ENVIRONMENTAL ASPECTS OF TRITIUM AROUND THE VINČA INSTITUTE OF NUCLEAR SCIENCES

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ABSTRACT

An overview of environmental distribution of tritium around the Institute of Nuclear Sciences Vinča during the period 1988-1994 is presented. Temporal and local variations of the specific tritium variations in precipitation (Usek, Zeleno Brdo), river waters (the Danube, the Sava and Mlaka Creek) as well as atmospheric water vapor are given. Estimates based on precipitation measurements have shown that 6.3 TBq of tritium activity should be released annually into the atmosphere from the Vinča Institute of Nuclear Sciences.

INTRODUCTION

Tritium is released from man-made source to the environment could be in the form of tritiated hydrogen gas (HT) and gaseous compounds, tritiated water (HTO) and aerosols of solid compounds. Transfer of tritiated water from the atmosphere to the surface of the earth occurs mainly by precipitation, but also by vapor exchange. Precipitation samples are of special interest because their tritium concentration together with the amount of precipitation governs the tritium activity fed into the soil and, consequently, have an impact on all the other media.

Due to possible pollution of the environment around the Vinča Institute of Nuclear Sciences with tritium originating from nuclear installations (the heavy water research reactor 6MW(th), RA, and the heavy water critical assembly reactor 1MW(th), RB), it is important to investigate the contribution of artificially produced tritium to surface waters and air in this region. For this reason a systematic study has been carried out since 1988. The purpose of this paper was to determine the distribution and level of tritium in the environment around vicinity of the Institute. Collected data should lead to a better understanding of tritium movement in the environment.

SAMPLING AND METHODS

Composite monthly precipitation samples were collected at three locations; the Vinča Institute of Nuclear Sciences (VIN), Usek, and the Meteorological Station Belgrade, Zeleno Brdo (ZB), (44°47'N, 20°32'E, altitude 243.2 m asl). The latter location is chosen at a distance of 7 km from the Institute in the main southeast wind direction as a reference site, and allows us to examine the behavior of the contribution of naturally formed tritium. The second off-side location, Usek is selected at the distance of 2.5 km from the Institute in the same wind direction. Precipitation was sampled by conventional rain gauge.

Tritium concentration in monthly composite samples (20-25 times per month) of river waters was measured at Belgrade in the Danube and the Sava at 1145 km and 6 km from their confluence, respectively. We, also, have checked the water of Mlaka Creek, that flows through

the Institute along the reactor boundary.

Environmental monitoring was extended to the air in 1993. Both HTO and HT tritium content in atmospheric water vapor is determined using a collecting unit, which is based on quantitative absorption/desorption on molecular sieve [1]. Sampling is continuous with an one-week integration period, during which 5-13 m^3 of air are passed through the collector.

The activity was measured by liquid scintillation counter (1219 Rack Beta Spectral) and in cases of precipitation after isotopic enrichment by electrolysis. Tritium activity was expressed in Bq/l water or Bq/m^3 air. Statistical errors of the measurements were $\pm 5\%$ for the concentrations above 2 Bq/l and up to $\pm 20\%$ below that value. Data on the amount of precipitation from the Federal Hydrometeorological Institute were used to calculate annual weighted mean concentrations and load in precipitation.

RESULTS AND DISCUSSION

The average weighted annual values for tritium concentrations in precipitation at studied locations, together with some IAEA/WMO network stations (Vienna, Zagreb, Budapest), over the period 1976-1994 are presented in Fig. 1. The tritium concentrations at Zeleno Brdo showed the decreasing time trend from 12.2 Bq/l (1978) to 3.2 Bq/l (1989). Year-to-year tritium monthly composite precipitation concentrations scatter of no more than ± 50 % from annual means. Concentrations observed at the reference location ZB are in agreement with current global levels measured at Vienna, Zagreb and Budapest [2, 3]. To the contrary, tritium content in precipitation at VIN exhibits in erratic time pattern with monthly values between 14.0 Bq/l (March 1993) and 59.4 Bq/l (August 1992) reaching the maximum annual value of 35.4 Bq/l in 1992. Much of the excess tritium comes in pulses and completely masks the normal seasonal variations. There is a decrease in specific activity with increase of source distance. Generally, concentrations at the off-site location Usek follow the pattern for VIN with the average annual value of 8.8 ± 3 Bq/l that is close to one at the reference site ZB (6.5 ± 3 Bq/l) suggesting a rapid diffusion around of the center of the discharge area.

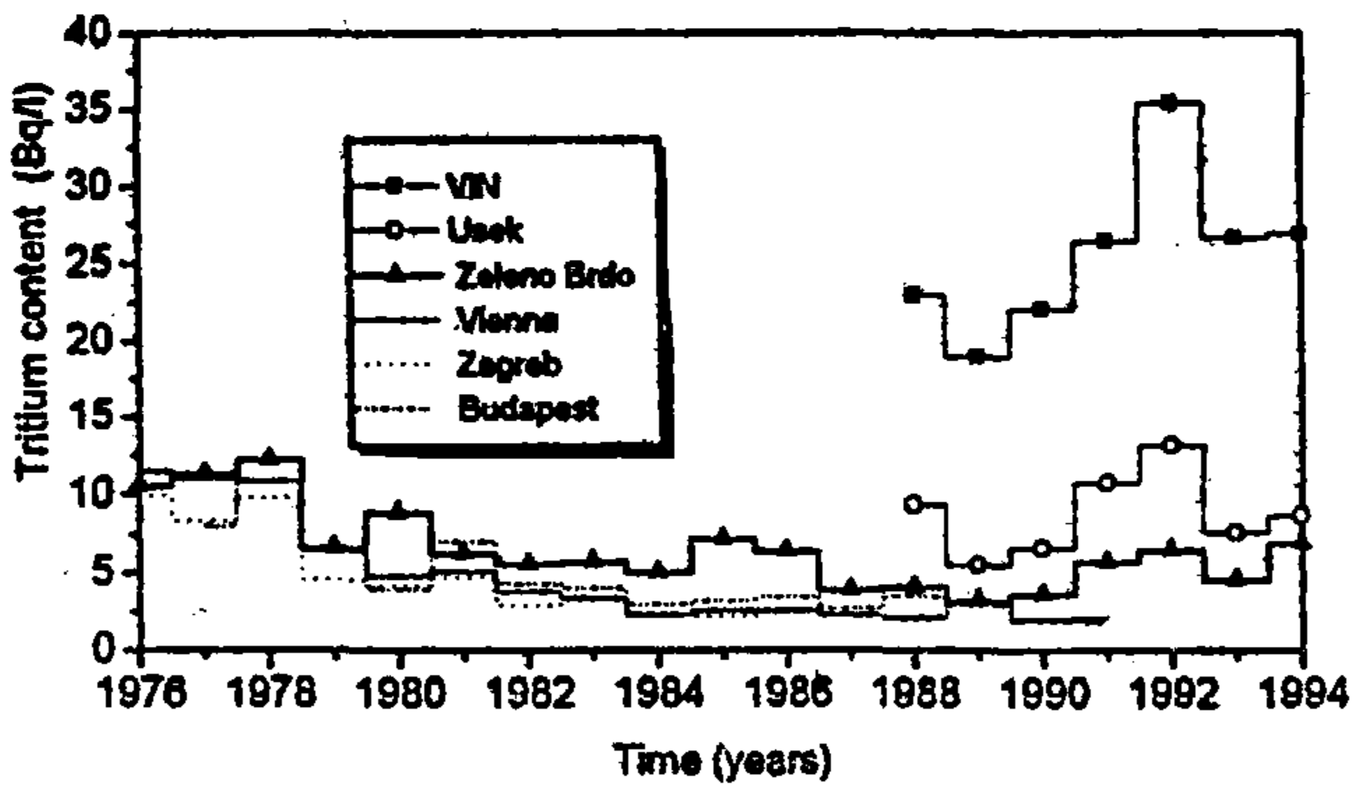

Fig.1. Annual weighted means of tritium content in precipitation at our locations together with some IAEA/WMO network stations

Data on monthly tritium concentrations in the Sava were from 1.5 to 8.1 Bq/l in April 1989 and June 1992, respectively (Fig. 2). At the same time obtained values for the Danube were larger between 3.7 Bq/l (April 1990) and 35.4 Bq/l (December 1992). Sometimes, irregular, extraordinary high values of unknown origin (12.5-35.4 Bq/l) were recorded in the Danube. Consequently, over the period 1988-1994 tritium abundance in the Danube was about 20% grater in average than in the Sava, excluding irregular high values. Mlaka Creek showed

remarkable fluctuations in tritium content from 38.2 Bq/l (May 1990) to 133.8 Bq/l (June 1992). Markedly increased tritium content not comparable with the values recorded in average annual precipitation (25±6 B/l) at VIN can be attributed to some influence by liquid discharge through shallow water-tables.

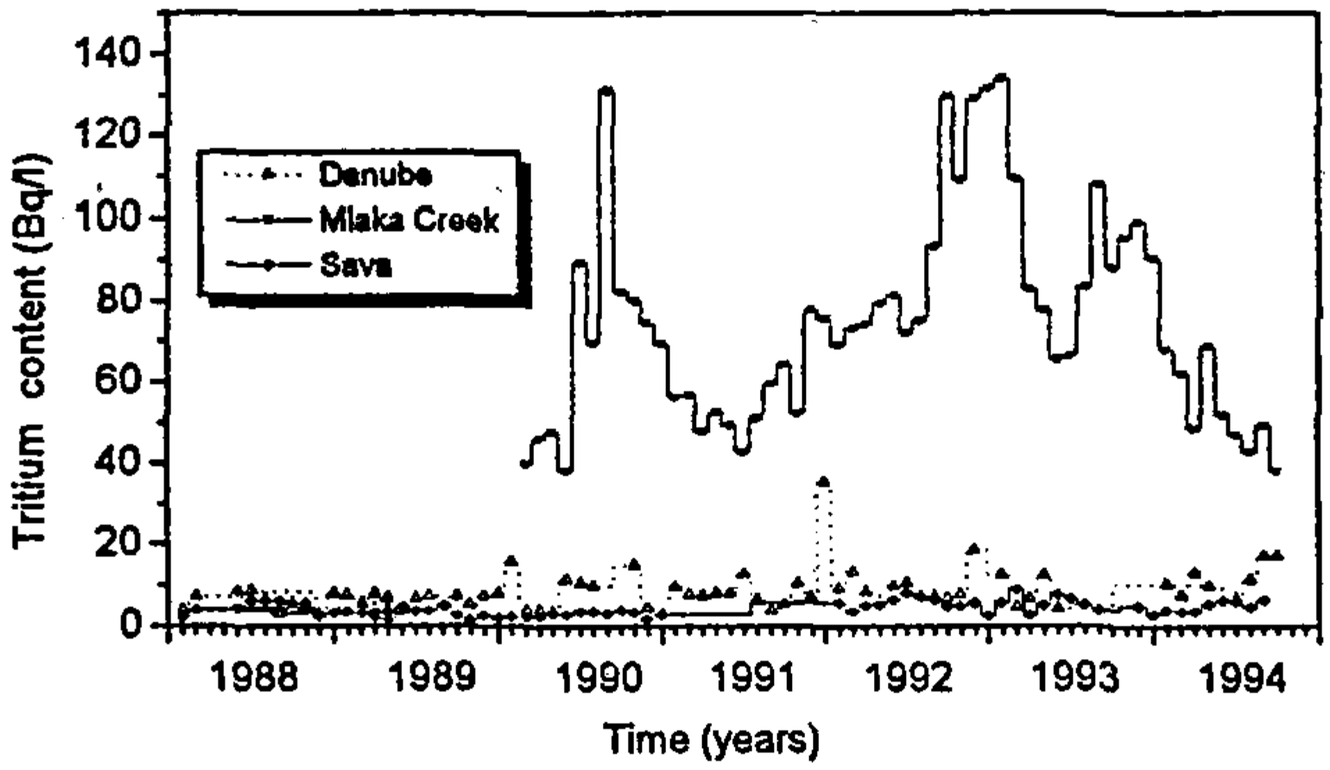

Fig. 2.Tritium content in river waters

The results on atmospheric tritium concentrations at ground level at VIN during 1993 and 1994 are plotted in Fig. 3. Data are in the range 1.5-67.1 Bq/m³ and 0.10-1.46 Bq/m³ for HTO and HT respectively. A pronounced summer maximum in August is noticeable for both tritium chemical forms. Values for HT (annual means 0.47 Bq/m³) and HTO (annual means 20 Bq/m³) and their ratio HT/HTO=0.03 indicate that HT is being emitted by the nuclear facilities. Concentrations are higher than the background level observed for general environment in Japan (HTO=0.02 Bq/m³ and HT=0.05 Bq/m³) [4]. Tritium concentration in air were comparable with those measured around the nuclear plant boundary up to 6 km distance [5].

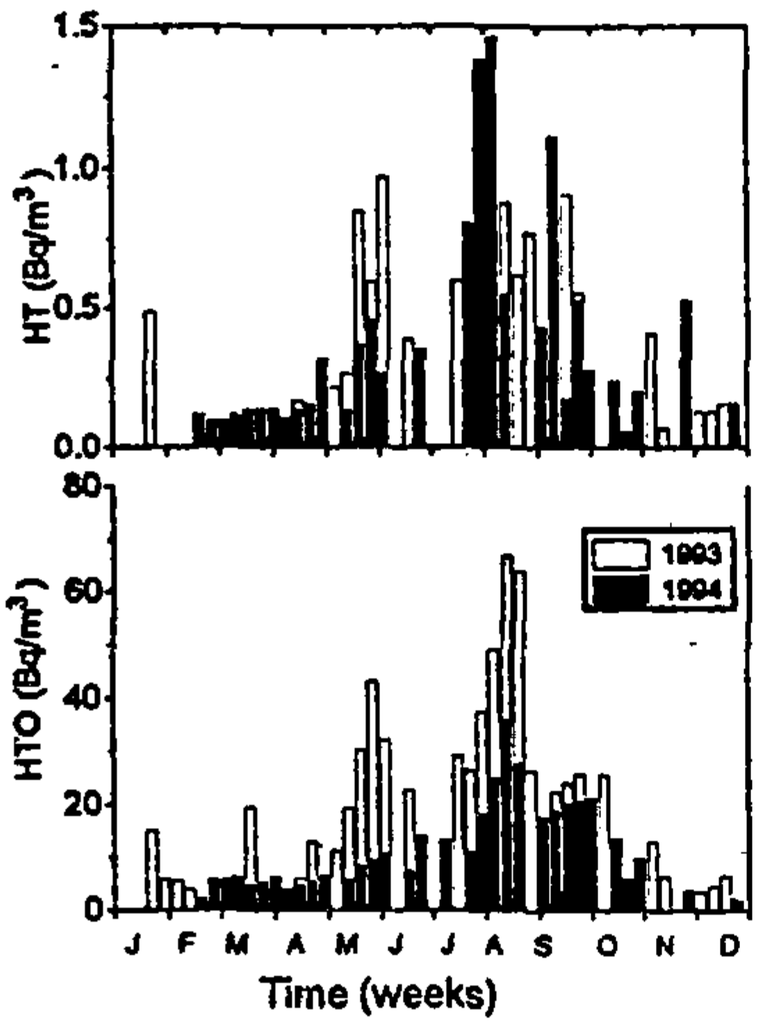

Fig. 3. Ground-level HT/HTO concentrations at the Vinča Institute of Nuclear Sciences

The mean values of surface load (tritium concentration in precipitation x amount of precipitation) at VIN clearly exceeded the values for more distant location ZB, because of the tritium emissions into the atmosphere from the Institute. Establishing the difference in surface load values found on the site of and outside VIN gives a mean surface deposition of about 1

kBq/m^2, i.e. 1 GBq/km^2) yearly from releases. A substantial part of the surface load is caused by fall-out. If the calculations are based on a mean surface load of 1 GBq/km^2 yearly from releases, a total annual load of about 3.14 GBq is obtained for an area of 1 km radius around VIN. Taking in account a found portion of $5x10^{-4}$ [6] of the tritium activity deposited by precipitation in the area of 1 km radius around the Karlsruhe Nuclear Research Center, a value of 6.3 TBq yearly is estimated to be released into the atmosphere from the Vinča Institute of Nuclear Sciences.

In order to make a rough evaluation of currently tritium releases some potential emitters are considered. The RB reactor with present equilibrium tritium concentration in the moderator (D_2O) of 1.85 MBq/l gives a total tritium activity of 11 GBq. The experiment in December 1994 demonstrated a rising of additional tritium make-up D_2O activity for 132 kBq caused by the total generated fission energy of 41 Wh. In addition, while RA used to be under operating conditions, its heavy water moderator and coolant system had a total activity of 540 TBq [7]. During standard reloading the measured tritium concentration of discharged air to the environment in the reactor hall was 0.74 MBq/m^3. Although RA is not operating unit, some regular facility examinations as the fuel inspection and vacuum distillation of D_2O still have been performed, leading to tritium release. Therefore, the preceding results suggest that the release of the remainder of reactor-generated tritium requires more sensitive and more accurate measurements.

CONCLUSIONS

The tritium measurements in precipitation for a time span of 19 years at the reference location ZB are consistent with general background levels. The apparent ZB station scatter differs by 30% from values found at the monitoring stations Vienna, Zagreb, and Budapest. During the period of observation (1988-1994) discharge of HTO vapor to the atmosphere has resulted in rising of average weighted annual tritium concentrations in precipitation to 25 ± 6 B/l at the Vinča Institute of Nuclear Sciences. Significantly higher tritium content in Mlaka Creek than other rivers indicates existing of liquid besides airborne effluents. The obtained results support the hypothesis that VIN is a regional source of tritium with several chronic emitters. Their contribution and the form of the release are necessary to be further investigated.

REFERENCES

[1] Miljević, N., Šipka, V., Gršić, Z., "Time variation of D, ^{18}O and ^{3}H in atmospheric water vapor and precipitation near the Institute of Nuclear Sciences Vinča", Second International Symposium and Exhibition of Environmental Contamination in Central and Eastern Europe, Budapest'94, Budapest, September 20-23, 1994, Symposium Program, p. 127.

[2] IAEA, Statistical Treatment of Data on Environmental Isotopes in Precipitation, Tech. rep. ser. no. 331, Vienna, 1992.

[3] IAEA, Environmental Isotope Data No. 10: World Survey of Isotope Concentration in Precipitation, Tech. rep. ser. no. 371, Vienna, 1994.

[4] Momosshima, N., Okai T., Inoue, M., and Takashima, Y., "Tritium monitoring around a nuclear power station in normal operation", Appl. Radiat. Isot. 38 (4), 263 (1987).

[5] Brown, R.M., "HT and HTO in the environment at Chalk River", Fusion Technology Vol.8, pp. 2539-2543, 1985.

[6] Konig, L.A., "Impact on the environment of tritium releases from the Karlsruhe Nuclear Research Center" in: Behavior of tritium in the environment, IAEA-SM-232/80, IAEA, Vienna (1979), p. 591.

[7] Ninković, M., Matić-Vukmirović Z., and Hadžišehović M., "Elements of tritium detection in air for heavy-water nuclear reactors", IBK report 528, Vinča, 1967.