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ANNUAL EXERCISES FOR DETERMINING THE PATHWAYS OF TRITIUM RELEASES FROM CERNAVODA NUCLEAR POWER PLANT WITHIN DOBROGEA ROMANIA. PART 2

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Abstract. The determination of the 'source-term' based on previous measurements of tritiated water in water samples (Part 1), as well as the possible effluent plumes from Cernavoda Nuclear Power Plant Romania led to a emission scenario as working basis for a future real-time simulation and sampling exercise within Dobrogea that will include event scenarios for radioactive atmospheric releases assessed by an expert software platform, developed at IFIN-HH.

Key words: Nuclear Power Plant releases, tritiated water, expert software platforms.

1. INTRODUCTION

Interpretation of the results of sampling campaigns from May 2017 and 2018, on the levels of tritium in inland waters in the central-northern part of Dobrogea highlighted the direct and indirect contribution of gaseous and liquid effluents from Cernavoda Nuclear Power Plant Romania (Cernavoda NPP) during scheduled shutdown. This influence has been observed beyond a limit of 5 km away from the source, on the Danube River downstream from Cernavoda to Tulcea, along the Danube–Black Sea and Poarta Alba – Midia Navodari Channels, on the Black Sea Coast at Mamaia–Constanta, and even along the artificial rainwater collection pools, near the A2 highway. This trend is closely related to source, requiring attention. Moreover, the existence of an interdependence along sampling points, between the values of tritiated water (HTO) expressed as tritium activity concentration in water samples was established. The correlation factors were calculated in the same year and in different places, but also for consecutive years in the same place. Annual distributions of tritium levels at one point were also anticipated, depending on the releases from the nuclear power plant throughout the year, taking into account the contribution of cosmogenic tritium. Using other experimental data collected during this period, apart from May, but also the experience of over 35 years in monitoring the Cernavoda nuclear objective, in this second stage of the exercise we tried to make post-event predictions of directions of the effluent plumes, depending on weather and climate conditions, in the normal operation mode of Cernavoda NPP [1–4].

Preparing of a future exercise on sampling – the real-time assessments will include event scenarios for atmospheric releases performed with a software platform, developed at IFIN-HH as part of a research project in the field of national security and safety [5], designed for anticipatory, prognostic assessments of a wide range of events involving radioactive, chemical and other hazardous substances. The software platform, (*CBRNE Software*) includes functions of event diagnostic and consecutive recommendation of response measures for an open and scalable variety of event scenarios [6]. In this particular case, the atmospheric release assessments performed with *CBRNE Software* will be developed in parallel with sampling of water upon the immediate recommendations, followed by sample processing and measurement of tritium as tritiated water (HTO), using liquid scintillation counting method (LSC method) at IFIN-HH, as described in Part 1 [4].

2. MATERIALS AND METHODS

2.1. Sampling points and sampling conditions

This two-part exercise is based on previous HTO determinations by LSC method, conclusions and correlations for two consecutive years (2017 and 2018) in central-northern Dobrogea, and on the experimental data acquired throughout 2018 in several sampling points in Cernavoda / Seimeni area such as Seimeni - dam / NPP Discharge Channel (N: 44.367875, E: 28.046411), Yahoo Best Cernavoda; artificial pool (N: 44.348428, E: 28.044542) (see section 3.1.). As 'zero point', samples having low-level tritium activities were taken from Inner lake Slanic-Prahova; Unirea Salt Mine; IFIN-HH microBq Lab (N: 45.235908, E: 25.942233). Geographical coordinates through the Google Earth application (Imagery Date: 2/21/2019) were obtained.

At the beginning, considering the normal operation regime, several distinct scenarios of liquid and gaseous effluent releases will be assessed, using as input data a series of values determined during this period around Cernavoda, as well as some general technical data on emissions of a CANDU type nuclear reactor.

Finally, the correlation of the two dispersion scenarios (atmospheric, by computer simulations, and water, by direct determinations), such as Cernavoda NPP current emissions, will result in a new perspective. Depending on the dominant weather and climate conditions at that time, this will include the contribution of HTO evaporation and dispersion by air humidity and the recombination of vapours with surface water from other areas (close range or far range).

All samples were taken in the same conditions as previous (see Part 1), from the shore or nearby, and from rainless periods.

2.2. Processing and measurements of water samples

Pretreatments consisted in a distillation stage at atmospheric pressure. Measurements of HTO tritium activity concentration were performed on Quantulus 1220™ ultra low-level liquid scintillation analyzer [7].

The Minimum Detectable Activity (MDA) was below 0.75 Bq/dm^3 ($\leq$ 7 TU). To achieve this performance, the vials containing distilled 'dead water' were measured four times longer than the samples. In this case, the MDA value was calculated according the recommendations written in the Reference Manual. As 'dead water', freshly-distilled Qlarivia™, depleted in tritium, was used.

The measured values were evaluated on the basis of a review on tritium levels in the environment, published in 2018 [8].

3. RESULTS AND DISCUSSION

3.1. Evaluations based on field campaigns results

The main results of the experimental input values are presented schematically in Figures 1 and 2. The description and / or ID number of the samples taken in that year were used. The values obtained in 2018 were compared with the landmark value of 6.6 TU obtained in the salt mine water from the Unirea Mine, Slanic-Prahova (Fig. 1). As higher values throughout the year 2018, those obtained every 2 months were compared, both in the waters of the Seimeni NPP Discharge Channel and in waters from the adjacent artificial pool, the mean value being 54 TU (Fig. 2). The higher values obtained in the isolated artificial pool show the prevalent contribution of tritium by air (gaseous effluents, evaporation and meteoric waters) compared to that by water, which is dominant in the values obtained for the discharge channel.

To begin with, using HTO tritium activity concentration values recorded between 2016 and 2019 at the IFIN-HH laboratory in water samples taken around Cernavoda NPP, an estimate was made of the total emission value expressed as BqT/m^3 water equivalent, over a period of time (in hours). This estimate is necessary in defining the 'source-term' subsequently used to generate real-time forecasts, based on input parameters.

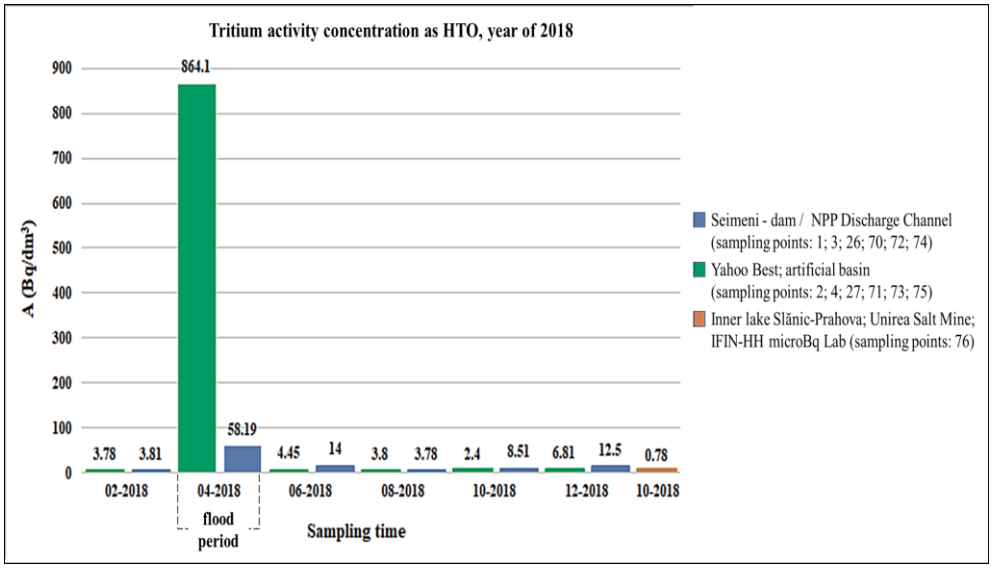

Fig. 1 – The values of the tritium activity concentration as HTO in 2018, determined for the Cernavoda 'source-term' and for the 'zero point', placed in the Unirea Salt Mine at Slanic-Prahova.

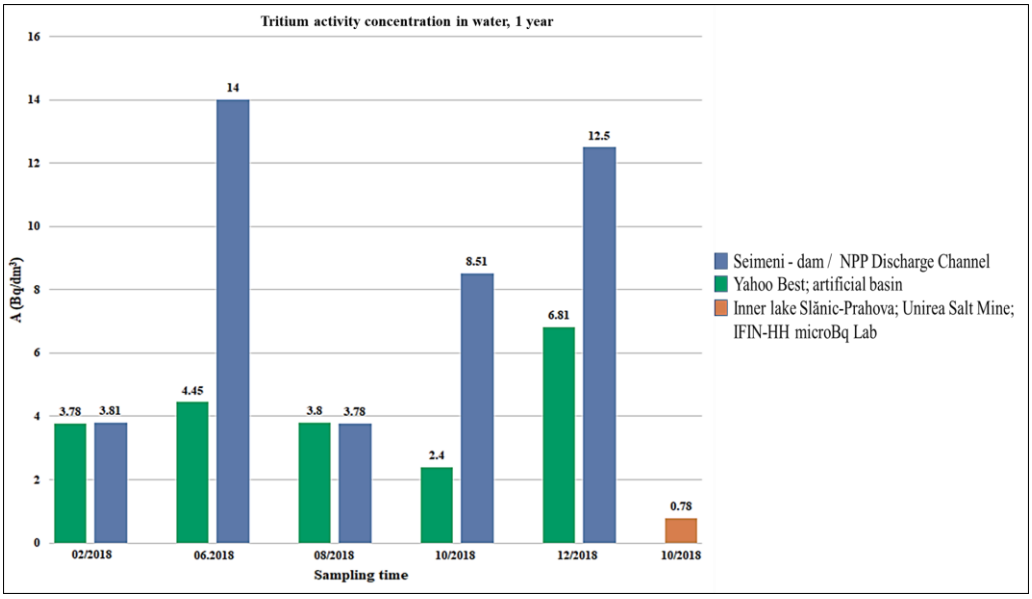

Fig. 2 – One year survey around Cernavoda NPP, 2018.

For the time period considered the emissions of the Cernavoda NPP are estimated to be as: through water in the Seimeni NPP discharge channel, into the inlet channel of the nuclear power plant and at the junction with the Danube–Black Sea channel and through air into the atmosphere and rain water.

Taking into account the data for water samples collected at the Seimeni NPP Discharge Channel (the main direct source of samples for releases in water form) and the Yahoo Pool (Fig. 3a), located approx. 2,280 meters S/SE on the Channel route (indirect source for releases into the atmospheric air and rainwaters), the following distribution shown in Table 1 emerged. It started in 2017, at the end of the scheduled shutdown of both Cernavoda NPP Units, which was considered as a starting point for a realistic post-event reconstruction of the definition of 'source-term'. The sampling from April 2018, where the highest values were obtained in both points, was also an outlier situation in terms of weather and climate which was taken into consideration.

We assume that: a) the water flow of the Channel is 25 m^3/s; b) the time interval is of 12 hours when the flood occurred at maximum levels, corresponding to a realistic value of the tritium activity concentration mediated (Table 1); c) the value for the water coming from the floods was taken on a point ('arrow', Fig. 3b)

further from the Channel route, at the edge of the flood perimeter, defined by a rectangle with approximate dimensions of l = 735 m; L = 3,450 m (Fig. 3b); d) the average depth of 1 m is taken into consideration to estimate a volume of water which will largely correspond to the tritium activity concentration determined at the sampling point ('arrow', Fig. 3b); e) Yahoo pool is a 'huge passive collector', considering that it has the same exchange characteristics as those used in the laboratory measurements. With these assumptions, the following input data emerged: a) before the flood, A_{in} = 2.5 Bq/dm^3 – after 30 days (cover-interval for the state of equilibrium to characterize the phenomenon of April 2018) a calculation method will be applied starting from A_{fin} = 58.2 Bq/dm^3; b) before the flood, A_{in} = 2.5 Bq/dm^3 – after 0.5 days or 12 hours (assuming that the equilibrium state that characterizes the phenomenon of April 2018 is reached) another calculation method will be applied starting from A_{fin} = 58.2 Bq/dm^3; c) for Yahoo ornamental pool, given that it is an indirect value that expresses the concentration in the air, A' will be expressed in Bq/m^3 of air, and the calculation formulas specific to 'passive collectors' will be applied [3].

Therefore, through the calculations shown below, it is possible to obtain an estimate in Bq/m^3 of water *versus* Bq/m^3 of air, to ascertain the distribution ratio of releases between the two main pathways of propagation.

Table 1

HTO tritium activity concentration around Cernavoda NPP, corresponding to the years 2017, 2018, 2019

Sampling point		**Sampling Time**	**Year**
Seimeni - dam / NPP Discharge Channel	**Yahoo Best; artificial pool**		
***A* (Bq/dm^3)**	***A* (Bq/dm^3)**		
24.83	–	May	2017
6.39	–		
11.95	–		
3.79	–		
4.39	–		
4.07	–		
314.82	**34.34**	June	
–	15.50	December	
3.78	3.81	February	2018
864.10	**58.19**	April	
4.45	14.00	June	
–	12.50		
4.52	6.80	August	
3.80	3.78		
2.40	8.51	October	
6.81	12.50	December	
3.45	7.44	April	2019
4.45	14.00	September	
10.85	9.73	October	
8.21	2.43	December	

Consequently, the following have been obtained: volume of water discharged at an average water flow of 25 m^3/s: 12×60×60×25 = 1,080,000 m^3; the assumed volume of water given the rectangle of the flood area: 735×3,450×1 = 2,535,750 m^3; experimentally determined average tritium activity concentration, in this volume: 865 Bq/dm^3 = 865,000 Bq/m^3. Compared to the route-source of water elimination from the nuclear power plant, considered at an associated average depth (therefore not real!) also of approx. 1 m, the ratio between the two volumes will be: 2,535,750 / 1,080,000 ≅ 2.35 times. Basically, this factor dilutes what the nuclear power plant discharges in this rectangle, for that moment of the flood. The tritium activity concentration at the release source on the waterway before the dam will be 865×2.35 = 2,033 Bq/dm^3 of water, 2,033,000 Bq/m^3 of water, respectively. For the sequence that generates tritium releases into the air (in the gaseous effluents, in the humidity of the atmospheric air), at equilibrium with the water exchange interface in the Yahoo ornamental pool (water at the initial value + the addition of air releases + the addition of atmospheric humidity including evaporation from the surrounding waters + contribution from previous meteoric

waters that reached the uncovered basin), the associated value of the water in the basin was 58.20 Bq/dm^3 (also, experimentally determined). Considering that the Yahoo ornamental pool operates as a 'passive collector' without precipitations in equilibrium with the environment at the time of sampling, then the following situations can occur: (i) for an interval of 30 days, before, during and after flooding $A' = F \times 58.20 / 5.6 \times 30 \times 10^{-3}$ = 3,465 Bq/m^3 of air (where F is the transfer factor = 10 times; 10% of the information is transferred); (ii) for an interval of 12 hours, before, during and after flooding $A' = F \times 58.20 / 5.6 \times 0.5 \times 10^{-3}$ = = 207,857 Bq/m^3 of air (where F = 10 times; 10% of the information is transferred). The simultaneous sampling time ratio will be: 2,033,000 Bq/m^3 of water *versus* 3,465 Bq/m^3 of air: 0.17 / approx. 0.20%; 2,033,000 Bq/m^3 of water *versus* 207,857 Bq/m^3 of air: approx. 10%, much more realistic compared to what is usually released as effluents in water and air from Cernavoda NPP. As a consequence, this value can be considered formally correct. If we consider 15% emissions of tritium in water and other pathways mentioned above: 2,033,000 + 304,950 + 207,857 = 2,545,807 Bq/m^3 water-equivalent. Basically, this unexpectedly high value of tritium activity concentration helps, through the specific conditions in which the sampling and determinations were made, to be considered as a starting point into the realistic post-event reconstruction of the 'source-term' at that time. The percentage by type of effluent, and the time in which the equilibrium occurred through the water / atmospheric air interface, can be given.

Fig. 3 – a) Sampling points around Cernavoda NPP; b) flood time around Cernavoda NPP, April 2018.

3.2. Evaluations based on the source-term calculation

Assuming that:

– *For tritium releases in water and air effluents, higher than the daily norm*, determined by water samples taken at the Seimeni NPP Discharge Channel and into the Yahoo ornamental pool, at a time of year favorable for dispersal, meaning: spring, wet season, maximum period of cosmogenic tritium; particular case: stationary weather-climatic conditions, higher release values coupled with massive floods on the Danube River and adjacent floodplains / channels / lakes. For intervals of 12 consecutive hours, as shown above in detail, the concentrations in water and into atmosphere predominate over the tritium activity concentrations encountered at this time of year, in the two forms of dispersion:

(**12**×60×60×25×2,545,807) / 3.7×10^{10} = 2.75×10^{12} / 3.70×10^{10} = 74.3 Ci

(**24**×60×60×25×2,545,807) / 3.7×10^{10} = 5.50×10^{12} / 3.70×10^{10} = 148.6 Ci

(**48**×60×60×25×2,545,807) / 3.7×10^{10} = 11.00×10^{12} / 3.70×10^{10} = 297.2 Ci

(**72**×60×60×25×2,545,807) / 3.7×10^{10} = 16.50×10^{12} / 3.70×10^{10} = 445.8 Ci.

Compared to the 'source-term' defined in the previous exercises, respectively 1.27×10^{10} Bq [9], the values estimated above are several hundred times higher, 200–1300 times respectively. Most likely, the first value would be associated with routine releases. Compared to the average of the usual values obtained over the years in the Seimeni NPP Discharge Channel, the value determined in the water for this event is approx. 200–400 times higher.

– *For tritium releases in water and air effluents within the daily norm*, determined by the water samples taken at the Seimeni NPP Discharge Channel and into the Yahoo ornamental pool, there are several conditions throughout the year. In January, February and March, when a low ambient temperature and low relative humidity occurred, the mediated experimental value of HTO tritium activity concentration (Bq/dm^3) is 3.78 (water) / 3.81 (air) ($F \cong 1$). In April, May and June, with relatively low temperatures and high relative humidity, in the conditions of cosmogenic tritium and shutdown scheduled at Cernavoda NPP, the mean values outside the shutdown period are 3.45 (water) / 7.44 (air) ($F \cong 2.15$). In July, August, September, with higher temperatures and higher relative humidity, the values are 3.80 (water) / 3.78 (air) ($F \cong 1$). In October and November, in the conditions of relative low temperatures and high relative humidity, average values of 4.45 (water) / 14.00 (air) ($F \cong 3.14$) are encountered. Finally, in the month of December, characterized by low temperatures and low relative humidity, the values are 6.81 (water) / 12.50 (air) ($F \cong 1.83$).

3.3. Radioactive effluent plumes assessments

The assessments were made based on the calculated 'source-terms' and taking into account the predicted weather-climatic conditions. For the atmospheric emissions simulations, a thematic module of *CBRNE Software* was run, addressing issues related to nuclear risks and threats that integrates, among others, the 'Close range plumes' application, part of the software platform's portfolio. This application is considered as a training-oriented simulator of radioactive atmospheric releases featuring a comprehensive capability to generate scenarios involving various source locations and types, nuclide mixes, release regimes, terrains, and weather conditions. Implementing an extended Gaussian plume model fits assessments at close range to sources [10]. The step-by-step working sequence requires input data regarding the source (location, nuclide mix, activity); release (duration, height, plume rise); exposure duration; terrain (scale, land type) and meteorology (dispersion system, wind speed and direction, ambient temperature and pressure). The output data resulting from running the 'Close range plumes' are delivered at interface as *interactive situation maps* representing the radioactive effluent plume footprint on the ground as well as the output data files of the evaluated cases including effective dispersion parameters, *dilution factor values* and *dose to distance dependencies* for a variety of doses. The input data were as follows: Source-term mix (nuclides): H-3 (tritium); Activity (kBq): 2.75×10^9, 5.5×10^9, 11×10^9 and 16.5×10^9; Release Duration (h): 12, 24, 48 and 72; Mouth Height (mAG): 50; Plume Rise (m): 0; Dispersion System: Briggs; Wind Speed (m/s at 10 mAG): 1 and 4; Wind direction (deg): 170; Ambient Temperature (^{o}C): 13. Four tritium release scenarios with different activities and release durations were assessed: Scenario 1: 2.75×10^9 kBq, 12 h; Scenario 2: 5.5×10^9 kBq, 24 h; Scenario 3: 11.0×10^9 kBq, 48 h; Scenario 4: 16.5×10^9 kBq, 72 h.

Figure 4 shows the interactive maps of the radioactive effluent footprint on ground from source and Fig. 5 shows the dose to distance dependencies of Total Effective Dose Equivalent (TEDE) for all four release scenarios, considering the above input data for activity (kBq), release duration (hours) and wind speed (m/s). Also, Fig. 5 shows that, for both wind speeds, all release durations and activities, the maximum values of TEDE are around 230 m from source and minimum values are around 10 km from source. The maximum impact will be given by a source with an activity of 16.5×10^9 kBq, a release duration of 72 hours, both wind speeds, at distance up to 1,000 m from source (see Scenario 4). The highest value of TEDE (5.60×10^{-3} mSv), can be in Scenario 4 too, at a wind speed of 1 m/s. It has to be noticed that, although the values of the tritium activity concentration (Bq/dm^3 water-equivalent) of the 'source-term' are relatively high, the values of TEDE are less than 0.01 mSv in all scenarios. The contribution of tritium becomes insignificant in terms of dose values over the period 12−72 hours, under the release conditions considered.

Also, the output data resulting from running the 'Close range plumes' application shows that, in all scenarios, at a distance of around 230 m from source, the value of Maximum Dilution Factor (s/m^3) is 2.26×10^{-5} for a wind speed of 1m/s, and 5.65×10^{-6} for a wind speed of 4 m/s and, and at a distance around 10 km from source, the value for Extreme Dilution Factor is 5.65×10^{-8} for a wind speed of 1m/s and 1.41×10^{-8} for a wind speed of 4 m/s, respectively. Thus, the values of Maximum Dilution Factor are more than 400 times higher than Extreme Dilution Factor, for all scenarios.

Summarizing, we took into account the real ground conditions from April 2018, that is, the presumption of 12 hours of emission, stable weather, relative humidity of 42% at 13 ^{o}C, average wind speed of 1 m/s, dominant wind direction from SE and a distance greater than 1,000 m from source.

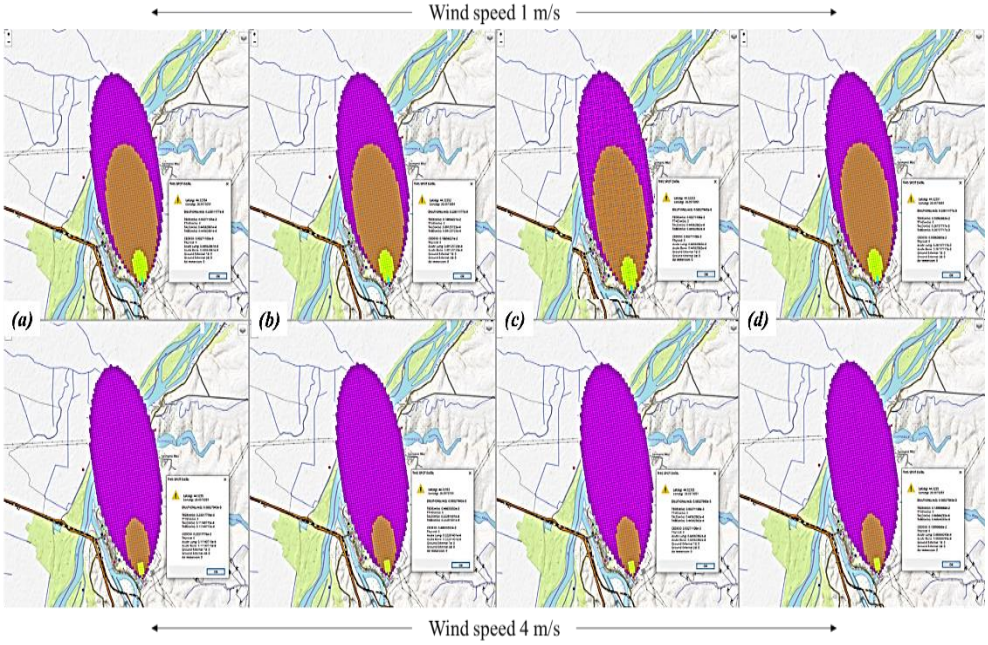

Fig. 4 – Interactive maps of the radioactive effluent plume footprint on ground from source for different activities, wind speeds and release durations:
a) Scenario 1: 2.75×10^9 kBq / 12 h; b) Scenario 2: 5.5×10^9 kBq / 24 h;
c) Scenario 3: 11.0×10^9 kBq / 48 h; d) Scenario 4: 16.5×10^9 kBq / 72 h.

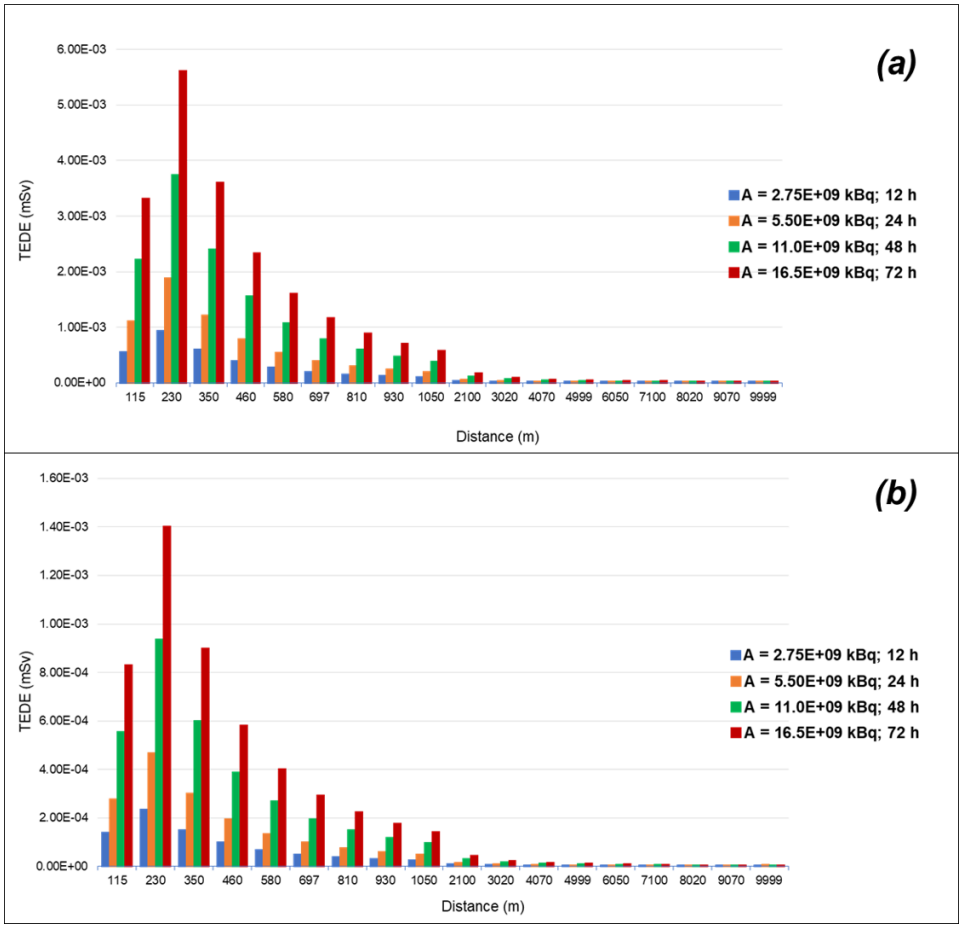

Fig. 5 – Dose to distance dependence of Total Effective Dose Equivalent (TEDE) for different wind speeds:
a) – wind speed of 1 m/s; b) – wind speed of 4 m/s.

Although the values measured in the water samples taken during floods are relatively high in terms of tritium activity concentration, exceeding the limit allowed in inland waters as a source of drinking water, they contribute insignificantly to the dose values and represent the most favorable of all simulations. For the other situations encountered in the sampling campaigns presented, the simulations become redundant. However, the 72-hour release scenario in stable, humid weather, and at an average wind speed of 1 m/s is considered to be the *'worst case scenario'*. It may be taken into account in a future exercise that will follow the evolution of the radioactive release in real time, depending on the dominant wind direction and its change in this interval. Moreover, Figure 4 shows that the area chosen for the 'source-term' assessment has been correctly defined, and Yahoo Best artificial pool, also considered, is approximately in the middle of the effluent plume in all situations. For the field conditions considered, the effluent plume extends beyond the Danube. Therefore it is worth taking a series of water samples on the other side of the river. A previous exercise established the existence of differences not only downstream / upstream, but also between the two banks of the river.

4. CONCLUSIONS

Based on a set of data acquired between 2016 and 2019 for the determination of tritium in environmental samples in Dobrogea, a first attempt of post-event simulation of the impact of releases from Cernavoda NPP was conducted. For the atmospheric assessments, the step-by-step working sequence required a set of input parameters such as the source characteristics, release information, exposure duration, terrain type and meteorological conditions during releases. By approximating all these parameters, and comparing with experimental results and conclusions on volumic activities, several release scenarios were built-up. Among all possible situations, the *worst case scenario* was for a source activity of 16.5×10^9 kBq, a release duration of 72 hours, and a wind speed of 1 m/s. However, even in the worst case resulted by simulation of the input data considered, the contribution of tritium released by gaseous and liquid effluents is insignificant in terms of dose values.

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