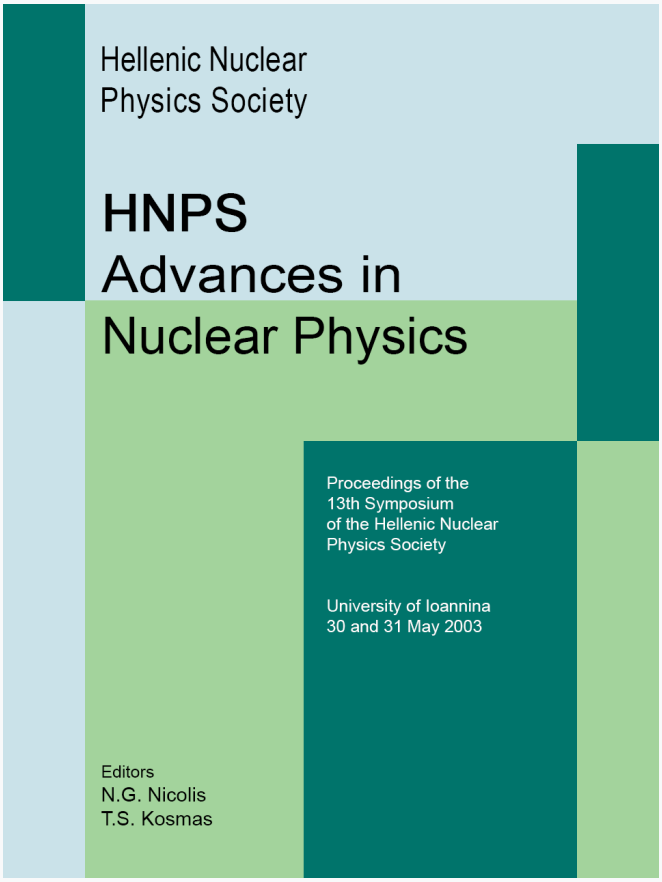
HNPS Advances in Nuclear Physics

Vol 12 (2003)

HNPS2003



Correlation of Tritium Concentration in Rainwater with Cosmic Rays: Preliminary Results

K. Stamoulis, K. G. Ioannides, A. Vlachogianni, c. A. Papachristodoulou

doi: 10.12681/hnps.3348

HNPS Advances in Nuclear Physics vol. 12, pp. 99-104 (2003)
HNPS2003
doi:10.12681/hnps.3348

Correlation of tritium concentration in rainwater with cosmic rays: preliminary results.

K. Stamoulis[1], K. Ioannides[1,2], A. Vlachogianni[3],
C. Papachristodoulou[4]

[1] *Radiochronology Center, University of Ioannina*
[2] *Nuclear Physics Lab., University of Ioannina*
[3] *Physics Department, University of Ioannina*
[4] *XRF Unit, University of Ioannina*

Abstract

The Radiochronology Center has been established in 1999 as a member of the Horizontal Laboratories network of the University of Ioannina. Recently it has added to its activities the development of a tritium measurements laboratory as part of the Radiochronology Center. The laboratory is equipped with a super low level background liquid scintillation counter, which is capable to measure very low concentrations of tritium for applications in radioprotection, dating and hydrology.
For the aims of the present continuing study rainwater samples are collected monthly. Each sample is distilled and then it is passed through an electrolysis process to increase the tritium concentration. Five mL of the enriched sample are mixed with 15 mL of a scintillation cocktail, specially designed for tritium measurements and its beta activity is measured for 200 min. The LLD of tritium in the samples is estimated 3 Bq/L or 27 TU. Our preliminary results show that, during the measuring period tritium concentration increased with time. The tritium values are correlated with the cosmic ray neutron flux data at ground level, available for Greece in the same period. The measured tritium concentrations in rainwater, which range from 36 ± 8 to 64 ± 8 TU, may be used for local hydrology studies.

1. Introduction

Establishment of the Tritium Laboratory

The Radiochronology Center was established in 1999 in the frame of the development of Horizontal Laboratories Network of the University of Ioannina. Since September 2002 the Radiochronology Center acquired a specially designed Laboratory capable to perform various radiation measurements. Among them, tritium and radium-radon measurements in various samples are included. The Center is equipped with a liquid scintillation analyzer from Packard (Tricarb 3170TR/SL). Various types of samples, water, soil and biological samples can be processed in order to pre-concentrate the radioactive element and transform the sample to the appropriate form for liquid scintillation counting. The Radiochronology Center participates in programs of the International Atomic Energy Agency (IAEA) and the Greek Atomic Energy Commission.

Tritium measurements in rain water samples

In the present study we collected rainwater samples to measure the tritium activity concentration. Tritium activity levels in rainwater are useful for hydrological studies. The Radiochronology Center is the first Laboratory in Greece to perform routinely tritium activity measurements.

Tritium (^3H) in the water cycle

Tritium (^3H) is a radioactive isotope of hydrogen and is produced in the upper layers of the atmosphere by the reaction of neutrons produced by cosmic rays and natural isotope of nitrogen ^{14}N :

$$n + {}^{14}N \rightarrow {}^{3}H + {}^{12}C$$

Tritium is oxidized by oxygen and is transformed to water. Thus following the route of the water from atmosphere to the ground it enters the water cycle. As it is a radioactive isotope (half life 12,3 years), it decays to ^3He giving an electron with E_{max} = 18,6 keV:

$$^{3}H \rightarrow {}^{3}He + e^{-}$$

Tritium is also produced in nuclear reactors, entering the environment by periodical release of the coolant water of the reactors. A major source of tritium pollution was hydrogen bomb detonations in the 60ies. This source raised tritium activity in the environment by a factor of 1000. Tritium concentration in water is measured in Bq/L or in Tritium Units (TU):

$$1 \text{ Tritium Unit (1TU)} = 0.118 \text{ Bq/L} = 1\ {}^{3}H/\ 10^{18}\ {}^{1}H$$

Before the tritium pollution the normal activity of tritium in rainwater was about 5 TU. During hydrogen bomb detonations the activity raised up to 6000 TU while after the Nuclear Tests Ban Treaty it continued decreasing and nowadays is about 10-100 TU (Figure 1).

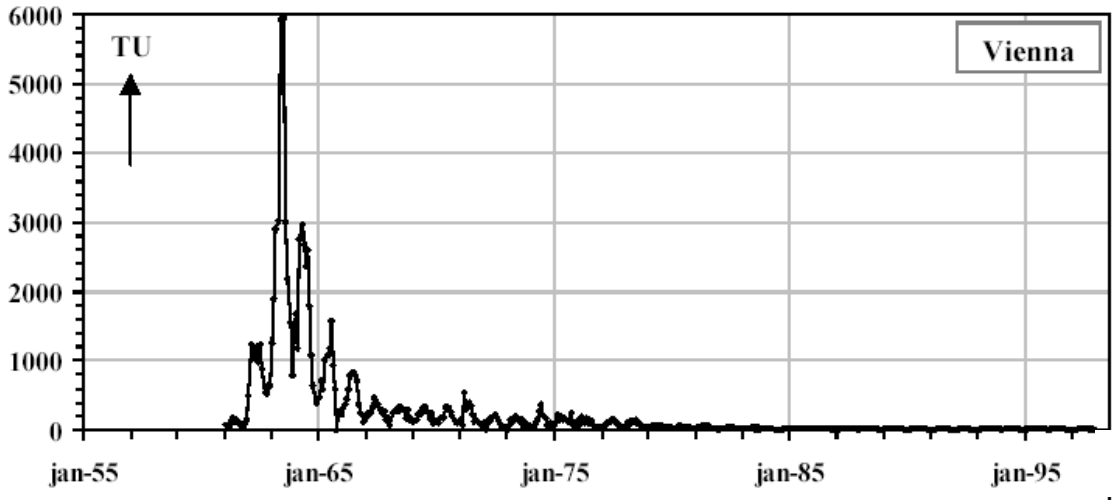


Figure 1 Tritium concentration in rainwater in Vienna (Mook G.W. 2000)

2. Materials and Methods

Liquid scintillation analyzer

For the liquid scintillation measurements (LSC) we used the Packard Tri-Carb 3170TR/SL equipped with various features suitable for powerful liquid scintillation analysis. It performs super low background counting for ^3H. Measuring 20ml of Ultima Gold LLT cocktail for 500 min produces 1.8 cpm background (spectral window 0-18,6 keV), which corresponds to a minimum detectable limit of 3 Bq/L or 27 TU for 5ml water sample and 15ml Ultima Gold LLT.

The analyzer is equipped with a sample changer with a capacity of up to 600 samples. Samples can be measured following different measuring protocols and assays. Using different protocols and assays samples can be measured in super low or normal count mode simultaneously, assaying the activity of one, two or three different radioactive alpha or beta emitting isotopes.
The counter's detecting arrangement consists of two photo-multipliers that surround the sample container and a specially designed BGO crystal serving as guard detector. The photo-multipliers are shielded with lead for lowering the cosmic rays background.
The radioactive material of the sample emits alpha, beta or gamma radiation that is absorbed by the scintillation molecules. These molecules are excited and emit photons, which are collected and amplified by the photo-multipliers. The number of emitted photons is proportional to the energy of the incident radiation. The electronic signal is amplified, digitized and counted according to the energy of the radiation that produced it.

Quenching effects
During the interaction of the solvent and the scintillator the excitation energy may be absorbed and the number of the emitted photons from the scintillator will be less than it should be according to the energy of the incident radiation. This reducing effect on the number of produced photons is called chemical quench. Also if the sample-cocktail solution is not clear many photons are absorbed before they reach the photo-multipliers. This effect is called color quench. Both these quenching effects decrease the efficiency of the detection system.
In order to calculate the quenching effect, various spectral indexes are used, which depend on the quench level of the sample. One of these indexes is the transformed Spectral Index of External Standard (tSIE), which is independent of various other factors that affect liquid scintillation counting of a sample (wall effect and cocktail volume). The Tri-Carb analyzer can calculate this index and is possible to determine the efficiency for the specific sample using the correlation between tSIE and efficiency of the counter.

Tritium enrichment of water samples
There are water samples that contain tritium in such activities that are below the detection limit. In this case the samples are enriched by electrolysis. This enrichment is based on the fact that heavier isotopes (deuterium and tritium) of hydrogen are not electrochemically separated as readily as hydrogen does.
80 ml of the water sample and 0.7 ml of H_2SO_4 are added in a glass tube and 3 A current is passed through the tube. The electrolysis tube is immersed in a water bath to maintain low temperature (less than 10 C). The Radiochronology Center is capable in electrolyzing 10 such samples in the same time.
The enrichment of the sample depends on the ratio of the initial to the final volume of the sample and the enrichment factor R. If T_f is the final tritium activity, T_i is the initial tritium activity and V_f and V_i are the final and initial volumes of the sample the initial tritium concentration of the sample is given by the equation (M. Kakiuchi, 1991)

$$T_i = T_f R \frac{V_f}{V_i}$$

The enrichment factor R is determined experimentally and is given by the equation (Hartley, 1972)

$$R = \{b-(b-1)e^{[\frac{(1-\frac{V_i}{V_f})}{b}]}\}\frac{V_f}{V_i}$$

where b is a parameter depending on the type of the electrolytic cell, the electrodes and the current density. This parameter was estimated experimentally and it was found b = 10,4.

3. Results and discussion

Tritium concentration in rainwater

The results of tritium concentration in rainwater samples are presented in the following Figure 2.

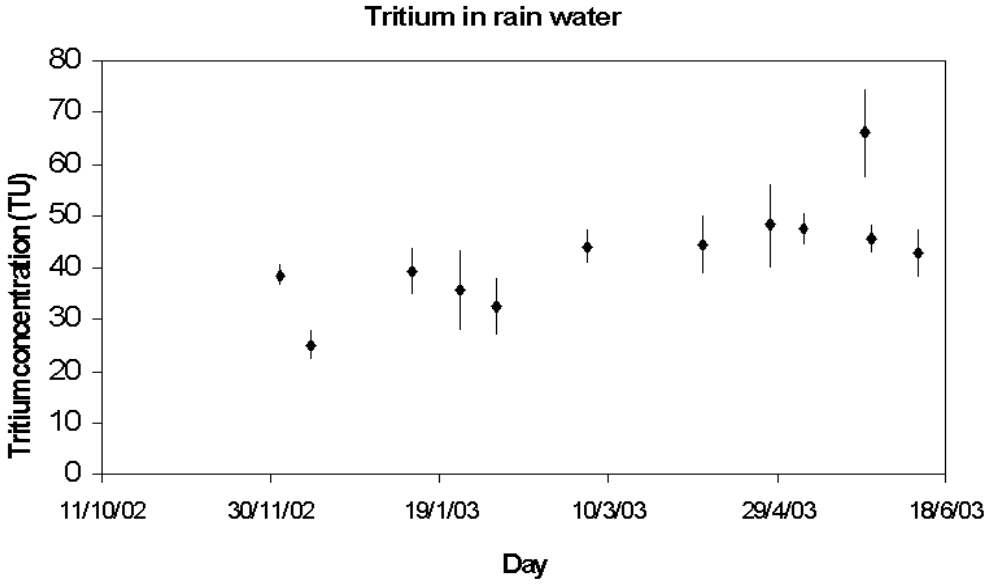


Figure 2 Tritium concentrations in rainwater samples for the period November 2002-May 2003

Tritium concentration showed an increase with time from about 35 TU or 4.3 Bq/L to 60 TU or 7 Bq/L, in a period of six months. This rather slow increase in this period of the year (winter-spring) is in accordance with observations of other studies (Mook G.W. 2000). Also these levels of tritium concentration in rainwater are in accordance with other published literature (Eriksson E. 1983; Garcia Lopez, 1994; The Scottish Environment Statistics, 1998; Blagojevich, 2003)

Temporal correlation of tritium concentration and cosmic ray neutron flux

Neutron flux collected at Institute of Terrestrial Magnetism, Ionosphere and Radiowave Propagation, in Moscow (IZMIRAN), for the period August 2001-May 2003 are presented in Figure 3. From September 2002 to May 2003 there is an increasing trend of the neutron flux. Because of the fact that tritium is produced in the upper atmosphere by the reaction of neutrons with the stable isotope of nitrogen ^{14}N, we attempted to correlate

the mean monthly neutron flux with the mean monthly tritium concentrations in rain water. The time dependence of the correlation can be calculated by using the equation

$$T(t)=aN_{mo}(t-t_d) + b$$

Where T(t) is the mean tritium concentration in month t, $N_{mo}(t-t_d)$ is the mean neutron flux in month $t-t_d$ and t_d is a time delay parameter (in months) estimated by fitting the data with the above equation. Time delay parameter is introduced because of the fact that tritium produced in the atmosphere, remains there before it comes down to the ground with the precipitation for a time period which is estimated by this time delay parameter. Figure 4 presents the correlation factor R^2 as function of time delay parameter t_d. The best correlation is obtained with t_d = 3 months delay and coefficient R^2 = 0,74.

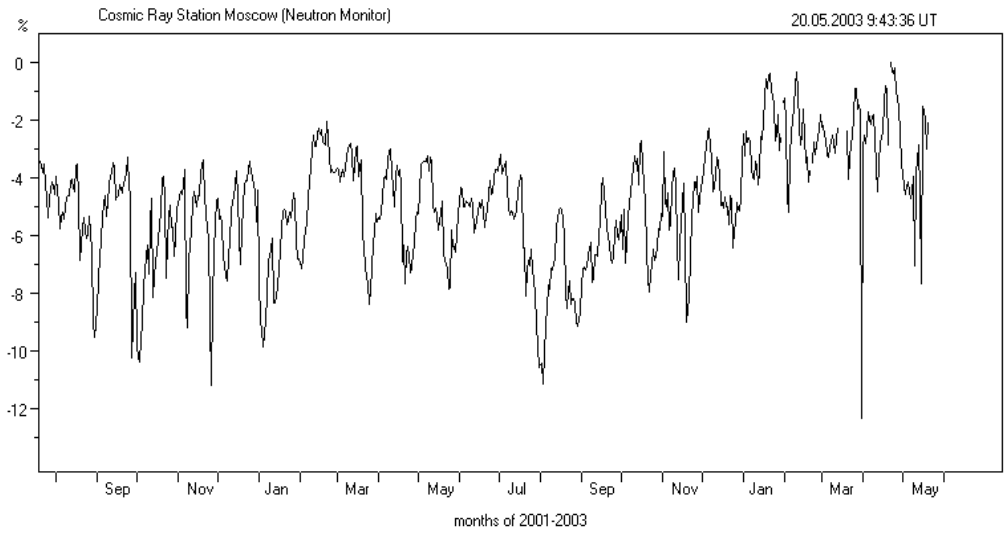


Figure 3 Neutron flux collected at Moscow, Institute of Terrestrial Magnetism, Ionosphere and Radiowave Propagation IZMIRAN.

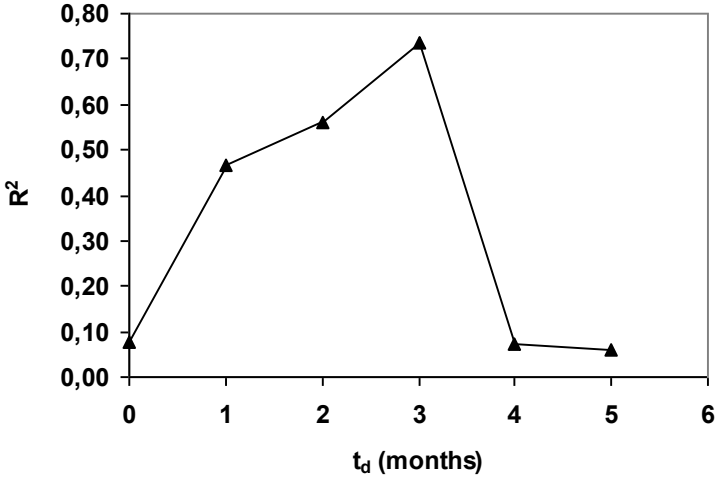


Figure 4 Relation between R^2 and time delay parameter t_d (months)

References

Blagojevich R.R ,Use of Tritium in Assessing Aquifer Vulnerability, 2003 http://www.epa.state.il.us/water/tritium.html

Eriksson E. (1983) Stable isotopes and tritium in precipitation. In Guidebook on nuclear techniques in hydrology, Vol. 91, 19–27. Technical Reports Series I.A.E.A.

Garcia Lopez, S., Cardenal, J., Cruz-Sanjulián, J. J. and Benavente, J. Natural and anthropogenic sources of tritium in carbonate aquifers to the south of Sierra Nevada (Spain). *J. Environm. Hydrology* **2**, pp. 2–31,1994.

Hartley P.E. Design and performance of tritium measurement systems using electrolytic enrichment. Nuclear instruments and methods, 100: 229-235, 1972.

Kakiucki M., Tanaka K., Kigoshi K. Natural Tritium Measurements : Determination of the electrolytic enrichment factor of tritium. Applied Radiation and Isotopes 42(8): 741-748, 1991.

Mook G.W. Environmental isotopes in the hydrological cycle, Principals and Applications. Vienna 2000.

The Scottish Environment Statistics, 1998
http://www.scotland.gov.uk/stats/envonline/_data/RADIOcaesiumandtritium.xls