	< 3	< 4	< 3
Dreissena	whole	Ham s/Meuse	-3.0	45.5	< 3	< 1.8	< 1.3	< 3	< 1.6	< 5	< 1.8	< 1.8	< 1.9
	whole	Heer	12.0	45.6	< 3	< 3	< 1.5	< 4	< 3	< 6	< 3	< 3	< 3
	whole	Waulsort	19.5	46.9	< 2.6	< 1.7	< 1.2	< 3	< 1.6	< 5	< 1.8	< 1.9	< 1.7
	whole	Dinant	31.0	47.3	< 2.8	< 1.7	< 1.1	< 3	< 1.6	< 4	< 1.7	< 1.8	< 1.6
	whole	Hun	39.5	48.4	< 2.2	< 1.7	< 1	< 3	< 1.8	< 4	< 1.6	< 1.5	< 1.7

Table 14: Activity concentration of artificial radionuclides in molluscs from the Meuse river (in Bq kg^{-1} dw ± 1 counting σ).

Two samples of *Anodonta* exhibited significant OBT content in their flesh in the order of 15 Bq kg^{-1} dw.

^{137}Cs could not be detected in contrast with some measurable samples in 1998.

4.B.5. Fish

Only ^{137}Cs has been detected in flesh from two fish samples (table 15): the maximum value amounted to 1.5 Bq kg^{-1} dw.

^{90}Sr was detectable in one sample (Heer: 5.3 Bq kg^{-1} dw) and ^{3}H was above detection limit in Dinant (13 Bq kg^{-1} dw).

species	organ	Location	Dist. (km)	DM %	^{3}H Bq kg^{-1}	^{54}Mn Bq kg^{-1}	^{57}Co Bq kg^{-1}	^{58}Co Bq kg^{-1}	^{60}Co Bq kg^{-1}	^{65}Zn Bq kg^{-1}	^{110m}Ag Bq kg^{-1}	^{134}Cs Bq kg^{-1}	^{137}Cs Bq kg^{-1}	^{90}Sr Bq kg^{-1}
Alburnus Perca Leuciscus	flesh	Heer	12.0	27.1	< 9	< 1.9	< 1.3	< 9	< 1.6	< 6	< 2	< 1.5	< 1.3	5.3±0.6
Alburnus Perca Leuciscus Rutilus	flesh	Hastière	15.0	25.3	< 17	< 4	< 3	< 17	< 3	< 11	< 5	< 3	< 3	< 4
Alburnus Perca Rutilus	flesh	Waulsort	19.5	25.4	< 13	< 3	< 1.9	< 12	< 3	< 8	< 4	< 3	1.5±0.5	< 4
Alburnus Perca Leuciscus Rutilus	flesh	Dinant	31.0	26.5	13±2	< 3	< 1.4	< 10	< 1.8	< 7	< 3	< 1.7	1.0±0.4	< 4
Perca Leuciscus	flesh	Hun	39.5	25.9	< 16	< 5	< 3	< 19	< 4	< 12	< 5	< 4	< 3	< 4

Table 15: Activity concentration of artificial radionuclides in fish from the Meuse river (in Bq kg^{-1} fw ± 1 counting σ).

5. DISCUSSION

Considering the potential releases from a pressurized water reactor (PWR) and the experience gathered during more than 30 years of radiological survey in the upper Meuse basin, a certain number of artificial radionuclides are systematically looked for in the different components of the river ecosystem in order to evaluate the impact on the population of the nuclear complex of Chooz.

In sediments, which accumulate radionuclides from the water, ^{137}Cs was found in every sample. The average contamination level downstream from the Belgian border is around 8.5 ± 2.3 Bq kg^{-1} dw, about a factor 3 less than in 1998. ^{134}Cs concentrations also decreased since 1992: in 1995 and 1998 not all values were above detection limits, and it is presently not detectable anymore.

The sample taken upstream from the NPP (at Ham-sur-Meuse) amounted to 1.8 ± 0.2 Bq kg^{-1} dw. The presence of higher ^{137}Cs concentration in bottom sediments downstream NPP can not just be due to erosion of contaminated soil particles from the slopes of the river and tributaries basin and to a past contamination from the global fallout and the Chernobyl deposit. At least, part of the ^{137}Cs was released from the NPP.

The ^{60}Co detected in all sediment samples in 1998 are now below detection limit, as expected from the ageing of the metallic parts of the reactor.

^{40}K activity amounts to about 410 Bq kg^{-1} dw and U-isotopes, all together, contribute to some 40 Bq kg^{-1} dw. ^{228}Th and ^{232}Th account each for about 40 Bq kg^{-1} dw. ^{226}Ra represents an activity concentration of 30 Bq kg^{-1} dw.

In aquatic plants, only few ^{137}Cs measurements were significant, but none of them exceeded 4 Bq kg^{-1} dw, which is much less than the ^{40}K content of about 500 Bq kg^{-1} dw.

In molluscs, only OBT was detected in two samples.

In fish, ^{137}Cs, ^{3}H and ^{90}Sr were the only artificial radionuclides exhibiting measurable concentrations. The maximum value was 13 and 5.3 Bq kg^{-1} dw for ^{3}H and ^{90}Sr respectively, which is however very low as compared to a natural ^{40}K content of 400 ± 10 Bq kg^{-1} dw.

6. CONCLUSIONS

The artificial radioactivity content of most plant and animal samples collected during the 2004 campaign in the upper Meuse lies below detection limits. The statistically significant values of ^{60}Co and ^{110m}Ag measured in 1998 have decreased because of the progressive stabilisation of the corrosion phenomena typical after the starting of a new installation. They are low and are largely exceeded by the natural radioactivity. Therefore, the aquatic releases from the Chooz nuclear site have only a marginal radiological impact on the population.

The sampling and measurement of bottom sediments and bio indicators is essential in the context of environmental survey as it allows revealing the presence of radionuclides of which the activity concentration in the water is too low to be detected. But, even for these samples, too many results are not statistically significant and can not be used to derive site specific transfer parameters. However, further decreasing the limits of detection seems very hard in many cases due to the limited availability of species and the limitations of the detection methods used.

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