# UNCERTAINTIES PREDICTING RADIONUCLEIDES DISTRIBUTION IN ICRP MODELS WITH RANDOM INPUTS

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**Abstract** .- Standards, such as ICRP 78, give criteria to establish bioassay programs assuming acute and chronic intakes. However, in real situations, the individual daily intake for occupational workers are usually a random variable. We have studied the daily intake data from workers exposed for a long period to  $UO_2$  particles. We find that the daily intakes and the concentration values can be fitted to the lognormal distributions. We used an improved method on the approximation of a sum of lognormal distributions to a lognormal distribution is in order to forecast bioassays and its uncertainties. This method can be useful to design and conduct the air control monitoring and the bioassay programs introducing statistical criteria to determine when an bioassay is required and how often should be applied.

# 1. INTRODUCTION

The internal doses for workers exposed to intake of radioactive aerosols, e.g. in uranium and plutonium processing plants, are usually estimated using air samplers. A bioassay program is also applied periodically to the workers monitored with lung counters and urine samplers, and less samplers. frequent with faecal The biokinetic model can be applied to establish a relationship between the individual intake and the bioassay program frequency. In fact, using the models and criteria of ICRPs, lung retention, urine and faecal excretion can be evaluated using the wellknown differential equation

 $\dot{\mathbf{q}}(t) = \mathbf{A} \, \mathbf{q}(t) + \mathbf{b}(t) \,, \, (1)$ 

where  $\mathbf{A} = \mathbf{K} - \lambda \mathbf{I}$ , being  $\mathbf{K}$  the matrix of transfer rates with elements  $k_{ij}$  representing the constant fractional transfer rate from compartment j to i, and  $\lambda$  being the radioactive decay constant. The vector  $\mathbf{q}(t) = \{q_1(t), ..., q_i(t), ..., q_n(t)\}$  represents the content in each compartment i of a n-compartmental system at time t and  $\mathbf{b}(t) = \{b_1(t), ..., b_i(t), ..., b_n(t)\}$  represents the input in each compartment i of the system at time t.

The solution of eqn (1) is given by eqn (2), being  $\mathbf{q}_0 = \{q_1(0), ..., q_i(0), ..., q_n(0)\}$  the content in each compartment *i* at time t = 0,

$$\mathbf{q}(t) = e^{\mathbf{A}t} \mathbf{q}_0 + \int_0^t e^{\mathbf{A}(t-\tau)} \mathbf{b}(\tau) d\tau$$
(2)

The solution of eqn (2) gives that the retention in each compartment,  $q_t(t)$ , is a sum of exponentials. The lung and whole body as well as the daily urine and faecal excretion after a time *t* are sums of contents of several compartments, that is,

$$r(t) = \sum_{\nu=1}^{l} c_{\nu} e^{-d_{\nu}t} \quad (3)$$

r(t) is called the intake retention function and the value of this function at a specific time t is known as the Intake Retention Fraction (IRF). This pattern can be applied for acute and chronic –constant- intakes [1].

IRF values and r(t) for many radioisotopes can be obtained using BIOKMOD available the web site in http://web.usal.es/~guillerm/biokmod.htm. Also, they are a web version in http://www3.enusa.es/webMathematica/Pub lic/biokmod.html, (sponsored by ENUSA Industrias Avanzadas, S.A). IRF values for acute and chronic intakes are widely used in bioassay programs. However, in real situations, the individual daily intake for occupational workers are usually a random variable (r.v.), this means that  $\mathbf{b}(t)$  in eqn (1) is a r.v.. These cases has

been studied by Ketcher and Robinson [2]

and others authors [3]. We are interested in the case when a random intake can be fitted to a known probability distribution function (pdf) in order to provide probability bands around the function r(t) for planning the bioassay program.

# 2. WORKERS EXPOSED TO RANDOM INTAKES

Let us consider a worker exposed to an environment of radioactive airborne. He intakes by inhalation an quantity  $I_j$ . each day *j*. Although the intake happens during a few hours every day from a practical point of view it can be assumed that  $I_j$  is an acute intake. Then, after a period the worker will have been exposed to multi-acute intakes  $I_1$ , ...,  $I_t$ . Then the retentions and the excretions functions after a time t will be the pattern of eqn (4) where the time has been discretized in days, where for convenience we call  $s_j(t) = r(t-j+1)$ ,

$$y(t) = I_1 r(t) + I_2 r(t-1) + \dots + I_t r(1) =$$
  
=  $\sum_{j=1}^{t} I_j r(t-j+1) = \sum_{j=1}^{t} I_j s_j(t).$  (4)

We are interested in studying y(t) and its uncertainties when  $I_i$  is a r.v. as it happens in the real situation. If the pdf of this r.v. can be fitted to the pdf of a known distribution some theoretical tools may be provided. With this purpose we have analyzed the intake data from a group of workers. They work at Juzbado Fuel Fabrication Plant and they has been exposed during a long period of time (usually a few years) to  $UO_2$  (<5% <sup>235</sup>U) radioactive aerosols. The Juzbado factory makes uranium fuel assemblies for light water reactors. We have found (see a previous work [4]) that the daily intake  $I_i$ for workers performing their activities in the same area for a long period of time (> 400 working days) can be modelized by a lognormal distribution  $LN(\mu, \sigma^2)$ , where  $\mu$ and  $\sigma^2$  are the mean and variance of the corresponding normal distribution. The pdf of the lognormal distribution is given by egn (5),

$$f(x) = \begin{cases} \frac{1}{\sigma x \sqrt{2\pi}} \operatorname{Exp}\left[-\frac{1}{2}\left(\frac{\ln x - \mu}{\sigma}\right)^{2}\right] & \text{if } x > 0 \\ 0 & \text{otherwise.} \end{cases}$$
(5)

The mean and variance of the lognormal distribution are

$$\mu_X = e^{\mu + \sigma^2/2}, \quad \sigma_X^2 = e^{2\mu + \sigma^2} (e^{\sigma^2} - 1), (6)$$

while the mean and variance of the associated normal distribution  $(\ln X)$  are

$$\mu = \ln \frac{\mu_X}{\left[1 + \left(\frac{\sigma_X}{\mu_X}\right)^2\right]^{1/2}},$$
$$\sigma^2 = \ln \left[1 + \left(\frac{\sigma_X}{\mu_X}\right)^2\right]. (7)$$

The following estimators derived form the transformation to the Normal distribution are widely used in practice:

$$\mu = \frac{1}{N} \sum_{i} \ln x_{i} ,$$

$$\sigma^{2} = \frac{1}{N} \sum_{i} (\ln x_{i} - \mu)^{2} , (8)$$

where *N* is the total number of observations

 $x_i$  from the lognormal distribution.

In our case the daily intake for a specific worker  $I_i$  plays the role of x and it has to be taken into account that for non working days, Saturdays and Sundays, it is assumed to be zero. Thus, in eqn (4) for each j corresponding to a working day  $s_i(t) I_i$  is the product of a constant and a r.v. It is well known that a r.v. proportional to a lognormal distribution is again lognormal. Therefore,  $s_i(t) I_i$  for working days follows a lognormal distribution. The sum of lognormal variables is not a known distribution. but there are some approximations to lognormal distributions. In particular, we have used the FentonWilkinson's approximation [5] for y(t) - eqn (4) - obtaining a lognormal distribution  $LN(\mu, \sigma^2)$  with mean and variance,

$$u_{1}(t) = E[y(t)] = \sum_{j} e^{m_{j}(t) + \mu + \sigma^{2}/2}, (9)$$
$$u_{2}(t) = E[y(t)^{2}] = \sum_{j} e^{2(m_{j}(t) + \mu + \sigma^{2})} + \sum_{j \neq j'} e^{m_{j}(t) + m_{j'}(t) + \mu + \sigma^{2}/2}, (10)$$

where  $m_j(t) = \ln[r(t - j + 1)]$ . Replacing  $m_j(t)$  in eqn (9) and eqn (10) it obtained that

$$u_{1}(t) = e^{\mu + \sigma^{2}/2} \sum_{j} r(j) = \mu_{I} \sum_{j} r(j), (11)$$
$$u_{2}(t) = u_{1}(t)^{2} + e^{2\mu + \sigma^{2}} \left( e^{\sigma^{2}} - 1 \right) \sum_{j} r^{2}(j) = (12)$$
$$= u_{1}(t)^{2} + \sigma_{I}^{2} \sum_{j} r^{2}(j)$$

where  $\mu_I$  and  $\sigma_I^2$  are the mean and variance of the lognormal distribution of the intake obtained using eqn (6). Thus, the approximated lognormal distribution for eqn (4) has parameters

$$\mu(t) = 2\log u_1(t) - \frac{1}{2}\log u_2(t), \quad .(14)$$
  
$$\sigma^2(t) = \log u_2(t) - 2\log u_1(t)$$

An approximation of the cumulative distribution function (cdf) of y(t) is then

$$P(y(t) \le y_0) \approx \Phi\left(\frac{y_0 - \mu(t)}{\sigma(t)}\right), (15)$$

where the Normal cdf. Probability curves for y(t) can be constructed for a fixed probability  $\gamma$ ,

$$P(u_{1}(t) - b(t) \le y(t) \le u_{1}(t) + b(t)) = \gamma ,$$
(16)

that is,  $b(t) \approx z_{\frac{\gamma+1}{2}} \sqrt{u_2(t) - u_1(t)^2}$ , where  $z_{\frac{\gamma+1}{2}}$  is the  $\frac{100(\gamma+1)}{2}$ -quantile of the Normal distribution. Some kind of probability region can be built for the stochastic process y(t),

$$y(t) \approx u_1(t) \pm z_{\frac{\gamma+1}{2}} \sqrt{u_2(t) - u_1(t)^2}$$
 .(17)

We can use these equations to evaluate y(t) an its uncertainties for workers exposed to a daily intake that can be adjusted for a lognormal distribution.

## 3. APLICATIONS

In this section the theoretical results given above are applied to bioassay programs. All examples are referred to workers exposed to intakes by inhalation of  $UO_2$  (class S) with enrichment 4.4 wt % of <sup>235</sup>U, AMAD  $= 5 \,\mu$ m, and radioactive decay constant near 0. The specific activity is 108 kBq/g U with 3.25% Bq of <sup>235</sup>U. The Dose Conversion Factor (DCF) for this type of particles is 6.66  $10^{-3}$  mSv/Bq. The fact that the individual intake could be represented by a lognormal distribution led us to study the method applied in Juzbado Fuel Fabrication Plant to estimate the intake. This method is similar to those applied in other uranium facilities.

**Example 1.-** A worker has been exposed during the last 2000 days to an intake represented by a lognormal distribution with mean  $\hat{\mu}_{I} = 3.3$  Bq U and standard deviation  $\hat{\sigma}_I = 5.1$  Bq U, estimated with the individual working day intakes. We wish to know whether the lung retention will grow over the Low Level of Detection (LLD) given by a lung counter by 92 Bq U. The lung counter usually measures the <sup>235</sup>U, but 1 Bq U is equivalent to 0.0325 Bq <sup>235</sup>U for an enrichment of 4.4%, then 92 Bq U is equivalent to 3 Bq <sup>235</sup>U). Also it is assumed that there is not intake during the weekends, i.e. it will be assumed that  $I_i = 0$  when j = 7k and j = 7 k-1, k = 1, 2, ...

Here we use the lung intake retention function for uranium ( $^{238}$ U,  $^{235}$ U,  $^{234}$ U) type S and AMAD = 5 µm [1],

$$r(t) = 0.01009 e^{-10.0 t} + 0.007959 e^{-2 t} + 0.01031 e^{-0.0301 t} + 0.01614 e^{-0.0201 t} + 0.03191 e^{-0.0011 t} + 0.004430 e^{-0.00022 t} + 0.001087 e^{-0.0001 t}.$$

Eqn (12) and (17) provide the mean and the probability region as follows:

$$\sum_{j=1}^{2000} r(j) = 25.795, \quad \sum_{j=1}^{2000} r^2(j) = 0.607,$$
$$u_1(2000) = 3.3 \sum_{j=1}^{2000} r(j) = 85.1$$

Then for  $\gamma = 0.95$  (one tail interval):

$$y(2000) \approx u_1(2000) +$$
  
+  $z_{\gamma} \sqrt{u_2(2000) - u_1(2000)^2} =$   
=  $u_1(2000) + z_{\gamma} \sigma_I^2 \sqrt{\sum_j r^2(j)} =$   
=  $85.1 + 1.645 \times 5.1 \sqrt{0.607} = 91.6 \,\mathrm{BqU}$ 

that is practically the lung counter limit of detection (92 Bq U). For this worker the lung retention and its uncertainties are shown in Fig. 1. It can be observed that he reaches the detection limit around 2040 days after starting the intake.

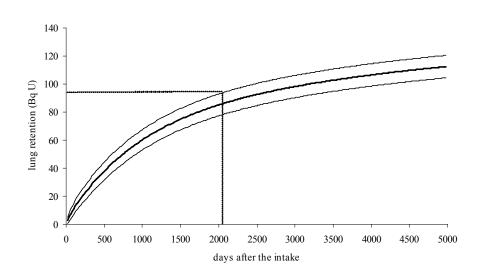


FIG. 1. Predicted values for a lognormal random intake ( $\hat{\mu}_1 = 3.3$  Bq and  $\hat{\sigma}_1 = 5.1$  Bq) and the probability bands with probability  $\gamma = 0.95$  for the lung retention of a worker exposed to a random intake. The detection limit for lung counting is also shown in the graph

**Example 2.-** A group of workers has been exposed for a long time to an daily, in Bq U, intake represented by a lognormal distribution with mean  $\mu_I = 0.43$  and  $\sigma_I^2 = 0.67$ . We want to establish the frequency with which a urinary sample should be taken.

The following criterion established in the Regulatory Guide 8.9 (apart 4.3) [6] will be applied: "In general, spot samples should be

collected frequently enough that there is no more than a 30% increase in the IRF between bioassay measurements".

Here we use the daily urine excretion function for uranium ( $^{238}$ U,  $^{235}$ U,  $^{234}$ U) type S and AMAD = 5 µm [1],

$$r(t) = -1.5 e^{-10 t} - 1.7 e^{-6.0 t} + 1.1 e^{-5.4 t} + 0.00016 e^{-2.0 t} + 2.7 10^{-6} e^{-0.34 t} + 3.5 10^{-6} e^{-0.14 t} + 6.4 10^{-7} e^{-0.099 t} + 1.6 10^{-5} e^{-0.097 t} +$$

3.2 
$$10^{-6} e^{-0.030 t} + 2.1 10^{-6} e^{-0.020 t} + 1.0 10^{-6} e^{-0.013 t}$$
  
+3.1  $10^{-6} e^{-0.001 t} + 4.3 10^{-7} e^{-0.00022 t} + 1.2 10^{-7} e^{-0.0001 t}$ .

From eqn. (11), (12) and (17) it is obtained

$$\frac{u_1(t+T) + z_{(\gamma+1)/2}\sqrt{u_2(t+T) - u_1(t+T)^2}}{u_1(t)} =$$
$$= \frac{u_1(t+T) + z_{(\gamma+1)/2}\sigma_I\sqrt{\sum_{j=1}^{j=t+T}r^2(j)}}{u_1(t)} = 1.30$$

at a specific value of t provides the time t + T for the next measurement. That is, t is the time from the first intake to the moment when a measurement is taken with the body counter. The time from this moment to the time when the next measurement should be made is T. The solution of eqn (18) will be applied for each worker. For instance, if a measurement is made at time t = 2000 the next one should be made T = 800 days later (Fig. 2).

(18)

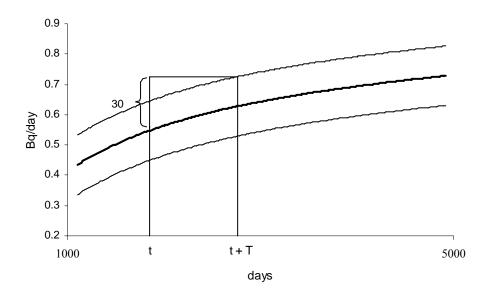


FIG. 2. Probability bands for  $\gamma = 0.95$  for the urine daily excretion of a worker exposed to a random intake.

### 4. **DISCUSSION**

The standards require that monitoring the intake of radioactive material and bioassay programs should be applied depending on radiological hazards, but they establish neither precise criteria of how this radiological hazards should be evaluated nor how often the bioassay should be applied. Standards, such as ICRP 78 [7], give criteria to establish bioassay programs assuming acute and chronic intakes. However, in real situations, the individual daily intake for occupational workers are usually a random variable. We provide formulae forecast the bioassay to measurement and its uncertainties taking into account random intake. In particular, we have found that the daily intake for a group of workers can be fitted to a lognormal distribution. This method can be useful to determine when a bioassay is required as well as the frequency in its application.

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