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ORGANIC BOUND TRITIUM PUBLIC DOSES DUE TO TRITIUM EMISSIONS FROM CERNAVODĂ NUCLEAR POWER PLANT: A CASE STUDY

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Abstract. In a CANDU type reactor, such those of Ceranvoda NPP (Romania), tritium is generated by thermal-neutron-capture reactions: ^{2}H (n, γ) ^{3}H mainly in moderator and heat transfer system. Further, during maintenance and normal operation, a very small amounts of tritiated heavy water (DTO) may escape and diffuse in the environment. Absorbed and metabolised by plants, DTO contribute to the formation of the Organic Bound Tritium (OBT) which finally enters food chain reaching human body. Our results, based on the experimental data regarding DTO environmental content in the vicinity of Cernavodă NPP, OBT/DTO ratio as well as food consumption have showed that for routine releases of DTO into air and water, the OBT contributes with less then 25% to the total public tritium dose, which, at its turn was, at Cernavodă city, less than 0.3 μSv y^{-1}.

Key words: Organic bound tritium, Public doses, Nuclear power plant, Radioactive pollution, CANDU.

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1. INTRODUCTION

The release of any potential radioactive pollutant into environment during routine operation of a Nuclear Power Plant (NPP) are the subject of an appropriate controls and assessments. Radiation protection of the public is based on the principles recommended by the International Commission for Radiation Protection (ICRP) [2], according to whom the protection is mainly achieved by control of the exposure sources. Source monitoring provide a means of assessing the radiation exposure of population groups. The assessed doses are used to demonstrate the compliance with authorized dose limits, *i.e.* 0.1 mSv y^{-1} in our case, but they could also be used for optimization purposes too.

The CANDU reactors are both moderated and cooled by heavy water D_2O, so that a great amount of tritium by neutron reactions with deuterium, boron or lithium as well as by ternary fission is generated. Activation of deuterium is by far the most important mechanism and is responsible for the production of about 89 TBq of tritium MWe^{-1} y^{-1} with respect to the only 0.7 MWe^{-1} y^{-1} by ternary fission [1]. Most of the tritium present in CANDU reactors is in the form of tritiated heavy water (DTO). Outside reactor, tritium could exists in one of three basic chemical forms: i. - elemental form (HT - gaseous), ii. - oxide form or free-water tritium (DTO - vapour or liquid), iii. - organically bound tritium (OBT) where an ^3H atom bounds non-exchangeably to a carbon atom in an organic molecule. The majority of tritium released from nuclear facilities is in the form of either elemental gas HT or tritiated water DTO, while the OBT represents only few percent of the total release tritium and occurs only when lubricating oils or solvents exchange tritium [3–5].

The HT emissions in the atmosphere leads to its the deposit and diffusion in porous soil where is oxidized by microorganisms to DTO. Here, some of DTO is taken by plants through the root system, and some is released into the atmosphere. DTO released into the atmosphere is taken up by animals and humans by inhalation and ingestion. Tritium that enters plants as DTO engages in various reactions in which water is a substrate, being partially converted into OBT. In the human body, tritium can enter in any of its chemical or physical forms, *i.e.* i. - HT by inhalation; ii. - DTO by inhalation, ingestion, or absorbed through the skin; and iii. - OBT by ingested foods. Here, in organism OBT could exists in two forms: exchangeable OBT and non-exchangeable OBT. Non-exchangeable OBT appears when tritium replaces hydrogen in carbon-hydrogen bond making it difficult to be replaced back by normal hydrogen while exchangeable OBT consists of tritium atoms (ions) bound to oxygen, nitrogen, phosphorus or sulphur atoms (ions), easily replaceable by hydrogen ions contained by water pool [6].

As OBT behaviour in the environment is relatively complicated and has a higher uncertainty than DTO, its accurate determination is useful for normal operations while its prediction should be very useful for accidental situations. The DTO and OBT dosimetry is related to the biological half- life of the tritium in the body. DTO behaves similar to water having a short half-life of about 10 d for adults [7]. On contrary, the non-exchangeable OBT has a longer biological half-life, attributed to the time required to break the molecular bonds to tritium, thereby contributing to a higher dose after ingestion [8]. For a better characterization of the effect of DTO and OBT on humans, the ICRP established for adults a dose conversion factor for ingestion DCF_{ig} of 1.8 10^{-11} Sv Bq^{-1} for DTO and of 4.2 10^{-11} Sv Bq^{-1} for non-exchangeable OBT [2](ICRP, 1996). The dose conversion factor for exchangeable OBT has not been universally established. Exchangeable OBT can be readily exchanged with hydrogen in the tissue free water (TFW) and should have the same

kinetics as DTO [9]. Regarding this, Ware and Allot [10] suggested that the exchangeable OBT dose conversion factor is likely to be between that of DTO and non-exchangeable OBT.

Romania has a fully functional CANDU type NPP located near Cernavodă city, in the South-Eastern Dobrudja, at the confluence of the Danube River with the Danube-Black Sea Canal. Provided with two 750 MW power units, the Cernavodă NPPs area was monitored since 1997, and thus allowing to collect a considerable amount of radioactivity concentration in environmental media data. The measurement of OBT concentrations in environmental samples is more difficult and complicated than the measurement of DTO, it is time-consuming, and requires specialized equipment and skills. The values obtained at environmental levels are subject to considerable uncertainty. Usually the sample dry matter remaining after freeze drying is washed repeatedly with tritium-free water to remove exchangeable OBT, and then freeze dried again. The dried material is combusted in a combustion bomb or furnace. The combustion water is distilled to neutralize and purify it and then counted in a LSC. Similarly, the ability to model OBT formation in plants, transfer to animals and humans, and dose per unit intake is limited by a lack of knowledge of the processes involved. Taking into account these considerations, and the lack of direct measurement of OBT in environmental samples at Cernavodă site, we tried to estimate the public dose due OBT using data recommended by different studies.

Accordingly, in this paper we present the results of our estimation of the public dose due to OBT by taking into account the original data regarding the DTO atmospheric content at Cernavodă City between 1997 and 2010, the annual food consumption in the most unfavourable hypothesis that all food was produced in the vicinity of Cernavodă city, as well as the literature data regarding both OBT to DTO ratio in foods and the OBT Dose Conversion Factors (*DCF*).

2. TRITIUM DOSIMETRY

Doses rate *D* (in Svy^{-1}) due to tritium absorption are usually calculated in the same way as doses due to other radionuclides, *i.e.* by multiplying the tritium intake rate by a dose conversion factor, according to the relation:

$$D = Q_T \cdot DCF, \tag{1}$$

where Q_T is the tritium intake rate (in $\mathrm{Bq\ y}^{-1}$) and *DCF* is the dose conversion factor (in SvBq^{-1}).

Due to different pathway, DTO should account for all exposure pathways *e.g.* inhalation, skin absorption, and food and water ingestion while OBT is only due to food ingestion. At the same time, DTO and OBT having different residence times in the body and consequently different dose conversion factor, needs to be treated

separately.

In the case of DTO, tritium intake rate Q_T is calculated by using its concentrations in air, drinking water and foodstuffs as well as the corresponding intake rates. In the case of OBT, the calculation involves only the activity concentrations and intake rates of foodstuffs.

The *DCF*s are generally calculated by taking into account a multitude of factors such as: i.-the distribution of tritium in the body following intake, ii. - conversion between DTO and OBT, iii. - energy deposition due to decay on the target cells, iv. - the radiosensitivity of the cells, v. - the rate at which tritium is lost from the body, vi. - the Relative Biological Effectiveness (*RBE*) of DTO and OBT. It is worth mentioning that the most hazardous forms of OBT in diet are the tritiated proteins [11], not produced in the human body, but taken up directly through the ingestion of animal products.

At present, there are different models to calculate tritium DCF in humans. The most often used is the ICRP model [12, 13], which assumes that the tritium is completely absorbed and uniformly distributed throughout all body tissues so that 97% of the DTO activity embedded in the body remain as DTO with a retention half-life in adults of 10 days while 3% is converted to OBT, which is excreted with a half-life of 40 days. The same model assumes that 50% of the OBT is converted to DTO. Within this model, both DTO and OBT *DCF* assumes a *RBE* equal to unity.

The advanced tritium models developed recently by [11, 14, 15] makes realistic assumptions regarding physiological parameters like the size of body, soft tissue bone pools, oxidation rates, proportion of carbohydrates, the amount of fat and protein in the diet. The tritium *RBE* ranges from 1.0 to 3.8 for DTO and from 1.0 to 11.6 for OBT. If the *RBE* is raised, the tritium dose will rise proportionately. According to the these models, there are distinct differences in the *DCF*s whose values depend on the age and gender: the *DCF* for females is about 20% higher than for males for DTO and about 50% higher for OBT [11, 15, 16]. Most of these models presume that the *DCF* for OBT is about 2.3 times larger than the *DCF* for DTO. At the same time, the OBT causes more harm than DTO because of its longer biological half-life and its potential to associate with radiosensitive cells like DNA molecules [17].

For routine releases of DTO in air and water, OBT contributes with about 20% or less to the total tritium dose to members of the public, to aquatic plants and animals, and to non-cereal terrestrial plants so that, the food ingestion remains the main vector of OBT in humans as in other animals. For this reason, an accurate estimation of OBT content in different kind of food as well as their daily consumption in humans are the most straightforward method to estimate, with sufficient accuracy, the OBT dose to population.

As the total amount of OBT in food is directly dependent on the environmental content of DTO, it is obvious that higher the DTO atmospheric content, greater the

Table 1.

Average *per capita* food consumption rate for Cernavodă site (in kg y^{-1}), DTO and OBT contents: c_{DTO} and respectively c_{OBT} (in 10^{-8} lg^{-1}) as well as the OBT to DTO ratio.

Food	Consumption rate	c_{DTO}	c_{OBT}	OBT/DTO ratio
Fruits, vegetables, cereals	251.8	93.2	3.2	1.0 to 1.8
Beef, meat	13.4	57.3	33.7	0.6 to 1.4
Pork meat	13.4	30.2	61.1	0.9
Milk	193.5	89.2	5.8	0.7 to 4.2
Poultry	13.4	65.5	25.2	0.6 to 1.7
Fish	7.3	75.7	13.4	2.4

OBT in food, and consequently, higher the intake dose. For this reason, the distance between the NPP stacks and food producing units plays a significant role.

3. MATERIALS AND METHODS

3.1. POPULATION

In this study, we have considered the average population of the Cernavodă city, located at about 2 km distance from the Cernavodă NPP. With a population of about 18,500 inhabitants and a surface of about 47 km^2, the Cernavodă was continuously monitored since 1997, when the first unit of the Cernavodă NPP was commissioned and staring to produce electricity. At the same time, we have considered the average food consumption per capita in the analysed period, *i.e.* 1997 to 2010 year.

As in the case of the Cernavodă inhabitants the annual consumption of different kind of foods are well known, (see Table 1), for our estimation of the OBT contribution to the total annual dose we have used the existing original data regarding the DTO activity concentration in air as well as in the main categories of food (Table 2) together with the literature data regarding OBT to DTO ratio [19] as well as the DTO DCFs [13, 15, 17, 20].

3.2. ANNUAL DOSE FOR INHALATION AND INGESTION

To calculate the annual dose through inhalation D_{ih}, we had used the following relation:

$$D_{ih} = c_{air} \cdot I \cdot OF \cdot DCF_{ih}, \tag{2}$$

where: c_{air} represents the DTO activity concentration in the air (in Bq m^{-3}); *I* stands for the inhalation rate (in m^3y^{-1}); *OF* represents the occupancy factor *i.e.* fraction of time an individual is exposed to the radioactive material in the air; DCF_{ih} is the dose conversion factor for inhalation (in Sv Bq^{-1}).

Table 2.

Numerical values of DCF_{ih} and DCF_{ig} (in 10^{-11} x Sv Bq^{-1}) for DTO used in the present work. (F, M and S denote fast, moderate and slow lung absorption respectively).

Age/gender	DCF_{ih}					DCF_{ig}		
	[13]	[15]	[20]			[13]	[15]	[20]
			F	M	S			
1 year	4.80	4.98	2.60	34.0	120	4.80	4.98	6.40
10 years	2.30	3.04	0.82	8.20	38.0	2.30	3.04	2.30
Adult/male	1.80	1.96	0.62	5.50	26.0	1.96	1.96	1.80
Adult/female	1.80	2.36	nd	nd	nd	2.36	2.36	nd

For our calculations, we have considered the numerical values of the annual inhalation rates *I* as mentioned in [18]: i. - 1400 m^3y^{-1} for infants up to one year old; ii. - 5500 m^3y^{-1} for children between one and 10 years old and iii. - 8400 m^3y^{-1} for adults. All inhalation rates were considered appropriate for average individuals and critical groups.

In the case of ingestion pathway, the annual dose through ingestion D_{ig} is described by the following relation:

$$D_{ig} = c_{food} \cdot I_f \cdot DCF_{ig}, \tag{3}$$

where: c_{food} represents the DTO activity concentration in food (in Bq kg^{-1}); I_f stands for the consumption rate (in kgy^{-1}); DCF_{ig} is the dose conversion factor for ingestion (in Sv Bq^{-1}).

To finish the calculations of OBT annual dose for average Cernavodă inhabitants, we have used the food consumption rates reproduced in Table 1, as well as the numerical values of DCF_{ih} and DCF_{ig} regarding DTO and respectively DCF_{ig} corresponding to OBT (Table 2).

4. RESULTS AND DISCUSSION

The experimental data regarding the activity concentration of DTO at Cernavodă city as reproduced in Table 3 and illustrated in Figure 1 showed that for the investigated period, *i.e.* 1997 to 2010, DTO activity concentration is distributed by following a lognormal distribution and its maximum value never overpass 2 Bq/m^3, as reported in [21]. As mentioned above, there is not an unanimous point of view regarding the numerical values of both dose conversion factor for ingestion and inhalation as well as for the OBT to DTO conversion ratio, but regardless this fact, it is still possible, starting from the experimental data concerning DTO activity concentration, average food consumption, as well as the different variants of dose conversion factors to estimate the OBT public dose for the Cernavodă city inhabitants.

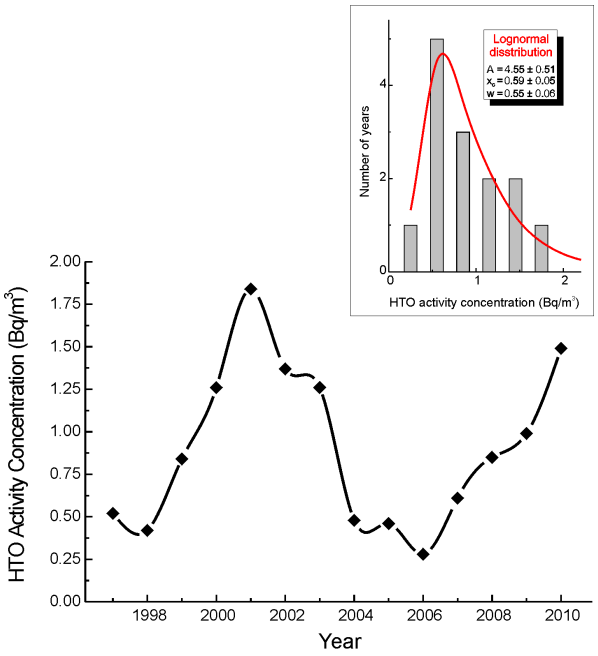

Fig. 1 – The annual average value of DTO activity concentration at the Cernavodă City between 1997 and 2010. The inset illustrate the corresponding distribution function (gray columns) as well as the lognormal best fit (continuous red line).

For this reason, we have computed the OBT public dose by using different models of OBT ingestion and transfer to humans such as those recommended by ICRP [2] or presented by Galeriu et al in Ref. [17]. The final results of our estimation are detailed presented in Table 2 and illustrated in Figure 2. As it results from these data, there are significant differences between the numerical values of OBT annual dose predicted by the mentioned models, differences which are in some cases about one order of magnitude, but, regardless this fact, the annual OBT dose never overpass 0.1μSv y^{-1}), which account for less than 25% of the annual dose due to total tritium intake within Cernavodă city area [21].

At the same, judging on the graphs reproduced in Fig. 1 and 2, apparently between average annual atmospheric release of DTO and OBT dose there is a weak correlation, which could be explained by the existing fluctuation in the annual food consumption.

Regarding our results it must be pointed out that the ability to model OBT formation in plants, transfer to animals and humans, and dose per unit intake is limited by insufficient knowledge of the processes involved. Moreover, the intake and the dose conversion coefficients depends on age and personal diet. In practice DTO concentration in many crops depends on recent DTO concentration on air and soil while

Table 3.

The OBT annual public dose (extent and average values) corresponding to different DCF_{ig} as defined by [2] - columns (2,3) and by [17] for male (columns 4,5) and female (columns 6,7). All public doses are based on the data reproduced in Tables 1 and 2

	Annual OBT public dose (μSv y^{-1})					
year	extent	average	extent	average	extent	average
1997	0.010 - 0.030	0.020	0.015 - 0.045	0.030	0.024 - 0.074	0.049
1998	0.090 - 0.028	0.018	0.013 - 0.042	0.027	0.021 - 0.068	0.045
1999	0.009 - 0.029	0.019	0.014 - 0.043	0.029	0.022 - 0.071	0.046
2000	0.090 - 0.028	0.018	0.013 - 0.042	0.028	0.022 - 0.069	0.045
2001	0.080 - 0.024	0.016	0.012 - 0.037	0.024	0.019 - 0.059	0.039
2002	0.070 - 0.021	0.014	0.010 - 0.032	0.021	0.016 - 0.052	0.034
2003	0.050 - 0.017	0.011	0.080 - 0.025	0.016	0.012 - 0.041	0.026
2004	0.012 - 0.031	0.021	0.018 - 0.046	0.032	0.029 - 0.075	0.052
2005	0.007 - 0.021	0.014	0.011 - 0.031	0.021	0.017 - 0.051	0.034
2006	0.011 - 0.034	0.024	0.017 - 0.055	0.036	0.027 - 0.090	0.058
2007	0.011 - 0.038	0.024	0.016 - 0.057	0.037	0.026 - 0.093	0.059
2008	0.009 - 0.031	0.020	0.014 - 0.046	0.030	0.022 - 0.075	0.049
2009	0.013 - 0.035	0.024	0.019 - 0.053	0.036	0.032 - 0.086	0.059
2010	0.012 - 0.035	0.023	0.019 - 0.053	0.036	0.032 - 0.086	0.059

OBT concentration reflects the history of several weeks. As the DTO sampling in the Cernavodă area is limited to few samples per month, the average values could be affected by sampling number and sampling time, but, regardless these considerations, the annual OBT dose represents a small percent of the total tritium exposure, which, at its turn, is significantly lower then the value of 1 mSv/y in the case of nonocupational exposure to ionizing radiation [22, 23].

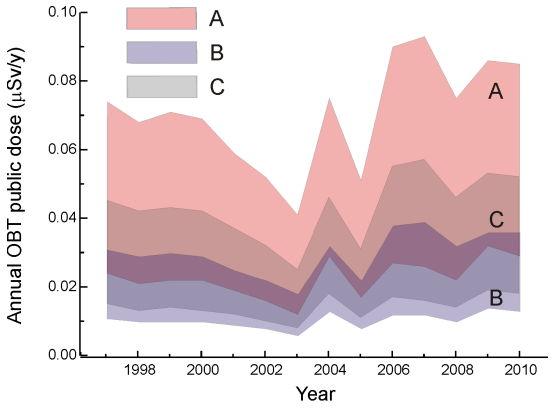

Fig. 2 – The annual OBT public dose at the Cernavodă City between 1997 and 2010, as computed by using different DCF_{ig} as defined in Ref. [17] for male (A) and female (C) as well as by Ref. [2] (B).

5. CONCLUDING REMARKS

A 14 years monitoring of the DTO emissions from the Cernavodă NPP allowed us to estimate, by using different models regarding the ingestion of the OBT, its contribution to the annual dose to the population of Cernavodă City. In this way and depending on the chosen model, the annual supplementary dose varied between 0.009 and 0.093 μSv y^{-1}), which represents maximum 25% of the annual does due to DTO intake, which, at its turn is significantly lower then the value of 1 mSv/y admitted by the present time Romanian Fundamentals Norms on Radiological Safety.

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