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Special Contribution

Validation of Popular Models Used in the Analysis of Specific Activity of Primordial Radionuclides in Environmental Samples through 1-D Analytical Modeling

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A scientific model may be considered agreeable if it conforms to given scientific laws, in addition to being able to procure data that can be replicated to an acceptable level when other time-tested and proven methodologies are applied. For first-time researchers such as graduate students, choosing the right model is not always easy and some may opt for a particular model not because they are assured of its correctness, but because it has been used by peers before them. Such tendencies can inadvertently lead to the propagation of flawed models across generations of researchers. In the field of environmental radioactivity, the "comparison" and "conventional" models are frequently used in the evaluation of specific activity of primordial radionuclides in solids. Through one-dimensional (1-D) analytical modeling, this paper shows that while the conventional model conforms to given scientific laws, the comparison model does not since it wrongly assumes a linear between the intensity of gamma radiation through a solid and mass of the solid. A modified version of the comparison model that corrects for difference in mass between the solids being compared (sample of interest and certified reference material) is advanced.

Key words: 1-D modeling, model, conventional model, one-dimensional modeling

1. Introduction

Following an experimental procedure, the data generated may require further processing, for instance through the application of a mathematical model, before the desired information is obtained. For a model to be considered agreeable, if should conform to given scientific laws, in addition to being able to generate data that can be replicated to an acceptable degree when other time-tested and proven methodologies are used. For first-time researchers such as graduate students, choosing the right model is not always easy and some may opt for a particular model not because they are assured of its correctness, but because it has been used by peers before them. Such tendencies may inadvertently lead to the propagation of flawed models across generations of researchers.

Various models are used in the evaluation of radioactivity levels in environmental samples. Some researchers[1-5] for instances opt for the comparison model shown in equation 1.1 while others[6-10] go for the "conventional" model shown in equation 1.2.

$$\frac{I_{ds}}{A_{sp,s}M_s} = \frac{I_{dR}}{A_{sp,R}M_R} \tag{1.1}$$

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$$A_{sp,s} = \frac{I_{ds}}{\gamma \eta M_s} \tag{1.2}$$

With

$$\eta = \frac{I_{dR}}{\gamma A_{sp,R} M_R} \tag{1.3}$$

Respectively, $A_{sp,s}$ and $A_{sp,R}$ (Bq/kg) represent specific activities of radionuclide of interest in sample, *s*, (specific activity not known) and certified reference material, *R*, (specific activity known), M_s and M_R (kg) masses of sample and certified reference material and I_{ds} and I_{dR} (counts/sec) the net gamma intensities transmitted through sample and reference material and registered by the detector. η is the absolute efficiency of the detection system while γ is the probability of emission of gamma ray of interest.

The main objective of this paper is to use one–dimensional (1-D) analytical modeling to test the correctness of the comparison and conventional models. This will provide upcoming researchers with the background information they may need when choosing the model to use for activity concentration analysis of environmental specimen. Besides, use of the correct model will generate data that reflects with high certainty the actual state of the environment in relation to activity concentration of primordial radionuclides.

2. Modeling

2.1. Background

The comparison model suggests that if for example a homogeneous radioactive solid of specific activity A_{sp} is divided into *z* components of unequal masses M_i, i = 1, 2, 3 ... z but of equal cross-sectional areas, and if I_{di}, i = 1. 2. 3 ..... z be the gamma intensity registered by the detector from each component, then;

$$\frac{I_{d1}}{A_{sp}M_1} = \frac{I_{d2}}{A_{sp}M_2} = \ldots = \frac{I_{dz}}{A_{sp}M_z} \tag{2.1.1}$$

This is on the condition that measurement of I_{di}, i = 1. 2. 3 ..... z is performed at the same gamma line while maintaining uniform detector characteristics and comparable positioning of the detector with respect to each component. Equation 2.1.1 implies that the ratio of the gamma intensity transmitted to the detector from the solid to the mass of the solid is constant, magnitude of mass. Equation 2.1.1 may in general be expressed as;

$$\frac{I_{di}}{M_i} = const \qquad i = 1.2.3.....z \tag{2.1.2}$$

There are a number of conditions that must be fulfilled for the conventional model to be used: (i) the detector must be calibrated for efficiency against energy within the range of energies of interest. This is because efficiency is energy dependent; for low energies, the efficiency increases with increase in energy before decreasing non-linearly with increasing energy after some energy threshold is reached. Efficiency-energy calibration is normally carried out using certified reference materials of known activities. (ii) The physical attributes of the sample whose activity is being sought must closely match those of the certified reference material. (iii) Measurement of I_{ds} and I_{dR} should be performed at similar gamma energies while maintaining uniform detector characteristics as well as detector positioning with respect to the sample and to the certified reference material.

Under these conditions, the conventional model reduces to;

$$\frac{I_{di}}{A_{sp,i}} = \frac{I_{dj}}{A_{sp,j}} \tag{2.1.3}$$

i and *j* being two solids that are similar in all aspects except radionuclide content.

In order for the comparison and conventional models to be considered correct, equations 2.1.2 and 2.1.3 must be in agreement with given scientific laws.

2.2. Model development

Consider a homogeneous solid sample, *s*, of uniform cross-sectional area *X*, mass *M*, height (mass-length) *a* and density ρ. Suppose the sample contains *N* uniformly distributed radioactive atoms of a certain radionuclide that decays by gamma emission. Let γ represent the probability that each radioactive decay will be accompanied by emission of gamma ray of a specific energy. If the decay rate (or activity, *A* (Bq)) of the sample is $\frac{dN}{dt}$, then the intensity of gamma radiation (I_s) generated within the sample is;

$$I_s = \gamma \frac{dN}{dt} = \gamma A \tag{2.2.1}$$

Specific activity of a radioactive material is defined as activity per unit mass. If A_{sp} be the specific activity of the sample, then $A = A_{sp} M$. Using this in equation 2.2.1 leads to;

$$I_s = \gamma A_{sp} M \tag{2.2.2}$$

Suppose, as shown in Figure 1, the sample is made up of *Q* thin identical sheets (elements) each of thickness δx such that $Q \cdot \delta x = a$. Suppose too that the gamma

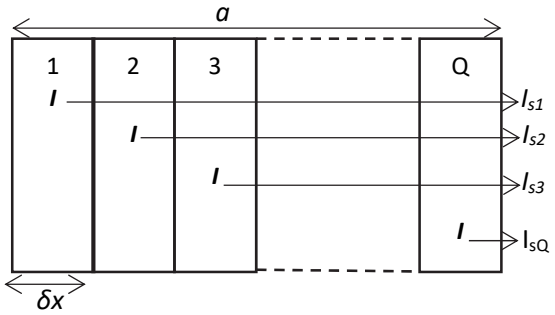

Fig. 1. Conceptual model for gamma generation and movement through the sample.

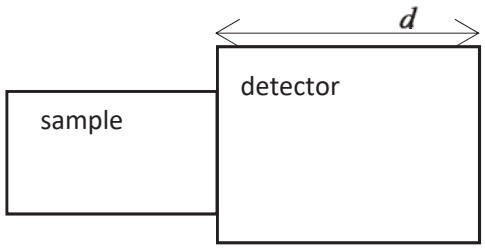

Fig. 2. Detector-sample arrangement cross-section.

intensity generated within each element is equal to I, and let I_{si} *(i=1, 2, 3.....Q)* be fraction of gamma intensity from each sheet i reaching the surface of the sample. By Beer-Lambert's principle, the small change in intensity (δI) across each element is given by;

$$\delta I = -\mu_s I \delta x \quad \text{or} \tag{2.2.3}$$

$$\frac{dI}{I} = -\mu_s dx \quad \text{when } \delta x \to 0 \tag{2.2.4}$$

Where μ_s is mass-attenuation coefficient of gamma radiation through the solid.

The intensity of gamma radiation (I_{s1}) from sheet 1 reaching the surface of the sample travels the entire mass-length a and is obtained by integrating equation (2.2.4) under the boundary conditions $I_{x=0} = I$ and $I_{x=a} = I_{s1}$. That is;

$$\int_I^{I_{s1}} \frac{dI}{I} = -\mu_s \int_0^a dx \tag{2.2.5}$$

$$\Rightarrow \ln\left(\frac{I_{s1}}{I}\right) = -\mu_s a \tag{2.2.6}$$

Equation 2.2.6 simplifies to;

$$I_{s1} = Ie^{-\mu_s a} \tag{2.2.7}$$

Intensity (I_{s2}) reaching the sample surface from sheet 2 covers a reduced distance equal to a-δx. To find I_{s2}, equation 2.24 is integrated with boundary conditions $I_{x=0} = I$ and $I_{x=a-\delta x} = I_{s2}$;

$$\int_I^{I_{s2}} \frac{dI}{I} = -\mu_s \int_0^{a-\delta x} dx \tag{2.2.8}$$

$$\Rightarrow \ln\left(\frac{I_{s2}}{I}\right) = -\mu_s (a - \delta x) \tag{2.2.9}$$

$$\Rightarrow I_{s2} = Ie^{-\mu_s (a-\delta x)} = Ie^{-\mu_s a} e^{\mu_s \delta x} \tag{2.2.10}$$

Similarly transmitted gamma radiation (I_{s3}) from sheet 3 covers a reduced distance a-$2\delta x$. Thus;

$$\int_I^{I_{s3}} \frac{dI}{I} = -\mu_s \int_0^{a-2\delta x} dx \tag{2.2.11}$$

$$\Rightarrow I_{s3} = Ie^{-\mu_s (a-\delta x)} = Ie^{-\mu_s a} e^{2\mu_s \delta x} \tag{2.2.12}$$

Following the pattern generated in equations 2.2.7, 2.2.10 and 2.2.12, transmitted gamma intensities I_{s4}, ..., I_{sQ} from, respectively, sheets, 4, .., Q can be expressed as;

$$I_{s4} = Ie^{-\mu_s (a-3\delta x)} = Ie^{-\mu_s a} e^{3\mu_s \delta x} \tag{2.2.13}$$

$$I_{sQ} = Ie^{-\mu_s (a-(Q-1)\delta x)} = Ie^{-\mu_s a} e^{(Q-1)\mu_s \delta x} \tag{2.2.14}$$

The total gamma intensity (I_{st}) transmitted to the surface of the sample s is given by;

$$I_{st} = I_{s1} + I_{s2} + I_{s3} + ... + I_{sQ} \tag{2.2.15}$$

Thus;

$$I_{st} = e^{-\mu_s a}(I + (Ie^{\mu_s \delta x} + Ie^{2\mu_s \delta x} + Ie^{3\mu_s \delta x} + ... + Ie^{(Q-1)\mu_s \delta x})) \tag{2.2.16}$$

As $\delta x \to 0$, $e^{i\mu_s \delta x} \to 1, (i = 1,2,3,....,(Q-1))$ and equation 2.2.16 reduces to;

$$I_{st} = e^{-\mu_s a}(I + I + ... + I) = e^{-\mu_s a}(QI) \tag{2.2.17}$$

Now, $QI = I_s$, I_s being the total gamma intensity generated within the sample. Equation 2.2.17 may therefore be written as:

$$I_{st} = I_s e^{-\mu_s a} \tag{2.2.18}$$

Using equation (2.2.2) in equation 2.2.18 and rearranging gives;

$$\frac{I_{st}}{\gamma A_{sp} M} = e^{-\mu_s a} \tag{2.2.19}$$

Suppose a detector of uniform cross-section area and length d is placed in contact with the surface of the solid sample being analyzed (Fig. 2) and that the entire gamma radiation (I_{st}) transmitted to the surface of the solid impinges on the detector surface. As the radiation travels the length of the detector, part of it (I_d) is absorbed and registered by the detector (and quantified in counts/sec) while the rest (I_{dt}) is transmitted. Thus, the gamma radiation transmitted through by the detector may be expressed as;

$$I_{dt} = I_{st} - I_d \tag{2.2.20}$$

By Beer-lambert's principle, the gamma intensity transmitted through the detector may also be calculated from;

$$I_{dt} = I_{st} e^{-\mu_d d} \tag{2.2.21}$$

Where μ_d is the gamma mass-attenuation coefficient of the detector.
Equations 2.2.21 by 2.2.20 are equivalent which means that;

$$\frac{I_d}{I_{st}} = 1 - e^{-\mu_d d} \tag{2.2.22}$$

So long as the detector characteristics and positioning with respect to the sample remain unchanged, the Right-Hand-Side (RHS) of equation 2.2.22 is a constant, say ε_{int}. The constant is equivalent to the intrinsic efficiency of the detector. Thus;

$$\frac{I_d}{I_{st}} = \varepsilon_{\text{int}} \tag{2.2.23}$$

Mutiplying equation 2.2.19 by 2.2.23 leads to;

$$\frac{I_d}{\gamma A_{sp} M} = \varepsilon_{\text{int}} e^{-\mu_s a} \tag{2.2.24}$$

From definition of density, $a = M/\rho X$. Using this in equation 2.2.24;

$$\frac{I_d}{\gamma A_{sp} M} = \varepsilon_{\text{int}} e^{-\beta M} \tag{2.2.25}$$

Where $\beta = \mu_s/\rho X$ is constant.

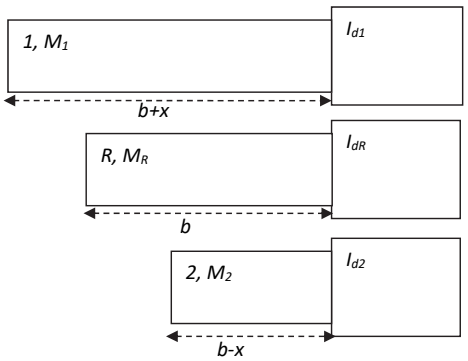

Fig. 3. Detector-sample arrangement.

3. Discussion

3.1. Model implications

To test the correctness of the comparison model, suppose the sample of specific activity A_{sp} is divided into two samples, 1 and 2, of un-equal masses M_1 and M_2 but of uniform cross-sectional areas. Let I_{d1} and I_{d2} represent the intensities of transmitted gamma radiation (same gamma line) from samples 1 and 2 respectively. By equation 2.2.25,

$$\frac{I_{d1}}{\gamma A_{sp} M_1} = \varepsilon_{\text{int}} e^{-\beta M_1} \tag{3.1.1}$$

$$\frac{I_{d2}}{\gamma A_{sp} M_2} = \varepsilon_{\text{int}} e^{-\beta M_2} \tag{3.1.2}$$

Dividing equation 3.1.1 by equation 3.1.2 gives;

$$\frac{I_{d1}}{M_1} = \frac{I_{d2}}{M_2} (e^{-\beta (M_1 - M_2)}) \tag{3.1.3}$$

Contrary to the 2.1.2;

$$\frac{I_{d1}}{M_1} \neq \frac{I_{d2}}{M_2} \Rightarrow \frac{I_{di}}{M_i} \neq const \tag{3.1.4}$$

In the conventional model, the physical characteristics of solids of interest are kept constant. With mass constant, the exponential part on the RHS of equation 2.2.25 reduces to a constant. This means that $\varepsilon_{\text{int}} e^{-\beta M} = const\,(=\eta)$. The constant η is equivalent to the absolute efficiency of the detection system. To test the correctness of the conventional model, consider two solids 1 and 2 that are identical in all aspects other that their radioactivity level. If A_1 and A_2 be their respective specific activities and I_{d1}

and I_{d2} the gamma intensities registered by the detector, then by equation 2.2.25;

$$\frac{I_{d1}}{\gamma A_1 M} = \eta \tag{3.1.5}$$

$$\frac{I_{d2}}{\gamma A_2 M} = \eta \tag{3.1.6}$$

Dividing equation 3.1.5 by 3.1.6 gives;

$$\frac{I_{d1}}{A_1} = \frac{I_{d2}}{A_2} \tag{3.1.7}$$

Equation 3.1.7 is equivalent to equation 2.1.3.

3.2. Modified version of the comparison model

Consider a solid of uniform cross-sectional area, mass M, mass-length a and specific activity A_{sp}. Suppose the sample is divided into three sub-samples 1, 2 and R of equal cross-sectional areas but different masses M_1, M_2 and M_R such that $M_1>M_R>M_2$. If mass-lengths of 1, 2 and R be $b + x$, b and $b - x$, and if I_{d1}, I_{d2} *and* I_{dR} be the gamma intensities registered by a detector placed in contact with the masses 1, 2 and R as shown in Figure 3, then by equation 2.2.24;

$$\frac{I_{d1}}{\gamma A_{sp} M_1} = \varepsilon_{\text{int}} e^{-\mu_s (b+x)} = \varepsilon_{\text{int}} e^{-\mu_s b} e^{-\mu_s x} \tag{3.2.1}$$

$$\frac{I_{dR}}{\gamma A_{sp} M_R} = \varepsilon_{\text{int}} e^{-\mu_s b} \tag{3.2.2}$$

$$\frac{I_{d1}}{\gamma A_{sp} M_1} = \varepsilon_{\text{int}} e^{-\mu_s (b-x)} = \varepsilon_{\text{int}} e^{-\mu_s b} e^{\mu_s x} \tag{3.2.3}$$

Assuming R to be the reference sample and dividing equations 3.2.1 and 3.2.3 by equation 3.2.2 leads, respectively, to:

$$\frac{I_{d1}}{\gamma A_{sp} M_1} = \frac{I_{dR}}{\gamma A_{sp} M_R} e^{-\mu_s x} \tag{3.2.4}$$

$$\frac{I_{d2}}{\gamma A_{sp} M_2} = \frac{I_{dR}}{\gamma A_{sp} M_R} e^{\mu_s x} \tag{3.2.5}$$

The exponential components $e^{\pm \mu_s x}$ in equations 3.2.4 and 3.2.5 represent the correction factors, with x as the difference between mass-length of the samples being compared. The exponent is negative if the mass-length of the sample of interest is greater than that of the reference material and positive if less than that of the reference material.

4. Conclusion

In order to test the correctness of comparison and conventional models commonly used in the evaluation of radioactivity levels in samples, 1-D analytical modeling of radiation transmission through a solid and eventual detection by a detector was carried out and the following observations made: (i) for uniform detector characteristics and positioning with respect to the solids being analyzed, the intensity of gamma radiation (I_{st}) reaching the solid surface is directly proportional to the intensity of gamma radiation (I_d) absorbed (registered) by the detector, (ii) the conventional model conforms to given scientific laws and for this reason can be considered to be valid, (iii) the comparison model cannot be considered valid since it wrongly assumes a linear relationship between mass of the sample and the intensity of gamma radiation registered by the detector (iv) the intensity of gamma radiation through a solid of uniform cross-sectional area reduces exponentially with mass, (v) the comparison equation can be corrected for difference in mass-length (between solids being compared) by the application of an exponential correction factor. The exponent is negative if the mass-length of the sample is greater than that of the reference material and positive if less than that of the reference material.

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Conflict of Interest

The authors declare that the reported study was performed in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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