Romanian Reports in Physics, Vol. 62, No. 4, P. 827–837, 2010

ATMOSPHERIC TRITIUM DYNAMICS AROUND CERNAVODA NUCLEAR POWER PLANT

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(Received November 15, 2009)

Abstract. Data concerning the distribution of ^3H in the atmosphere in the vicinity of Cernavoda Nuclear Power Plant (NPP) during 1997–2007 monitoring are presented and discussed. The annual average concentration of the atmospheric tritiated water varied between 1.3 and 13 Bq/m^3 in the immediate vicinity of NPP, exponentially decreasing with distance from the Cernavoda NPP and falling to the current background level at 30 km off-site. Within Cernavoda town, the total supplementary dose due to tritium intake fluctuated between 0.27 and 0.69 μSv/y, three orders of magnitude lower than the maximum permitted dose for nonocupational exposure. The role played by atmospheric precipitation in ^3H spreading was proved by the existing weak positive correlation between tritium fall-out and atmospheric absolute humidity.

Key words: tritium, nuclear power plant, Cernavoda, atmosphere, vegetation, soil, interstitial water.

1. INTRODUCTION

The CANDU technology Cernavoda Nuclear Power Plant (NPP) is dedicated to produce electrical as well as thermal energy in a safe and efficient manner for at least 30 years. CANDU reactors are both moderated and cooled by heavy water. During routine operation of such type of reactor, various gaseous, liquid, and solid radioactive wastes are generated, which, in spite of existing protections systems that minimize their output, are continuously discharged in small quantities and at very low concentrations as gaseous and liquid effluents in environment.

Among them, tritium represents the main component. Tritium is generated in CANDU reactors by neutron reactions with deuterium, boron, lithium and by ternary fission, but activation of deuterium is by far the most important mechanism. In this way, about 89 TBq of tritium is produced annually for each MW of electricity compared to only 0.7 TBq generated by ternary fission per MW of electricity and per year. Most of the tritium present in CANDU reactors is in the form of tritiated heavy water (DTO) [1].

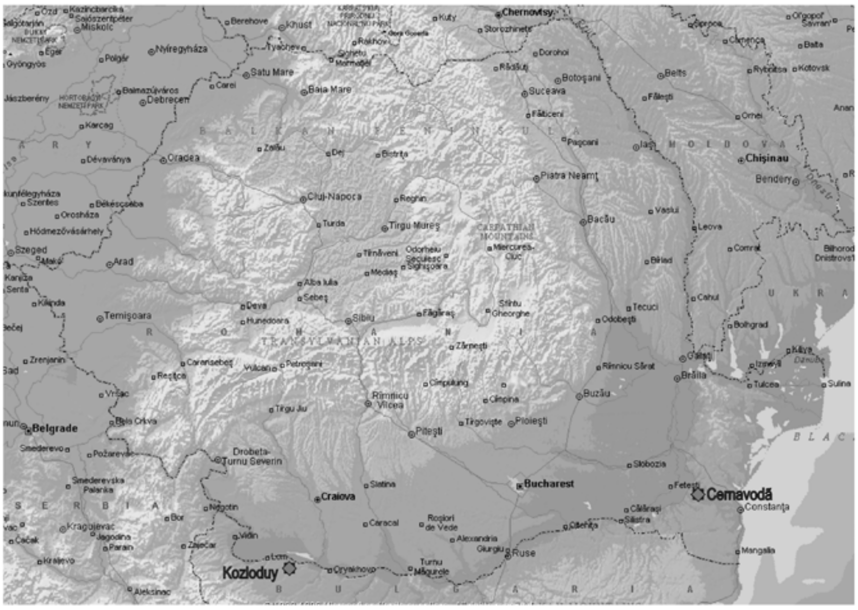

Fig. 1 – Map showing the location of Romanian Cernavoda as well as Bulgarian Kozlodui NPP.

Tritium (half-life of 12.33 y) undergoes β^- decay to ^{3}He by emitting a low-energy β^- particle, with an average energy of 5.7 keV (maximum 18.6 keV) and a maximum range of 6 mm in dry air and about 8 μm in tissue. For this reason, outside body, tritium contribution to radiological hazard is negligible, as its β^- particle cannot penetrate the outer layer of the skin while, once entered in the body either by inhalation or by absorption through the skin, the energy of β^- particle deposited in body tissue has very harmful consequence. As within a CANDU reactor, tritium is usually bound in a molecule of heavy water, when escapes, it creates in the air a source of DTO vapours. Inhaled or absorbed through exposed skin, DTO produces an internal, occupational radiation hazard in the station and through normal ventilation flows contribute to the station's tritium emissions [2-6].

Twelve years of experience in operating Cernavoda CANDU NPP have shown that ^{3}H represents more than 80% of the total released radioactivity.

Monitoring of the Cernavoda site was performed between 1984 and 1996 within the Preoperational Environmental Radiation Monitoring Program whose main aim was to define background radiation by analyze environmental. It is worth mentioning that within this program the fallout from Chernobyl accident was detected in April of 1986. Finally, the Environmental Radiation Routine Monitoring Program for Cernavoda NPP started in March 1996.

Further, in this paper we will present and characterize the distribution and trends of ^{3}H atmospheric concentrations around the Cernavoda NPP.

2. MATERIALS AND METHODS

2.1. ATMOSPHERIC DISCHARGE OF DTO FROM CERNAVODA NPP UNIT 1

The Cernavoda NPP is situated in the Dobruja area, in the vicinity of the town of Cernavoda, counting for more than 20,000 inhabitants (Fig. 1). The natural cooling system is provided by the right arm of the Danube River and by the Danube-Black Sea Canal. The area is primary rural terrain with a mixture of residential and agricultural areas.

Table 1

The numerical values of the annual discharges [TBq/year] of DTO in gaseous effluents from Cernavoda NPP Unit 1

year	DTO release	year	DTO release
1996	1.4	2002	286.0
1997	25.5	2003	171.0
1998	50.8	2004	198.0
1999	85.3	2005	246.0
2000	208.0	2006	350.0
2001	180.0	2007	230.0

Tritium has been continuously discharged mainly in tritiated water form into the atmosphere from the Cernavoda NPP, *via* a 30 m high stack. The discharge rate of DTO has been determined as the weekly-averaged rate by the ^{3}H activity measurement of water contained in the airborne effluent at the stack from Cernavoda NPP Unit 1, corresponding data being illustrated in Table 1 as annual average values and in Fig. 2 as both weekly and annual values.

2.2. SAMPLING

Tritium concentration in air was measured at environmental monitoring sites placed around the plant, three within power plant and eight at distances between 1 and 30 km (Table 2). Airborne water vapors were collected continuously by drawing air through molecular sieve (13X type) while sampling were performed with an integration period of one month, during which 4–6 m^3 of air are passed through the collector. From this, the tritium content was determined by liquid scintillation counting. The air sampling equipments used between 1996–2001, were manufactured by Nuclear Research Reactor Institute from Pitesti. In 2001 in order to decrease sampling errors, air sampling equipments were changed with CAS - LV 118 ECNV- model (F&J Speciality Products Inc.) equipped with a back-up power supply a and digital flow integrator.

2.3. TRITIUM ACTIVITY MEASUREMENT

For the determination of ^{3}H concentrations in the atmosphere, the absorbed water was removed from molecular sieves by heating it at 350 °C. 8 ml of extracted and purified water was homogenously mixed with 12 ml of liquid scintillator Ultima Gold LLT in a 20 ml vial followed by ^{3}H activity measured with a Beckmann LS1801 liquid scintillation spectrometer (between 1996 and 2000) and by a more sensitive Quantulus 1220 (from 2001 till present). Always the counting time was 400 min. Although the quenching level of all samples was quite constant, the quenching correction in the activity determination was made using a quenching curve derived beforehand from the measurement of a series of quenched standards.

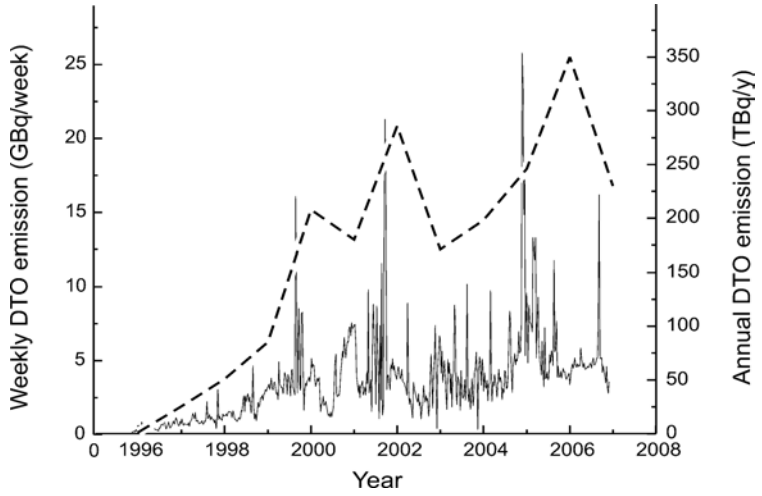

Fig. 2 – Weekly (continuous line) and yearly (dotted line) DTO emission as measured in the vicinity of Cernavoda NPP stack.

In these conditions, typical background count rate and counting efficiency for these measurements were about 5 cpm and 18–20%, respectively. By considering the minimum detectable activity of ^{3}H as equal to three times the standard deviation of the background counting rate, in the present system minimum activity was estimated to roughly 2.5 Bq l^{-1} (10 Bq/l for the previous Beckmann LS1801 system) [7–10].

It is worth mentioning that the ^{3}H concentrations below the minimum detectable concentration in the monthly monitoring were considered as having the minimum detectable concentrations. Since the number of data below the detectable concentration was very small, we considered that these data did not affected the estimation of the annual concentrations.

3. RESULTS AND DISCUSSION

3.1. ATMOSPHERIC DTO CONCENTRATIONS

The results of our multi-annual monitoring are presented in Table 1 and illustrated in Fig. 2. By analyzing these data it follows that the DTO emission, excepting some local maxima corresponding to the first years of exploitation, slowly increased between 1996 and 2007 reaching its present value.

To estimate at which extent released DTO was found in the atmosphere in the vicinity of NPP, in Table 2 we have reproduced the atmospheric ^3H concentrations, as annual averages based on weekly monitoring data corresponding to all stations (see also Fig. 3) The same data were used to calculate the multi-annual atmospheric DTO release at the place of monitoring stations, illustrated in Fig. 4 as a function of distance between NPP stack and each station.

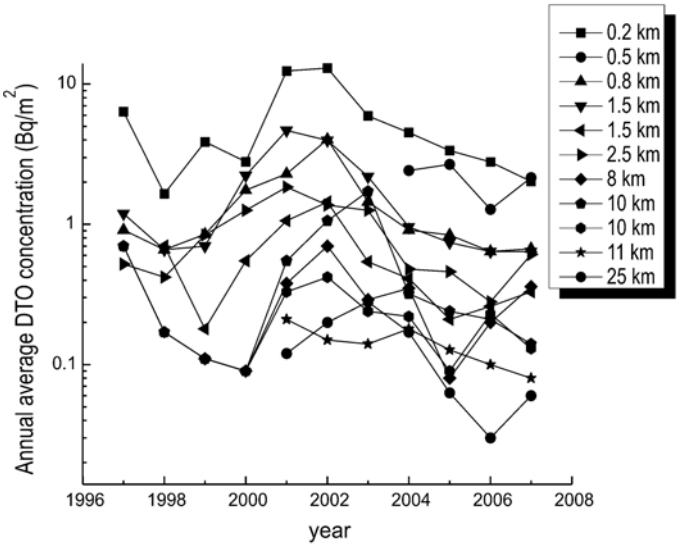

Fig. 3 – Time evolution of the average annual ^3H concentration [Bq/m^2] for all monitoring stations (—►— Cernavoda town).

As atmospheric humidity plays an important role in DTO deposition, in Fig. 5 we have illustrated the time evolution of absolute humidity beginning with 1996 and ending with 2007 year. Moreover, for the same reason we have calculated, for each station the correlation coefficients between ^3H fallout and absolute humidity (Table 3).

From these data it can be remarked that the multi-annual average DTO concentrations fall rapidly with the distance with respect to NPP stack, faster in the vicinity of central stack until Cernavoda town and slower in the case of the most remote monitoring station, *i.e.* ADB-01 located at about 30 km faraway, so that the annual values of ^{3}H concentrations are similar to those measured in the preoperational program 1994–1996 by IFIN-HH Bucharest (0.186 and 0.032 Bq/m^3). Better this dependency was described by a sum of two exponentials, characterised by half-attenuation distances of 0.17 ± 0.07 km nearby central stack and 3.3 ± 0.7 km after the town of Cernavoda (Fig. 4). This fact suggests that at this distance, the influence of the Cernavoda NPP Unit 1 tritium effluents is unimportant and consequently, ADB-01 data can be regarded as background ^{3}H level around the Cernavoda.

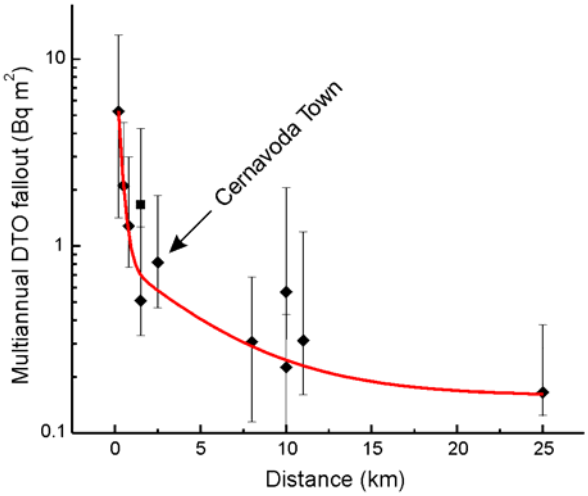

Fig. 4 – Multi-annual average DTO concentration as function of distance between NPP stack and monitoring station, better described by a decay-type exponential.

Table 2

Annual ^{3}H averages atmospheric concentrations around the Cernavoda NPP [Bg·m^{-3}]

Year	Location					
	Distance from the NPP [km]					
	ADI-12	ADI-13	ADI-11	ADI-05	ADI-06	ADI-08
	0.2 km	0.5 km	0.8 km	1.5 km	1.5 km	2.5 km
1997	6.36± 0.5	no data	0.91± 0.07	1.19 ± 0.29	< 0.62	0.52± 0.09
1998	1.65± 0.1	no data	0.66± 0.06	0.66± 0.06	0.70 ± 0.06	0.42± 0.07
1999	3.87± 0.1	no data	0.85± 0.07	0.70 ± 0.06	0.18 ± 0.04	0.84± 0.07

Table 2 (continued)

2000	2.79± 0.1	no data	1.75± 0.09	2.23 ± 0.13	0.55 ± 0.06	1.26± 0.08
2001	12.43± 0.1	no data	2.29± 0.03	4.68 ± 0.05	1.06 ± 0.01	1.84± 0.03
2002	13.00± 0.2	no data	4.03± 0.06	3.96 ± 0.05	1.44 ± 0.02	1.37± 0.02
2003	5.92± 0.07	no data	1.45± 0.02	2.19 ± 0.02	0.54 ± 0.008	1.26± 0.02
2004	4.52± 0.05	2.42± 0.03	0.91± 0.01	0.96 ± 0.01	0.41 ± 0.006	0.48± 0.07
2005	3.35± 0.05	2.68± 0.04	0.84± 0.01	0.74 ± 0.01	0.21 ± 0.004	0.46± 0.007
2006	2.78± 0.06	1.28± 0.02	0.64± 0.01	0.64 ± 0.01	0.26 ± 0.005	0.28± 0.005
2007	2.02± 0.03	2.15± 0.03	0.67± 0.01	0.64 ± 0.01	0.33 ± 0.01	0.61± 0.01

Year	Location Distance from the NPP [km]				
	ADI-09 8 km	ADI-02 10 km	ADI-04 10 km	ADI-10 11 km	ADB-01 25 km
1997	< 0.70	0.70 ± 0.44	0.70± 0.13	< 0.70	0.70 ± 0.12
1998	< 0.17	0.17 ± 0.03	0.17± 0.06	< 0.17	< 0.17
1999	0.11± 0.04	0.11 ± 0.04	0.11± 0.04	< 0.11	< 0.11
2000	0.09± 0.05	0.09 ± 0.04	0.09± 0.04	< 0.09	< 0.09
2001	0.38± 0.009	0.55 ± 0.01	0.33 ± 0.01	0.21± 0.01	0.12 ± 0.01
2002	0.70± 0.001	1.06 ± 0.02	0.42± 0.01	0.15± 0.003	0.20 ± 0.01
2003	0.29± 0.010	1.72 ± 0.03	0.24± 0.004	0.14± 0.003	0.28 ± 0.005
2004	0.35± 0.007	0.32 ± 0.005	0.22± 0.004	0.18± 0.003	0.17 ± 0.003
2005	0.08± 0.003	0.24 ± 0.008	0.09± 0.003	0.127± 0.003	0.063 ± 0.002
2006	0.20± 0.008	0.21 ± 0.003	0.23± 0.005	0.10± 0.002	0.03 ± 0.001
2007	0.36± 0.008	0.14 ± 0.003	0.13± 0.003	0.08± 0.002	0.06 ± 0.002

Another important monitoring station, ADI-08, located in the Cernavoda town, 2.5 km off NPP stack was used to calculate public supplementary doses due to Unit 1 DTO emissions. At this location, the annual ^{3}H concentrations are two to 10 time lower than those registered in the immediate vicinity of NPP, but still higher then above mentioned background (Table 2, Fig. 4).

With regard to this, it is worth mentioning that, according to our monitoring data concerning the annual intake of ^{3}He by ingestion and inhalation within Cernavoda town, the total supplementary dose due to tritium varied between 0.27 and 0.69 μSv/y [V. Simionov, unpublished results], a value significantly lower than the maximum admitted value of 1 mSv/y in the case of nonocupational exposure to ionizing radiation [11, 12].

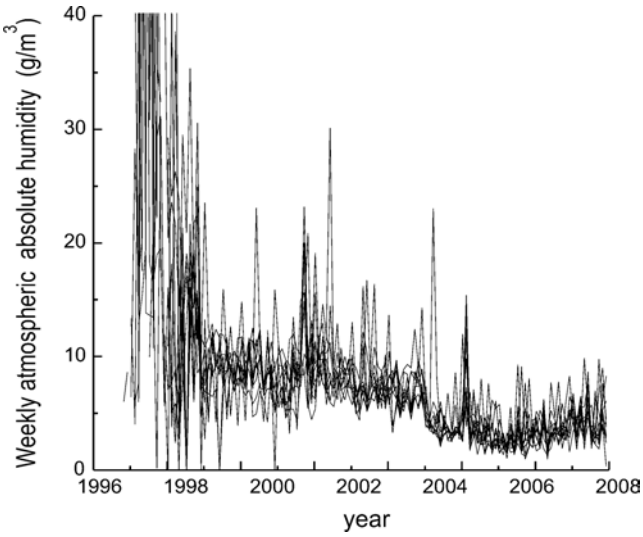

Fig. 5 – Weekly atmospheric absolute humidity corresponding to all observation points around Cernavoda NPP between 1996 and 2007.

Concerning both DTO released into atmosphere and ^{3}H fall-out recorded at all monitoring stations, we have noticed two contrary tendencies, while atmospheric release slowly increased between 1997 and 2007, the tritium fallout recorded by all stations showed a slightly decline for the same period of time. In our opinion, this peculiarity concords with the role played by atmospheric precipitations in transporting radioactive gases emitted by Cernavoda NPP (Fig. 5). This assumption is partially confirmed by the existing positive correlations between atmospheric humidity and DTO fall-out for some of the observing points (Table 3).

Table 3

The correlation coefficients between the long term DTO fall out and humidity as recorded by observations stations around Cernavoda NPP. Significant correlations at $p < 0.05$ are represented in italics

ADI-12	***0.19579***	**ADI-05**	0.03836	**ADI-09**	***0.38363***	**ADI-10**	***0.97865***
ADI-13	***0.29304***	**ADI-06**	***0.21591***	**ADI-02**	***0.29753***	**ADB-01**	***0.45228***
ADI-11	0.06051	**ADI-08**	***0.48105***	**ADI-04**	0.11022		

At the same time to verify at which extent the atmospheric dispersion around Cernavoda is more or less uniform, we have investigated the correlation existing between the long term DTO fall-outs at each observation points. The results reproduced in Table 4 as a matrix of correlation coefficients showed positive correlations at $p < 0.05$ (95% probability) only in few cases, this fact arguing for a more complex short-distance dispersion of tritium in the vicinity of NPP.

Table 4

Matrix of the correlation coefficients of the long term DTO fallout as recorded by observation stations around Cernavoda NPP. Significant correlations at $p < 0.05$ are represented in italics

	ADI-12	**ADI-13**	**ADI-11**	**ADI-05**	**ADI-06**	**ADI-08**	**ADI-09**	**ADI-02**	**ADI-04**	**ADI-10**
ADI-13	-0.0689									
ADI-11	***0.6254***	0.0558								
ADI-05	***0.4889***	-0.1181	***0.4684***							
ADI-06	***0.5220***	-0.1106	***0.4564***	***0.2966***						
ADI-08	***0.1840***	0.0827	***0.3521***	***0.2414***	***0.2838***					
ADI-09	***0.3718***	0.0490	***0.4199***	***0.3004***	***0.2517***	***0.1712***				
ADI-02	***0.2820***	-0.0441	***0.3437***	***0.1328***	***0.2064***	0.0731	***0.2323***			
ADI-04	***0.3029***	-0.0661	***0.2928***	***0.4078***	***0.3211***	0.1237	***0.3124***	***0.2190***		
ADI-10	-0.0040	-0.0427	-0.0258	-0.0191	0.0177	-0.0612	***0.2491***	0.0100	***0.2391***	
ADB-01	0.1199	-0.0467	***0.1962***	0.0308	0.1098	0.0595	***0.3229***	***0.2373***	***0.3988***	***0.6550***

Similar results we have obtained by analyzing the long term distribution of humidity for the same observation points, as results from a similar matrix of correlation coefficients (Table 5) which shows the same absence of significant positive correlations.

The long term data concerning the discharge rate of DTO from Cernavoda NPP and its average atmospheric concentration at each observing point we have calculated K_a, the long-term Atmospheric Dispersion Factors (ADFs):

$$K_a = \frac{C}{Q}, \tag{1}$$

where C is the annual average tritium-in air concentration [Bq/m^3] and Q is the average gaseous tritium release rate [Bq/s].

Table 5

Matrix of the correlation coefficients of the long term atmospheric humidity as recorded by observation stations around Cernavoda NPP

	ADI-12	**ADI-13**	**ADI-11**	**ADI-05**	**ADI-06**	**ADI-08**	**ADI-09**	**ADI-02**	**ADI-04**	**ADI-10**
ADI-13	***0.7110***									
ADI-11	***0.5604***	***0.5984***								
ADI-05	***0.7280***	***0.7826***	***0.6530***							
ADI-06	0.1663	0.0128	0.14836	0.1082						
ADI-08	***0.4919***	***0.5289***	***0.56703***	***0.6970***	0.1304					
ADI-09	0.1590	0.2306	0.17432	0.1742	-0.0942	0.1433				
ADI-02	***0.3979***	0.2689	0.27304	***0.3068***	0.1562	0.2066	0.0787			
ADI-04	***0.4014***	***0.5023***	***0.32808***	***0.6856***	-0.0478	***0.5335***	0.1737	0.2662		
ADI-10	0.2453	***0.3018***	0.19004	0.2795	0.2056	***0.3486***	-0.0205	***0.3669***	0.2209	
ADB-01	***0.4043***	***0.4440***	***0.34988***	***0.5247***	0.1511	***0.3860***	***0.5381***	***0.5275***	***0.4377***	0.1843

Table 6

Numerical values of the annual dispersion factor in the vicinity of Cernavoda NP

Location	Distance from the NPP [km]	Atmospheric Dispersion Factor x 10^{-6} s/m^3									
		1998	1999	2000	2001	2002	2003	2004	2005	2006	2007
ADI-12	0.2	1.03	1.31	0.39	2.21	1.43	0.65	0.72	0.43	0.25	0.26
ADI-13	0.5	--	--	--	--	--	--	0.39	0.34	0.12	0.27
ADI-11	0.8	0.29	0.23	0.27	0.42	0.44	0.16	0.14	0.11	0.06	0.08
ADI-05	1.5	0.34	0.28	0.34	0.84	0.44	0.25	0.15	0.10	0.06	0.08
ADI-06	1.5	--	0.06	0.05	0.20	0.16	0.06	0.07	0.03	0.02	0.04
ADI-08	2.5	0.21	0.27	0.19	0.34	0.15	0.14	0.08	0.06	0.03	0.08
ADI-09	8	--	--	--	0.08	0.08	0.03	0.06	0.01	0.02	0.05
ADI-02	10	--	--	--	0.11	0.12	0.19	0.05	0.03	0.02	0.02
ADI-04	10	--	0.05	0.02	0.07	0.05	0.03	0.04	0.01	0.02	0.02
ADI-10	11	--	--	--	0.05	0.02	0.02	0.03	0.02	0.01	0.01
ADB-01	25	--	--	--	0.03	0.02	0.03	0.03	0.01	0.00	0.01

The results of these calculations, reproduced in Table 6 show an almost monotonous decrease of the dispersion factor in the vicinity of Cernavoda NPP, in accordance with the increasing release of the DTO by the first unit of NPP.

4. CONCLUSIONS

The long term data concerning the distribution of ^{3}H in the vicinity of Cernavoda NPP between 1997 and 2007 as well as the corresponding data regarding atmospheric absolute humidity has showed a constant increase of the DTO amount released into atmosphere together with a steady decrease, for the same period of time, of the atmospheric humidity. These two factors finally determined a moderate decline of the amount of DTO registered at each of 11 observation points around NPP.

The same data showed an exponentially decrease of the ^{3}H fall-outs with the distance with respect to NPP, reaching the background level at 30 km off-site. It is worth mentioning that within Cernavoda town, the total supplementary dose due to tritium intake fluctuated between 0.27 and 0.69 μSv/y, three orders of magnitude lower than the maximum premised dose for nonocupational exposure.

In addition we have noticed a weak positive correlation between atmospheric humidity and DTO fall-outs, this fact attesting a relatively complex atmospheric circulation around Cernavoda NPP, in concordance with two other observations revealing the existence of weak correlations between observing points concerning the long term data regarding DTO fall-outs as well as atmospheric humidity.

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