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Study of the comparative dynamics of the incorporation of tissue free-water tritium (TFWT) in bulrushes (*Typha latifolia*) and carp (*Cyprinus carpio*) in the Almaraz nuclear power plant cooling reservoir

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ABSTRACT

The Almaraz nuclear power plant (Spain) uses the water of Arrocampo reservoir for cooling, and consequently raises the radioactive levels of the aquatic ecosystem of this reservoir. From July 2002 to June 2005, monthly samples of surface water, bulrushes (*Typha latifolia*) and carp (*Cyprinus carpio*) were collected from this reservoir. They were analyzed to determine the temporal evolution of the levels of ^{3}H in surface water and of its transfer from the surface water to free-water in the tissues (TFWT) of the aforementioned two organisms. The tritium levels in the surface water oscillate with a biannual period, with their values in the study period ranging between 53 and 433 Bq/L. The incorporation of tritium to bulrushes and carp was fairly similar, the respective mean concentration factors being 0.74 and 0.8 (unitless, as Bq/L tissue water per Bq/L reservoir water). The temporal evolution of the levels fairly closely followed that observed for the surface water tritium, although detailed analysis showed the dominant periodicity for the bulrushes to be annual. This difference reflects the influence on the incorporation of tritium to bulrushes of diverse environmental and metabolic factors, especially evapotranspiration and the seasonal growth of this plant.

1. Introduction

In order to guarantee the existence of a sufficient volume of water to cool the Almaraz nuclear power plant (henceforth ANPP) during its operation, the artificial Arrocampo reservoir ($V = 3.5 \times 10^7$ m^3) was constructed in its proximity on the right bank of the River Tagus. All the liquid effluent from the ANPP is fed into this reservoir, whose volume is maintained practically constant by inputs of water from the River Tagus at the height of the Torrejón-Tagus reservoir ($V = 1.6 \times 10^8$ m^3). This input is greater during the summer than during the winter to compensate the greater evaporation losses and to reduce the temperature of the water sufficiently for its use as coolant. For this same reason, water is forced to circulate around the length of the reservoir to favour heat exchange with the atmosphere. The mechanisms of input and recirculation of the water, together with the activities evacuated by the ANPP itself, determine the temporal evolution of the artificial radionuclide activity levels in the reservoir. This temporal evolution has been quantified in previous studies for diverse radioactive cations (Baeza et al., 1991), and also characterized and modeled with respect to tritium (Baeza et al., 1997). This latter study managed to satisfactorily describe the dynamics of the temporal evolution of the tritium content of the Arrocampo water for the period 1994–1997. The behaviour is clearly seasonal in that the greatest activity levels are recorded in the winter, and the lowest during the hottest months.

We have also characterized the spatio-temporal behaviour of the tritium levels in the water of diverse sections of the Tagus river more or less distant from the ANPP, and hence affected to a greater or lesser degree by the releases of tritium most of which are from the ANPP (Baeza et al., 2001). Again, we observed that the temporal evolution of tritium in the water downstream from the ANPP has a markedly seasonal behaviour (Baeza et al., 2004). This seasonality is modified with respect to that observed in the Arrocampo reservoir because of the forced transit of water along this section of the Tagus, which consists of a quasi-connected succession of dams whose management even leads in certain situations to the water flowing contrary to its natural direction. We modeled the temporal evolution of tritium for some of these ecosystems, obtaining satisfactory results (Baeza et al., 2002a, 2006). In particular, these models were applied to the hydrodynamic distribution of tritium,

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in the river basin – the Tagus – with a very high variation in flow regime (Baeza et al., 2002b).

Nevertheless, we had yet to consider the transfer of the levels of tritium present in the water of Arrocampo to other characteristic links in the trophic chain of that ecosystem. Studies of this type are particularly interesting because once tritiated water is present in an ecosystem it is initially taken up by living organisms and then partially eliminated, the biological half-life for humans, for instance, being some 9.7 days (NCRP, 1979). Also, during its incorporation, diverse metabolic reactions, such as those that occur in photosynthesis, transform the tissue free-water tritium (TFWT) into organically bound tritium (OBT) (Diabaté and Strack, 1990, 1993, 1997). The present work analyzes the transfer of tritium from surface water to two different species that are abundant in the Arrocampo ecosystem – carp (*Cyprinus carpio*) and bulrushes (*Typha latifolia*). The carp is an edible freshwater fish whose analysis is interesting from the point of view of radiological protection. The interest in the bulrush is because of its frequent use as a bio-indicator in radiological environmental monitoring programs of lake and river ecosystems, in addition to its intrinsic interest in determining the dynamics of the transfer of radionuclides in these ecosystems. The primary objective of the present study was therefore to determine the tritium levels in the tissue free-water (TFWT) of a species of aquatic plant – the bulrush – and of a species of fish – carp – that are common in the Arrocampo reservoir. From these levels, we shall attempt to characterize comparatively their different temporal evolution, and evaluate the environmental and metabolic factors that influence the dynamics of the uptake of water to these two freshwater species.

2. Experimental and methods

2.1. Analysis of the tritium activity in water, bulrushes, and carp

The samples analyzed in the present work were collected monthly in the Arrocampo reservoir (longitude = 5°40′27.98″W, latitude = 39°50′20.90″N USO 30) during the period July 2002–June 2005. This reservoir provides the water flow necessary to cool the ANPP during its operation. This nuclear plant consists of two 930 MWe PWR-type units, and is located on the right bank of the River Tagus, in the Region of Extremadura, 180 km west–southwest of Madrid, the capital of Spain. Initially, the sampling was only of surface water and bulrushes. This was later extended to include collection of carp, of which only the muscle was used to determine the tritium levels. Finally, with the same monthly frequency, an *in situ* study was made of the growth of the bulrushes in the reservoir (details below). For the bulrush samples, only the aerial parts from about 3 cm above the water level were collected. The amounts of each sample of water, bulrushes, and carp muscle used in the analyses were 1 L, 500 g-fresh, and 100 g-fresh, respectively. All samples were stored frozen immediately after collection until assay.

Given the physico-chemical characteristics of the water samples, the only pre-treatment necessary was filtration (Baeza et al., 1997) before the corresponding determination of the activity of ^{3}H by liquid scintillation spectrometry. For the carp and bulrush samples, it was necessary to extract the free-water to determine the tissue free-water tritium (TFWT) by means of a pre-treatment consisting of an extractive Dean–Stark type distillation with cyclohexane (LIBRA, 2002). This extraction procedure is less troublesome to apply than other methods of extracting of the free-water fraction, such as liophilization or thermal desorption *in vacuo* (Failor et al., 1998; Choi et al., 2002), since the equipment used is much simpler. One starts with an appropriate aliquot of the sample – 50 g-fresh for both the bulrushes and the carp muscle. This is triturated and mixed with cyclohexane, followed by distillation for 12 h. Under these conditions, the water forms an azeotrope with the cyclohexane in the vapour state, allowing the free-water to be extracted from the sample. Once the still miscible steam has condensed, the resulting cold phases – aqueous and organic – are immiscible, and therefore easily separable. The former is conserved under refrigeration until continuing with the process of sample preparation. This aqueous phase obtained from the bulrushes or the carp, as with the filtered surface water, is later assayed by liquid scintillation spectrometry, with no need for prior electrolytic concentration (Baeza et al., 1999) or any other treatment. Indeed, the values of quenching obtained for the free-water extracted from the biological samples are similar to those of the surface water of the Arrocampo reservoir. Using the Optiphase Hisafe 3 cocktail in the proportion (aqueous sample/scintillator) 8/12, with counting times of 800 min, in an LKB Quantulus 1220 device equipped with a special background reduction system, the minimum detectable activity in the determination of ^{3}H present in all three types of sample was 1.4 Bq/L.

For the biological samples, one must also taken into account what fraction of the water is recovered by the Dean–Stark distillation method. We determined these to be $(72 \pm 3$ (S.D.))% and $(80 \pm 2$ (S.D.))% for the bulrushes and the carp, respectively.

2.2. Study of the growth of the bulrushes

During the last months of the sampling period, we simultaneously determined the average growth of the bulrushes of the Arrocampo reservoir, studying plants in the same zone as that of the monthly sampling. To this end, a sufficiently stable metal reference platform with a vertical pole was constructed, and fixed permanently to the bed of the reservoir during the period December 2004–June 2005, very close to where the bulrushes were collected. Monthly determinations of the mean height of the bulrushes were made from 80 individual measurements of this parameter for randomly chosen individuals, distributed homogeneously around the platform.

2.3. Data analysis

For the analysis and interpretation of the measured tritium activity levels, in addition to the basic statistical treatment, we applied methods of time series analysis (smoothing, test of randomness, and harmonic analysis). Data smoothing of a time series allows one to distinguish long-term from short-term variability. We used the simple moving average smoothing method, in which each datum of the original series is replaced by the average of l data to the left, l data to the right, and the central datum. The number of data used in the average is thus $n = 2l + 1$, which is the length of the moving average (Diggle, 1990). To test whether the time series were indeed random series of values, we used the Box–Pierce test which is based on the analysis of the sum of the squares of the first autocorrelations (Diggle, 1990).

We performed the harmonic analysis by means of obtaining the periodogram and the integrated periodogram of the time series. The periodogram is based on a Fourier analysis of the data. To this end, one assumes that the data time series $A(^{3}H,t)$ is formed by the superposition of sinusoidal components of different frequencies. The intensity of the periodogram $I(f_i)$ is defined as:

$$I(f_i) = \frac{2}{N}\left\{\left[\sum_{t=1}^{t=N} A(^{3}\mathrm{H},t)\cos(2\pi f_i t)\right]^2 + \left[\sum_{t=1}^{t=N} A(^{3}\mathrm{H},t)\sin(2\pi f_i t)\right]^2\right.$$

with $i = 1, 2, 3, \ldots q$ where $q = (N-1)/2$ for N odd and $q = N/2$ for N even. The periodogram is the plot of $I(f_i)$ against f_i, where $f_i = i/N$ is the i-th harmonic of the fundamental frequency $1/N$, up to the Nyquist frequency of 0.5 cycles per sampling interval (which corresponds to the smallest identifiable wavelength of two samples). Since $I(f_i)$ is obtained by multiplying $A(^{3}H,t)$ by sine and cosine functions of the harmonic frequency, it will take on relative large values when this frequency coincides with a periodicity of this frequency occurring in $A(^{3}H,t)$. As a result, the periodogram maps out the spectral content of the series, indicating how its relative power varies over the range of frequencies between $f_i = 0$ and 0.5. The integrated periodogram is the normalized cumulative sum of the periodogram ordinates against the frequencies from 0.0 to 0.5, and serves to determine whether or not the spectral components of the periodogram are significant (Hewit, 1992).

3. Results

Table 1 lists the results for the ^{3}H concentrations found in the three types of sample, together with the bulrush-water and carp-water concentration factors (CF's, unitless, as Bq/L tissue water per Bq/L reservoir water) for that radionuclide. As can be seen the tritium levels in the water of the Arrocampo reservoir in the sampling period varied from a minimum of 53 Bq/L to a maximum of 433 Bq/L, with a mean of 131 Bq/L. The mean activity of tritium for the entire triennium was markedly greater in the surface water than in the bulrushes and the carp, with the respective means being $(131 \pm 79$ (S.D.)) Bq/L, $(96 \pm 68$ (S.D.)) Bq/L, and $(103 \pm 50$ (S.D.)) Bq/L. It is also noteworthy, however, that the two biological values are very similar to each other, although it must be borne in mind that, because of the design of the sampling, there were fewer monthly samples collected for the case of the carp. The mean CF's were normally less than unity for both bulrushes and carp – for the bulrushes varying between 0.306 and 1.27 with a mean of 0.74, and for the carp between 0.44 and 1.78 with a mean of 0.8.

The temporal variation of the tritium activities in the three sample types is shown in Fig. 1. For the case of the surface water, in