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RESEARCH ARTICLE

HOSPITAL RADIOACTIVE WASTE TREATMENT BY PHYTOREMEDIATION

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ABSTRACT

Nuclear medicine is a medical practice that uses low-level radioactive isotopes for diagnosis, treatment and therapy in oncology. However, inadequate management of radioactive waste is a problem that prompts the current authors to suggest more sustainable alternatives for final disposal of such waste. Phytoremediation is one of these sustainable methods, which is economical and efficient in managing radioactive waste from hospitals. Usually, hospital pretreatments require a lot of space for storage or special procedures to dispose of such waste in ways that are both optimal and legal, in order to avoid impacts on society and the environment. In the current research, we studied *Phragmites australis*, which is a bio accumulative plant for heavy metals and radiopharmaceuticals, such as iodine-131 at plant root level. The plant has shown that phytoremediation at 144 hrs for experimental unit (1) has an increase of absorption activity of 0.1258 MBq and the plants morphometry did not show any change.

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INTRODUCTION

Nuclear medicine is a medical practice that uses radioactive isotopes emitting gamma and beta rays of low intensity for diagnosis, treatment and therapy. Some diseases covered are: bone pain, evaluation of the musculoskeletal system, thyroid glands, infections in the urinary, cardiovascular, pulmonary, digestive and central nervous systems, and cancer treatment.

One consequence of the use of nuclear medicine is the separation and immobilisation of large volumes of waste containing radioactive isotopes from hospitals. This represents a serious difficulty in their storage and handling, because the high solubility of the waste in any environment can cause external radiation and radioactive contamination of people who are around and the environment itself (Palau et al., 2010). Management of radioactive waste can be inadequate and costly, which allows to suggest a more economic and sustainable alternative such as phytoremediation; this method uses plants or microorganisms to remediate contaminated areas. Some radioactive isotopes that have been studied, such as barium (Ba), actinium (Ac), radon (Rn), and rare elements

that can be found on Earth, showed that they are accumulated by plants such as *Japonica gleichenia*, *Dicranopteris dichotoma* and *Struthiopteris niponica*, during their growth and all of which identified as accumulators of the aforementioned isotopes (Chao and Chuang, 2011). The method of phytoremediation using a plant called *Phragmites australis* (*P. australis*), is widely known since it has been confirmed that it hyper accumulates metals, heavy metals and lixiviates. It is also used in artificial wetlands for the treatment of industrial discharges, demonstrating its bio accumulative efficiency at root level. Earlier studies demonstrated knowledge that *P. australis* accumulates chromium, copper and zinc on the stem and rhizomes, and nickel at foliar level (Bragato et al., 2009, 2006). In this research, the radioisotope iodine-131 (I-131) generated from residual waste in the area of oncology of a hospital located in the State of Mexico was tested at the root level of *P. australis* and its accumulation was measured and analysed. This technique resulted in a sustainable alternative for disposal of this radioisotope, which is used to obtain a thyroid gamma graph and for treatment of patients in the hospital with thyroid cancer. The administration dose, although very small, generates radioactive waste which is arranged in specific areas until it reaches half-life of iodine-131 (eight days). These doses are then dissolved in an oral solution of sodium iodide-131 (NaI-131). Trials to spread *P. australis* were conducted in a greenhouse at CINVESTAV-

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IPN, Mexico City, and the kinetics was carried out at the premises of the hospitals according to the recommendations considered by Solís-Domínguez *et al.* (2007). The spread of *P. australis* was hydroponic for 6 months and the dosage was performed in a liquid solution of NaI-131.

MATERIALS AND METHODS

P. australis was collected at the root level of adult plants in a town in Cuautla de Morelos, Morelos State, Mexico. The roots were constantly washed until dirt was removed and they were free of any other particles or parasites in the roots (method employed by WANG and JIA, 2009). After cleaning the plants, these were separated by units with respect to their bulbs and placed in Long Ashton nutrient solution (Hewitt, 1966) to begin the propagation of young plants for experimentation. The spread of *P. australis* was carried out under greenhouse conditions with a minimum/maximum temperature range of 13/40 °C and relative humidity of 63/71, with photo-period of 12 hrs of light and 12 hrs of darkness (12/12). The nutrient solution was changed weekly during 6 months in order to obtain the considered sizes (37 to 46 cm), with the number of leaves being 7 and root size of about 36 cm. Once *P. australis* was propagated and ideal conditions were reached for, the next step was to make the dosage of I-131. This dosage was held in the premises of the hospital. Four experimental units and one unit control were used, and these units were composed of 200 mL of distilled water, 5 plants of *P. australis* with saline solution of NaI-131 for each one, plus the control unit was used alone and it contained 5 plants *P. australis* and 200 mL of distilled water.

conclusion at 192 h. The experiment was undertaken in the area of Nuclear Medicine at the hospital, and data on environmental conditions were recorded by security equipment, such as the activimeter and other equipment that measures radiation (Geiger Inspector Alert™ Radiation Monitor - 15520 series). Finally, data were recorded from the experimental units. Once the experimental data were obtained, a statistical analysis was performed using WinQSB and Microsoft Excel programmes.

RESULTS AND DISCUSSION

Regarding nutrient absorption, calibration curves were determined with NaCl at different concentrations (Table 1) and the TDS was quantified in the Long Ashton solution, hence, the adsorption rate was determined for the *P. australis*.

Table 1. Calibration curve with NaCl

| NaCl (gr/50ml) | ppt |
|----------------|------|
| 0.001 | 0.02 |
| 0.002 | 0.04 |
| 0.003 | 0.05 |
| 0.004 | 0.07 |
| 0.005 | 0.08 |

Averages were performed to obtain general TDS data and based on the regression coefficient and a model of simple linear regression equations the consumption of the substrate was also determined, which corresponds to 1.98 ppt of TDS for the *P. australis*. An analysis of variance (ANOVA) was carried out with the WinQSB programme (Table 2).

Table 2. ANOVA calibration curve for NaCl

| Variable Name | Media | Standard deviation | Regression coefficient | Standard error | t value | p value |
|---------------|------------|--------------------|------------------------|----------------|----------------|---------|
| Dependent | ppt | 0.052 | 2.387467E-02 | | | |
| Y – intercept | Constant | | 6.9999E-03 | 3.3166E-03 | 2.1106 | 0.1252 |
| 1 | Conc. NaCl | 0.003 | 1.581139E-03 | 15 | 0.9996 | 15.001 |
| | Se= | 3.1623E-03 | R ² = | 0.9868421 | R – adjusted = | 0.9824 |

During 8 consecutive days (half-life of NaI-131), the activity in the experimental units was measured using an activimeter Capintec CRC® -10R Radioisotope Calibrator. The activity was monitored in 3 mL of deionised water with NaI-131 in each of the experimental units and considering the controlling unit. Cuts were also made in each experimental unit of root, leaf and stem, and readings were taken in each of these sections to determine the amount of adsorbed NaI-131 for the *P. australis*. Similarly, *P. australis* was under observation for its phenotypic conditions in each of the sections (root, leaf and stem), in order to look for the presence of any mutation, intoxication or death due to the addition of the radioisotope. Total Dissolved Solids (TDS) were determined for two months every third day by measuring the electrical conductivity using a conductivity meter (Serial No. 1209531, Oakton Acron series, Eutech Instruments). Based on the data obtained from the conductivity, a calibration curve was performed with sodium chloride (NaCl) in 50 mL of distilled water by a ratio of conductivity: ppt with equipment Hanna DiST® Waterproof (Sigma-Aldrich) and subsequently the consumption of substrate *P. australis* was determined in order to assess the correspondence with the adsorption capacity of the plant. The activity was initially registered at the time of dosing the experimental units to determine the initial activity and measurements were continued approximately every 24 h until

The values obtained with the activimeter were analysed using Eq. (1).

$$A = A_0 e^{-\lambda t} \dots\dots\dots (1)$$

Where:

- A: Activity calculated (Bq)
- A₀: Initial activity (µCi)
- λ: Disintegration (h) According to Eq. (2)
- t: Time lapsed (h)
- t_{1/2}: I¹³¹ half-life time

$$\lambda = \frac{\ln 2}{t_{1/2}} \dots\dots\dots (2)$$

By comparing the observed data with the calculated data, the NaI-131 saline solution absorbed by *P. australis* was determined, and also the amount absorbed in the area of the plant (root, stem and leaves). In Table 3, the initial activities of NaI-131 are observed at the time of dosing for each of the experimental units (1, 2, 3 and 4). The kinetics lasted 192 hours (equivalent to half-life of I-131) in the deionised water mixed with saline NaI-131 and roots at different times, as described in Table 4.

Table 3. Initial Activity

| Experimental Unit | Activity (MBq) |
|-------------------|----------------|
| 1 | 0,3663 |
| 2 | 0,481 |
| 3 | 0,3922 |
| 4 | 0,3293 |

The data time reported for the kinetics of I-131 with intervals of approximately 24 hrs to 192 hrs, showed some environmental disturbances during the readings, that can be explained by the change in climate conditions, occupation of the activimeter in other samples, work-shift change of hospital staff, among others. Using a selection criteria for statistical data, some of the sampled time data were discarded. The data presented in Table 4 are shown in Figs. 1 and 2, where the behaviour of I-131 measured in NaI-131 solution is observed (Figure 1), and also the behaviour at root level of I-131 (Figure 2). Figure 1 shows the decay of NaI-131 saline solution over time, where the behaviour and the nature of the radioisotope I-131 are explained as it decays, even though it is homogenised in a liquid solution (deionised water).

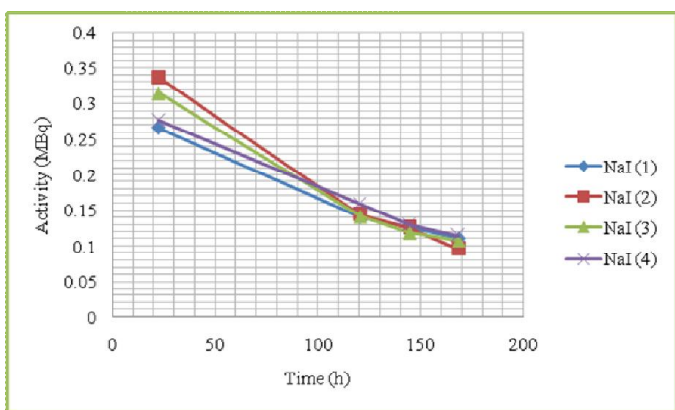


Figure 1. Behaviour of NaI-131 solution

In order to confirm the results, the calculation of the kinetic study was performed using Eq. 1, which determines the activity, ideally calculated by considering as initial concentration a sample of 370 MBq/mL, the concentration at which the drug came to the hospital. For this particular research the calculated activity should be assessed using Eq. (3):

$$C_{Act} = M_{ActNaI-131} + M_{Act\ root} \dots\dots\dots (3)$$

Where:

- C_{Ac} - Calculated activity
- $M_{ActNaI-131}$ - Measured activity with activimeter in NaI-131 solution
- $M_{Act\ root}$ - Measured activity with activimeter at root level

Table 5 shows the comparison of the calculated activity and measured activity in solution of NaI-131 and root. The values show agreement and I-131 is present either in solution or at the root of the plant, since the half-life of I-131 fails to translocate to stem and leaves.

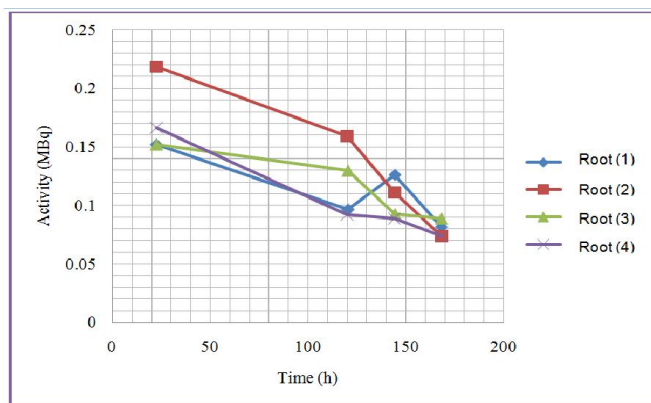


Figure 2. Adsorption of NaI-131 solution at root level of P. australis

Table 4. Observed activity during 192 hours

| TIME (h) | NaI-131 (MBq) | | | | Root (MBq) | | | |
|----------|---------------|--------|--------|--------|------------|--------|--------|--------|
| | 1 | 2 | 3 | 4 | 1 | 2 | 3 | 4 |
| 22.43 | 0.2664 | 0.3367 | 0.3145 | 0.2775 | 0.1517 | 0.2183 | 0.1517 | 0.1665 |
| 120.40 | 0.1406 | 0.1443 | 0.1406 | 0.1591 | 0.0962 | 0.1591 | 0.1295 | 0.0925 |
| 144.47 | 0.1258 | 0.1258 | 0.1184 | 0.1295 | 0.1258 | 0.111 | 0.0925 | 0.0888 |
| 168.34 | 0.111 | 0.0962 | 0.1073 | 0.1147 | 0.0814 | 0.074 | 0.0888 | 0.074 |

Table 5. Comparison of activity calculated and subtraction of measured activity in NaI-131 solution and that bioaccumulated at root level of P. australis

| TIME (h) | Calculated activity (MBq) | | | | $M_{ActNaI-131} + M_{Act\ root}$ (MBq) | | | |
|----------|---------------------------|--------|--------|--------|--|--------|--------|--------|
| | 1 | 2 | 3 | 4 | 1 | 2 | 3 | 4 |
| 22.43 | 0.3377 | 0.4436 | 0.3617 | 0.3037 | 0.1803 | 0.2557 | 0.1822 | 0.1921 |
| 120.40 | 0.2370 | 0.3112 | 0.2538 | 0.2131 | 0.1969 | 0.2915 | 0.2374 | 0.1831 |
| 144.47 | 0.2173 | 0.2854 | 0.2327 | 0.1954 | 0.1455 | 0.1368 | 0.1136 | 0.1065 |
| 168.34 | 0.1994 | 0.2619 | 0.2136 | 0.1793 | 0.0993 | 0.0975 | 0.1079 | 0.0901 |

Figure 2 presents the concentration adsorbed at root level of *P. australis* and the saline concentration NaI-131 added to each of the experimental units. This figure shows the decay of I-131, but at some time, approximately at 144 hrs experimental unit (1), there showed an increase in adsorbed activity of 0.1258 MBq, which is evidence of a positive adsorption of *P. australis* (Table 4). In the case of experimental units (2), (3) and (4), no form of adsorption was observed, therefore, decay of the radioisotope continued for each experimental unit.

The calculations of concentrations consider the same times and conditions used in the prior kinetics (Table 4). During 192 hours experimental units were observed in order to find some kinds of phenotypic effects on *P. australis* when exposed to I-131, in root, leaves and stem of each of the experimental units. The changes observed in *P. australis*, both in colouration and hydration were presented by the surrounding environmental conditions to which they were exposed. For instance, when carrying out the dosages in the area of nuclear medicine at the

hospital, environmental conditions showed slight variations of low temperature due to air conditioning. This was confirmed with the experimental control unit which also showed a gradual loss of colour and dehydration in leaves, which shows that *P. australis* did not present changes due to the dosing with I-131.

Conclusions

Proper selection of plants to use with phytoremediation allows optimisation of treatment and / or pretreatment of contaminants. In this study, a plant known as *P. australis* showed rapid spreading and adaptation and it was demonstrated that the plant is an excellent bioaccumulator of contaminants such as metals, heavy metals, and lixiviates; and currently is proposed as an excellent model for bioaccumulation of radiopharmaceuticals. This research is a contribution to the limited information and experimentation conducted with radiopharmaceuticals; the other significant contribution being that which uses fungi to bioaccumulate radioisotopes with long half-lives. The investigation showed that *P. australis* is a bioaccumulative plant for the radioisotope I-131, which concentrates it at root level. I-131 was not detected in stems and leaves during the duration of the study, possibly due to the short half-life of I-131 and the low concentration used. Phenotypic effects developed in *P. australis* were not because of the dosage of I-131, but are attributed to the environmental conditions under which the experiment was conducted, as demonstrated by the experimental control unit. This proposal is considered as being quite feasible and economic for management of hospital radioactive waste, because it can deal with such waste as a pretreatment during conventional treatments. Also, it optimises storage space, as compared with traditional treatments for the management of radiopharmaceuticals. Despite the short half-life of I-131, which limits a greater adsorption of *P. australis*, it can be a sustainable alternative to be applied *in situ*.

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