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

Corina Anca SIMION[1], Aurora RANCA[2], Vasile PATRASCU[3], Iuliana Madalina STANCIU[1,4]

[1] "Horia Hulubei" National Institute for Research and Development in Physics and Nuclear Engineeering, 30 Atomistilor St., 077125 Magurele Ilfov County, Romania
[2] Murfatlar Vineyard – Murfatlar Viticulture-Vinification Research and Development Station (SCDVV Murfatlar), 2 Bucuresti St., 905100 Murfatlar Constanta County, Romania
[3] "Grigore Antipa" National Institute for Marine Research-Development, 300 Mamaia Blvd., 900581 Constanta, Romania
[4] Technische Universität München, Physik Department, 85748 Garching, Germany

Corresponding author: Corina Anca SIMION, E-mail: anke@nipne.ro

Abstract. Possible scenarios, correlation factors and propagation intervals of direct releases in liquid effluents and through indirect contributions by gaseous effluents from Cernavoda Nuclear Power Plant Romania to the tritium inventory in water samples from the central-northern area of Dobrogea are proposed. Part 1 focuses on experimental data.

Key words: Nuclear Power Plant releases, tritiated water, Liquid Scintillation Counting method.

1. INTRODUCTION

As a first step in preparing a future exercise of sampling-simulation in real time, this paper presents new useful information on the direct and indirect propagation/dispersion through inland waters, of tritium releases from the Romanian Cernavoda Nuclear Power Plant (Cernavoda NPP).

Because the main directions of dispersions over inland waters of nuclear releases around the town of Cernavoda are in the north-northeast direction (Danube River, downstream of the Seimeni NPP Discharge Channel), east-northeast (Poarta Alba - Midia Navodari Channel) and east-southeast (Danube - Black Sea Channel), the defined perimeter will be placed within the central-northern part of the Dobrogea region, mainly between the right bank of the Danube River (to the west), the Danube Delta (to the north), the Black Sea Coast (to the east) and the Carasu Valley (to the south). The following points were chosen in the sampling strategy: (i) the town of Murfatlar (famous for its wines) for the region along the Danube - Black Sea channel; (ii) the interface zone between Constanta and Mamaia (the most important Romanian Black Sea resort) for the seaside area; (iii) the Tulcea Port for the Danube River, due to its importance and some particularities of the entrance of the river into the Delta, and (iv) the town of Cernavoda.

The exploratory sampling exercises were developed on the basis of previous results and of the expertise of the involved teams [1, 2]. In order to complete information and define as accurately as possible the 'term-source' for tritium, both needed in the second part of this exercise, due to the lack of relevant data on the actual releases of tritium in the environment, some of the already published Cernavoda NPP surveys and Annual Reports were taken as benchmarks [3, 4].

The current experiments involve monitoring over two consecutive years (2017 and 2018). The variation of the values of the tritium activity concentration present as tritiated water (HTO) in water samples were determined by daily grab sampling, at the same times, in Murfatlar, Constanta, and Tulcea. These values were compared to those obtained in the Cernavoda perimeter or on the Danube downstream, and also to those determined on environmental samples in other areas of Romania that were considered as 'zero points' of the tritium level. The trends for the years 2016 and 2019 for the Dobrogea region will be taken into account too.

2. MATERIALS AND METHODS

2.1. Sampling points and sampling conditions

The idea of this two-part exercise was triggered by the 10th anniversary of the actual commissioning of U2 at Cernavoda NPP.

For the reasons described in previous chapter, five representative areas were targeted:

- Cernavoda / Seimeni, with sampling points sustained by IFIN-HH team: direct influence of gaseous and liquid effluents (Fig. 1a; Flags A, R, S, C, Q);
- Danube River downstream Cernavoda to the river entrance into Danube Delta, with sampling points sustained by IFIN-HH team; Tulcea: point Tulcea-2 was chosen in the most exposed area of the Harbor, compared to Tulcea-1, located in a gulf area, away from the river course, with a logistic sampling point near Tulcea Harbor, sustained by volunteers: indirect influence of gaseous effluents + direct discharges of liquid effluents (Fig. 1b; Flags D, E, F, G, I);
- Danube – Black Sea Channel and Poarta Alba – Midia Navodari Channel with a logistic sampling point at Murfatlar Vineyard – Murfatlar Viticulture-Vinification Research and Development Station (SCDVV Murfatlar): indirect influence of gaseous and liquid effluents (Fig. 1c: Flags B, O);
- Tabacarie Lake – Constanta/Mamaia, with a logistic sampling point near Grigore Antipa National Institute for Marine Research-Development Constanta: indirect influence of gaseous effluents + without influence of liquid effluents + direct influence of the coastal maritime area (Figure 1c; Flag H);
- Artificial basins along A2 Highway from Bucharest to Constanta; Dobrogea area, between Medgidia and Murfatlar directions, with sampling points sustained by IFIN-HH team: indirect influence of gaseous effluents + without influence of liquid effluents (Fig. 1c, Flags B, J–P).

The other sampling points mentioned in this paper were sustained by the contribution of IFIN-HH team and occasionally other volunteers from these regions. The rainwater contribution was not taken into consideration. Samples were taken after periods without rain or during rains, the latter case being mentioned in text.

Thus, during May 2017, a first exploratory campaign took place, especially since both units entered the scheduled shutdown, one after the other, this aspect being a special feature that deserves to be investigated. At the end of the 2017 campaigns, the seawater sampled from 2 Mai Beach (43.789098^{o} N, 28.581135^{o} E), at the southern end of the Romanian Coast, was taken as a landmark. The whole year 2017 cannot be considered a year with special phenomena from the point of view of the tritium inventory, by comparing with the previous year.

As is the case for 2018, the field campaigns being resumed to the first four areas from the previous year. At the end of the campaigns, the water from the inner infiltration lake located in Unirea Slanic-Prahova Salt Mine was taken as a landmark; working point microBq Lab IFIN-HH (45.235908^{o}N, 25.942233^{o}E) [5]. The year 2018 cannot be considered a year with special phenomena by comparison with the general trend recorded in Dobrogea for 2019.

The coordinates of the sampling points from 2017 and 2018 campaigns, obtained using Google Earth™ (Imagery Date: 12/2010) were: Seimeni – dam / NPP Discharge Channel – **Flag A (**44.367875, 28.046411); Danube – Black Sea Channel; Mile 23 Murfatlar – **Flag B (**44.169129, 28.408464); Danube - Black Sea Channel; before SEIRU – **Flag C (**44.307714, 28.054743); Capidava – **Flag D (**44.495000, 28.087303); Topalu – **Flag E (**44.527992, 28.046863); Daeni-Frecatei – **Flag F (**44.851617, 28.110329); Tulcea-1 / **Flag G (**45.183690, 28.807584); Tabacarie Lake – **Flag H (**44.211790, 28.641470); Tulcea-2 / **Flag I (**45.182048, 28.804911); Valu lui Traian – Cumpana / **Flag J (**44.142950, 28.514320); Highway; after parking area on Constanta route; artificial basin – **Flag K (**44.140310, 28.456040); Highway on Danube - Black Sea Channel; water from artificial basin – **Flag L (**44.138156, 28.440798); Danube - Black Sea Channel; water from channel, near the artificial basin – **Flag M (**44.138011, 28.439611); Highway; near Petrom Station; artificial basin – **Flag N (**44.151470, 28.356090); Poarta Alba – Midia Navodari Channel; 5 km downstream – **Flag O (**44.236001, 28.448143); Medgidia; under Bridge – **Flag P (**44.250095, 28.261423); Danube – Black Sea Channel; SEIRU – **Flag Q (**44.302595, 28.055475); Yahoo Best Cernavoda; artificial basin – **Flag R (**44.348428, 28.044542); Cernavoda NPP – **Flag S (**44.321892, 28.057901). The Flags were associated to the sampling points as the samples were grabbed.

All samples were taken from the shore or nearby, either from quiet areas outside the watercourse or from the water's edge, not deeper than 50 cm from the surface. Regarding sampling stage, the tests performed at IFIN-HH in 2017, but also the recommendations in the literature imposed a special care on the conditions of sampling, transport and storage until the time of processing [6, 7].

Fig. 1 – Sampling points areas from 2017 and 2018 campaigns, obtained using Google Earth™ (Imagery Date: 12/2010):
a) Cernavoda – Seimeni;
b) Danube River downstream Cernavoda to the river entrance into Danube Delta;
c) Danube – Black Sea Channel and Poarta Alba - Midia Navodari Channel; Tabacarie Lake – Constanta / Mamaia. Artificial basins along A2 Highway from Bucharest to Constanta; Dobrogea area, between Medgidia and Murfatlar directions.

2.2. Processing and measurements of water samples

Regarding pretreatments in the laboratory stage, working conditions and special measures are governed by literature recommendations as well by the standards in force [8, 9].

Thus, the pretreatment consisted of distillation at atmospheric pressure using the Dimroth-Claisen separation system, and avoiding the contact with the air from laboratory. The improvements were largely described in a previous publication [9]. The distillates were added into 20 mL PE vials, in a volume ratio of 5:15 to the scintillation cocktail, in this case Ultima Gold uLLT®. The measurement basic conditions on Quantulus 1220™ were: fixed window (5–320 channels), SQP(E) single label, sample preparation error 1.6%, 10×100 min/vial/sample. The Minimum Detectable Activity (MDA) was initially below 1.06 Bq/dm^3 (≤ 9 TU; 1 TU = 0.118 Bq/dm^3). The continuous improvement of the measurement conditions lowered this threshold value below 0.75 Bq/dm^3 (≤ 7 TU). Being a relative measurement, the LSC method finally used as 'dead water', practically free of tritium compared to the analyzed samples, freshly-distilled Qlarivia™ double-distilled water in a boron-silicate laboratory glassware installation, specially designed and used only for this purpose. These performances may be considered as representing reasonable values in terms of MDA, taking into account the studied region and the global or regional general trends in this area [6, 7, 10–12].

The campaign scheduled for May 2020 was postponed for the same period in 2021, due to the COVID-19 Pandemic and the traffic restrictions imposed.

3. RESULTS AND DISCUSSION

3.1. Evaluations based on field campaigns results

The results of the sampling campaigns from 2017 and 2018 are presented schematically in Figs. 2–5. To each sampling point (Fig. 1), i.e., description and/or ID number of the sample taken in that year were used. The values in the graphs have occasionally been associated with other values from 2016–2019 interval.

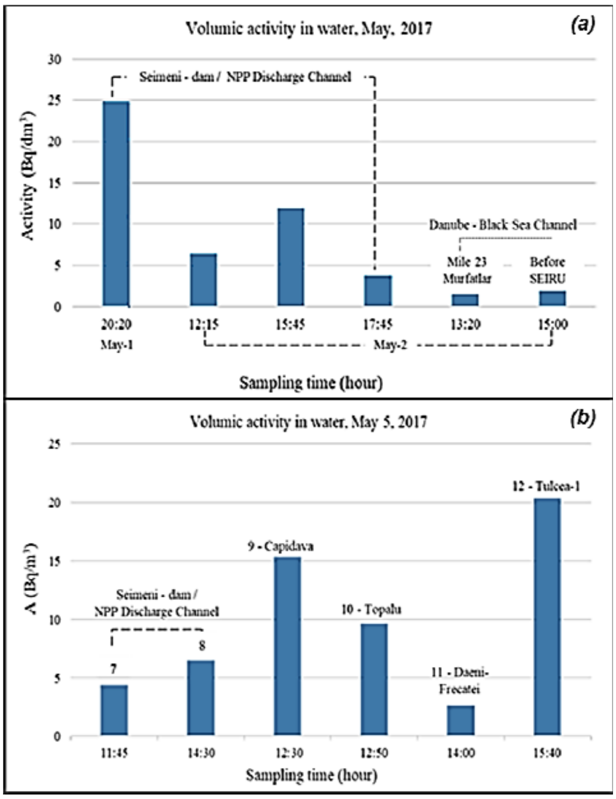

Fig. 2 – HTO activity concentration in water taken at different hours, May 2017:
a) Seimeni dam / NPP Discharge Channel and Danube – Black Sea Channel;
b) on May 5th, downstream of Cernavoda; logistic sampling points 7 to 12.

The primary conclusions for the 2017 campaign (Figs. 2–4) were: there is a noticeable variation in the values of the HTO activity concentration in the waters of the NPP Discharge Channel, formally correlated with the days / hours of sampling, and the flow. The effects could be detected downstream, up to Tulcea and there is a difference between Tulcea-1 and Tulcea-2. On the main river, due to the flow rate, the values are relatively uniform and lower than those obtained in quiet areas of the gulf, towards the shore. Remote effects could be detected at all sampling points and there was a correlation between the values obtained, over time. The values of the HTO activity concentration determined in the Lower Danube Basin in the time interval (2010–2018) are generally between 7 TU and 33.5 TU on the main watercourse [6, 7, 10, 11]. Compared to these, and to the mean value determined at the IFIN-HH laboratory in water samples on the Dobrogea area in 2016, of 64.6 TU for shallow surface waters, the values recorded in 2017 do not exceed the ranges, the average of the values, except for points 60, 61, 62 (Fig. 4), being 37.3 TU. The higher value determined in the discharge channel in the middle of the exercise (Fig. 4) could be correlated with the entry of the second unit in the scheduled shutdown, and therefore the reiteration and extension of the sampling interval in 2018 was justified.

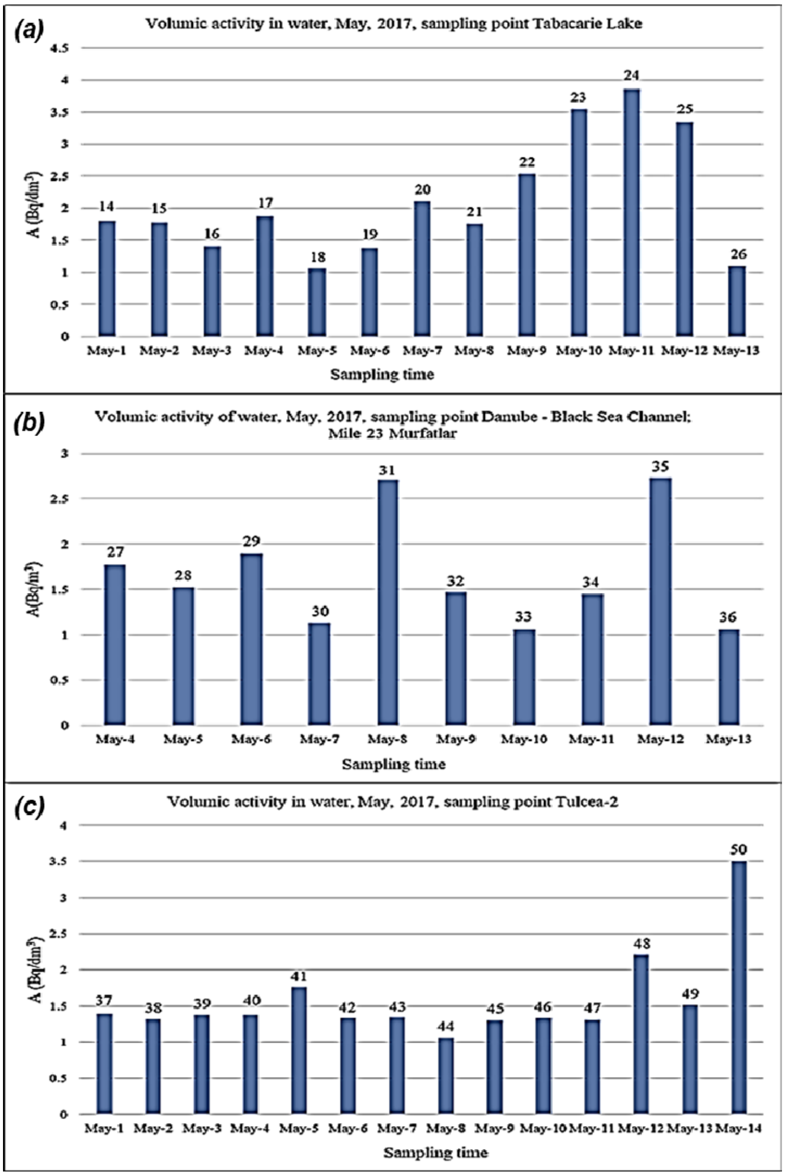

Fig. 3 – HTO activity concentration in water taken in different areas, May 2017:
a) Constanta-Mamaia area, Tabacarie Lake; logistic sampling points 14 to 26;
b) Murfatlar area, Danube-Black Sea Channel, Mile 23; logistic sampling points 27 to 36;
c) Tulcea Harbor area; logistic samplig point 37 to 50.

However, a higher value is noted at the Seimeni NPP Discharge Channel dam before May 2nd, 2017 (Fig. 2a). For this point, on the first day of the scheduled shutdown period of the first unit, a timing distribution appears that highlights the dilution regime using Danube water from the Intake Channel, for the release of liquid effluents into environment. Nevertheless, the levels of tritium, expressed as dose units for one person from population (microSv/year), are far below the maximum limits allowed for the normal functioning of Cernavoda NPP [4]. Three days after entering the scheduled shutdown period, the results recorded on May 5th, 2017 at several sampling points along the Danube River downstream show fluctuations, suggesting previous releases in environment. The cumulative interpretation of Figs. 2a and 2b, according to the geographical positions of the sampling points (Fig. 1), suggests that direct releases into liquid effluents downstream of the Seimeni are felt on the Danube River, on the shore, in shallower calming places, in the coming days, and that the possible higher releases from the beginning of this time interval can be highlighted even in the gulf area of Tulcea Harbor, after at least 4 days. In the Constanta / Mamaia area, the water samples taken in Tabacarie Lake between 1 and 13 May 2017 also show fluctuations beyond the variations given by the measurement statistics, with an increasing trend that reaches a maximum on May 11th, after which it starts to decreases (Fig. 3a). In principle, the maximum at approx. 3 days without raining could mark the duration of transmission mainly by air of the effects of gaseous effluents in the given meteo-climatic conditions, at the seashore. Slightly different in appearance, the graph for the values recorded in the Danube – Black Sea Channel at Murfatlar shows the cumulative influence of liquid and gaseous releases from Cernavoda NPP, showing fluctuations at 2-4 days, and two highs on May 8th and 12th, after which it shows a descending trend (Fig. 3b). In the exposed area of the Tulcea Harbor, after two relatively small maxima, a visibly higher maximum appears on May 14th. It could represent a stronger release around May 11th, also felt in the other two locations (Fig. 3c).

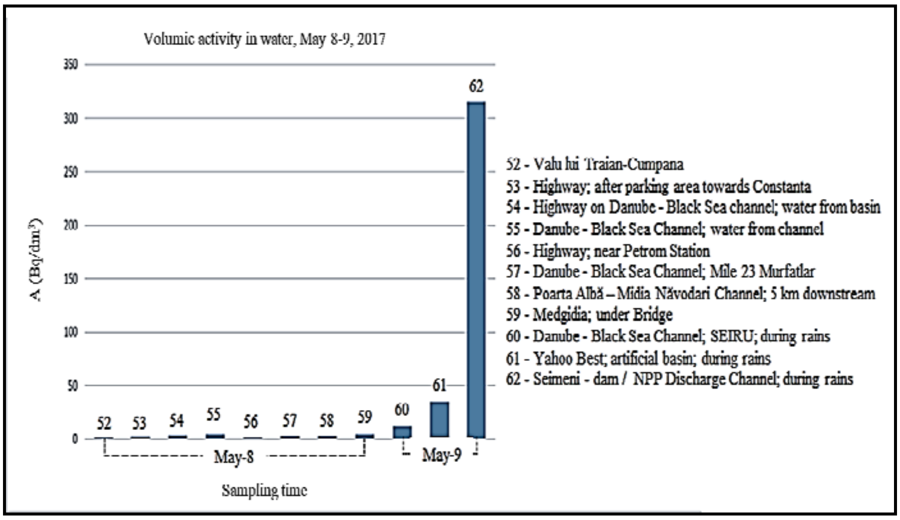

Fig. 4 – HTO activity concentration in water, May 8-9, 2017; logistic sampling points 52 to 62.

During this period (1–14 May), on May 8th, samples were taken from the artificial basins associated with the A2 Highway (Figure 1c). Depending on their proximity to the Danube – Black Sea Channel, these values increase, although they are placed at about the same distance from Cernavoda area. Moreover, the value in the basin near the Channel is the highest, but it is lower than that of the water from the Channel himself, which is placed at approx. 100 m distance (Fig. 4). Although it is closer to the nuclear power plant, Sample No 56, also associated with an artificial basin near the A2 Highway, has values similar to those furthest from the water source in the Channel (Sample No 52). Fluctuations in the channels are also highlighted (Samples No 55, 57, 58, 59). Resuming, there are fluctuations in the HTO activity concentration on these channels on 2–4 consecutive days, as well as for the same day, but at different distances from Cernavoda NPP. In principle, there is a cumulative effect of gaseous releases from Cernavoda NPP at the interface with water luster, but which combines, depending on the distance and the weather-climatic conditions, with the increased atmospheric humidity by evaporation near the waters flowing through the connecting channels of Cernavoda NPP with the Black Sea, but also with rainfalls. On May 9th, 2017, in a situation of atmospheric humidity close to 100% and on a front of significant rains, the values obtained around Cernavoda best show the cumulative contribution of the nuclear power plant to the tritium inventory in the neighboring waters (gaseous releases, liquid releases, re-evaporation of water bodies and meteoric waters). An important maximum was reached in the waters of the Seimeni NPP Discharge Channel (Sample No 62). This value, possibly associated with the scheduled shutdown of the other unit at Cernavoda NPP, was most likely felt in the following days in Tulcea, Constanta, Murfatlar, as presented above. As landmark in 2017, the seawater from 2 Mai Beach was chosen. Being located at the southern end of the Romanian Coast, its HTO activity concentration value was under MDA (≤ 1.06 Bq/dm^3).

In 2018, the similar comparative analysis of the results obtained from the simultaneous exercises from 1 to 21 May, carried out in Tulcea (with several maxima placed at about 4 days interval), Constanta (with several maxima placed at 3 days interval), and Murfatlar (with several maxima randomly placed between 4-6 days apart), showed evolutions similar to those of the previous year, distinguishing an important maximum in all locations around May 10th, with decreasing trends after May 21st (Figs. 5a–c).

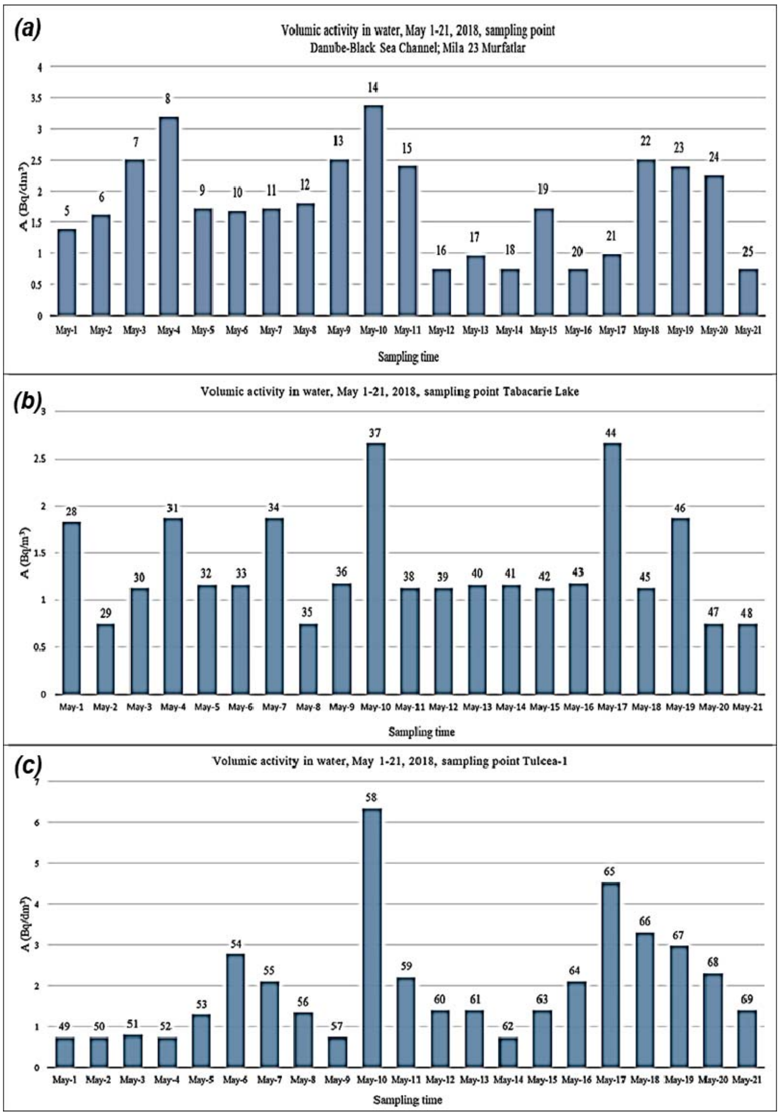

Fig. 5 – HTO activity concentration in water taken in different areas, May 2018:
a) Murfatlar area, Danube – Black Sea Channel, Mile 23; logistic sampling points 5 to 25;
b) Constanta – Mamaia area, Tabacarie Lake; logistic sampling points 28 to 48;
c) Tulcea Harbor area; logistic samplig point 49 to 69.

The values obtained in 2018 were compared with the landmark of 6.6 TU, obtained in the salt mine water from the Unirea Mine, Slanic Prahova. The mean value for 2018 was 23.24 TU.

4. CONCLUSIONS

Compared to 2017, the values of the HTO activity concentrations in 2018 for the same location were placed in a weak anti-correlation for Tulcea and Murfatlar, and were relatively correlated for Constanta, Lake Tabacarie.

The comparison per single year between locations, established by the Correl / Excel 2010 function, showed that in 2017 Constanta and Murfatlar were weakly anti-correlated (–33.22%), Constanta and Tulcea were poorly correlated (13.62%), while Murfatlar and Tulcea were semi-correlated (48.68%), most likely these results showing the dominant contribution of liquid effluents and their derivatives (evaporation and meteoric waters). For 2018, the correlations go much better: Constanta and Murfatlar (27.85%), Constanta and Tulcea (64.74%), Murfatlar and Tulcea (29.63%). This time, the dominance is most likely given by the releases in the gaseous phase and by the predominant wind direction, which correlates much better the values in the central-northern area of Dobrogea.

In the case of the annual evolution of tritium levels at Murfatlar, the influence of nuclear activities at Cernavoda NPP impresses a complex interdependence after a polynomial of degree 6 ($R^2 = 0.9016$), as stated by correlating all records for the period (2009–2019). There is a direct interconnection between the total amount of precipitation in a year, higher temperature which favorizes the evaporation, and the average level of tritium – which means that there are other routes of penetration of this radionuclide through humidity of the air and rainfall, apart from the releases from the nuclear power plant located within a radius of 32 km [4].

All these preliminary conclusions and observations show that a systematic analysis using a second independent searching method of possible interdependencies over a certain time interval between these locations become justified.

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