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APPLICATIONS OF EXPERIMENTAL DESIGN TECHNIQUES, THROUGH THE USE OF DIAGNOSTIC STATISTICAL FUNCTIONS AND GRAPHICAL METHODS, TO IDENTIFY THE RANDOM ERROR THAT COULD HAVE OCCURRED IN THE TRADITIONAL DECAYING VALUES OF TECHNETIUM-99

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ABSTRACT

Objective: Use of Experimental Design Statistics to identify the random errors that are occurring in the established or traditional decaying values of the disintegrations of Technetium-99 per time. ***Methodology:*** Usage of the conventional Tc-99 data, and its subsequent graphing. Plotting of logarithmic transformations, to check the order the reaction law aimed at the identification of experimental errors. Adjusting time series regression diagnostic techniques and the Durbin-Watson statistic, to test for autocorrelation in the residuals in statistical regression analysis, to check for first order autocorrelation causing experimental errors. ***Results and discussions:*** The Tc-99 disintegrations are negatively autocorrelated, thus precluding precision of medical clinical applications. ***Conclusions:*** The disintegrations values are negatively autocorrelated, preventing precise measurements. The established decaying calculations have undetected experimental faults and are not in compliance with the Central Limit Theorem, which says that as the sample sizes get larger, the distribution of means calculated from repeated sampling will approach normality, thus preciseness. ***Recommendations:*** A revision of the established disintegration values is recommended by refining the laboratory techniques and/or by increasing the mean sample size to make the data more accurate.

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INTRODUCTION

This is a study of nuclear chemistry associated with the disintegrations of the radioisotope Technetium-99 as a function of time. Technetium-99m is a metastable nuclear isomer of technetium-99 used in millions of medical diagnostic procedures, making it the most commonly used radioactive tracer isotope in Nuclear Medicine. The artificial radioisotope technetium-99, which decays by beta emission, is the most widely used isotope in medical applications. The data is expressed in disintegrations per minute versus time. Technetium-99 (99Tc) is an isotope of technetium which decays with a half-life of 211,000 years to stable ruthenium-99, emitting beta particles, but no gamma rays. Technetium-99m (99mTc), a metastable nuclear isomer used in nuclear medicine is a short-lived (half-life about 6 hours) and is used in nuclear medicine. The common isotope of technetium is Tc-99, but it is the metastable isotope Tc-99m.is the most pursued. Here, the 'm' in Tc-99m stands for metastable having an excited nucleus. In any event, this investigation is centered in applying experimental design techniques to the traditional statistical distribution values of the disintegrations of Technetium-99, to identify the experimental errors that could have occurred during the old-fashioned calculated disintegrations of this isotope. In fact, this research is using scientific and persuasive statistical arguments to accumulate evidence to show the traditional statistical distribution disintegration values of Tc-99 are not in compliance with the Central Limit Theorem, because they lack normality or symmetry, thus not precise, according to the conventional disintegrations of the actual data used. In this connection, the Central Limit Theorem (CLT) is a statistical theory saying that given a sufficiently large mean sample size from a population with a finite level of variance, the mean of all samples from the same population will be approximately equal to the mean of the population. This means that, as the random sample size increases, the distribution of means calculated from repeated sampling will approach the normal distribution, thus, precluding experimental errors. The current literature of applications of experimental statistical design techniques to the established or traditional disintegrations of Tc-99 is very scarce or none at all. The findings of this study suggest the established decaying calculations of the disintegrations of

Technetium-99 could have inherent experimental errors, because they are negatively auto correlated. In this instance, a negative autocorrelation implies that if a particular value is above average, the next or previous value is more probable to be below average. This condition can lead to experimental errors. To this end, this research is persuading medical researchers to revise (if they have not done so), the actual calculated values used in clinical applications of this radioisotope. This is because of the faults found in this research related to the traditional or conventional values of the disintegrations of Tc-99. Technetium-99m (symbolized as 99mTc) is used in millions of medical diagnostic procedures, making it the most popular medical radioisotope used. According to the Environmental Protection Agency (EPA) of the United States, this source of information describes Technetium (chemical symbol Tc) as a silver-gray, radioactive metal that occurs naturally in very small amounts in the earth's crust, but is primarily artificially made. Technetium-99m is a short-lived form of the isotope Tc-99 that is used as a medical diagnostic tool. This isotope has a short half-life (6 hours) and does not remain in the body or the environment for a long time. However, Technetium-99 can pose a health risk when it enters the body because Tc-99 concentrates in the thyroid gland and the gastrointestinal tract. Yet, the body constantly gets rid of Tc-99 in feces. As with any other radioactive material, there is an increased chance that cancer or other adverse health effects can result from exposure to radiation (Environmental Protection Agency, 2017).

Furthermore, conferring to a *Georgia State University* study, Technetium-99m is a widely used radioactive tracer isotope used in Nuclear Medicine. Its gamma ray energy of about 140 keV is convenient for detection (Georgia State University, ND). Hence, the fact that both its physical half-life and its biological half-life are very short, this leads to very fast clearing from the body after an imaging process. In accord to this information, Technetium-99 is produced by bombarding molybdenum ^{98}Mo with neutrons. The resultant ^{99}Mo decays with a half-life of 66 hours to the metastable state of Tc. This process permits the production of 99mTc for medical purposes in the form of pertechnate (TcO_4^-). Still further, regarding the properties of Technetium-99m, bestowing to the organization *Radioactive Isotopes*, the Tc-99 is adequate for medical applications, because of its properties which have ideal characteristics for medical scanning. For example, it has a very short half-life of 6 hours which is long enough to examine metabolic processes, yet short enough to minimize the radiation dose to the patient. Besides, this isotope emits low energy gamma radiation that minimizes damage to tissues, but can still be detected in a person's body by a gamma ray sensitive camera (Radioactive Isotopes, ND). Accordingly, Technetium is reasonably reactive; it can be reacted to form a compound with chemical properties that leads to concentration in the organ of interest such as the heart, liver, lungs, bones or thyroid gland. Technetium-99m decays by a process called "isomeric"; which emits gamma rays and low energy electrons. Since there is no high-energy beta emission, the radiation dose to the patient is low. The low energy gamma rays it emits, easily escape the human body and are accurately detected by a gamma camera. Conferring to this information, the chemistry of Technetium-99 is so versatile; it can form tracers by being incorporated into a range of biologically-active substances to ensure that it concentrates in the tissue or organ of interest. The short half-life of the isotope allows for scanning procedures that collect data quickly. The fact that both its physical half-life and its biological half-life of Tc-99 are very short, it leads to very fast clearing from the body after an imaging process. A further advantage is that the gamma is a single energy, not accompanied by beta emission, and that permits more precise alignment of imaging detectors. Still further, applying mathematics to the disintegration of TC-99, the radioactive decay followed by this isotope, where the rate of decay of the nuclei is directly proportional to the number of active nuclei present at that instant, is called a first order reaction. A first order chemical reaction refers to the rate of a reaction which is directly proportional to the concentration of the reacting substance. First-order reactions often have the general form A → products. The differential mathematical rate for a first-order reaction is: $\text{Rate} = -d[A] / dt = k[A]^n$. Where k is proportionality constant called rate constant, n is the first order of the reaction, $[A]$ is the concentration of the substance after a time t has passed and t is the time.From our perspective, regarding first order reactions, it is well-known that all radioactive decaying processes follow first order reactions (including pesticides decay or biochemical oxygen demand kinetics). The rate laws for most reactions have the general form: $\text{Rate} = k[\text{reactant } 1]^m [\text{reactant } 2]^n$, where m and n are called reaction orders and their sum is the overall reaction order. In this way, a first order reaction is defined as a reaction in which the rate of the response is directly proportional to the concentration of the reactant species, at any instant. In this context, to know if a chemical reaction is truly a first order, the plotting of the logarithmic values, as a function of time yields a straight line, with all the points situated exactly on the line (provided there are no experimental errors, a situation that did not occurred in the decaying values of Tc-90). Outweighing the previous mathematical reaction functions, if we write the instantaneous rate law for such processes, the mathematical description of a first reaction can be written as $dC/dt = -kC$, where k is a constant and C is the instantaneous concentration of the reactant species. After the algebraic manipulation of this differential equation, the integration of $\text{Ln } [A]/[A]_o = -kt$, yields $[A] = [A]_o \exp\text{-}(kt)$. In this instance, it was precisely this kind of mathematical reasoning of first order reactions that our research utilized to identify the experimental errors that the current distribution data of the disintegrations of Tc-99 could have.

Furthermore, about the history of Technetium, rendering to the organization *JPNM Physics Isotopes*, element 43 was predicted on the basis of the periodic table and originally reported to have been discovered in 1925 and named "masurium." The element was actually discovered by Perrier and Segre in Italy in 1937 in a sample of molybdenum which had been bombarded by deuterons by none other than E.O. Technetium was the first element to be produced artificially, but it is has not been found in nature and appears to be appropriately named. Ahead, on the uses of Technetium, regarding this substance in the environment, convening to the source *Lenntech*, since Technetium is produced in big quantities in nuclear reactors, it is adding to the planetary burden of unwanted radioactive waste, because some technetium escapes to the environment via its use in medical diagnosis (Lenntech, 2019). On the other hand, relating the current literature on the applications of experimental design statistical techniques, this research used time series regression analyses. Rendering to the organization *Research Methodology*, regression analysis is a quantitative research method used when the study involves modeling and analysis of several variables, where the relationship includes a dependent variable (*Y*) and one or more independent variables (*X*) (Research Methodology, 2019). Similarly, *Neter et al.* (1996) discuss their views on regression models, the diagnostics and remedial measures and give an overview of tests involving residual plots. Moreover, *Devore* (2000) discourses probability regression and correlation models and the functions for estimating model parameter. Likewise, *Montgomery* (2005) provides an overview of the topic related to the fitting of regression models. This statistical scientist also confers some basic statistical concepts of noise in the results of samples that have variations. Accordingly, this noise is usually called "experimental error" which is a statistical error that arises from variation that is uncontrolled and unavoidable. Similarly, the organization *Business Dictionary* defines the term, "experimental errors," as errors that may occur in the execution of a statistical experiment design. In view of that, these types of experimental errors include human errors, mistakes in data entry; systematic errors, or mistakes in the design of the experiment itself (*Business Dictionary*, ND). This notation also refers to random errors caused by environmental conditions or other unpredictable factors. In this context, our research is focusing on the "experimental error," as a very important issue occurring in the measurements of the traditional disintegrations of the isotope Tc-99. Furthermore, the *College of Education and Integrated Studies of Cal Poly Pomona*, discuss statistical uncertainties, when there are measurements of radioactive decays in the extents of recorded counts per minute. They add that, when "we understand the statistical nature of the decaying process, we can estimate the variations in the number of recorded counts, and this variation is called statistical uncertainties of measurements." Still further, the *Free Dictionary* by *Farlex* defines the experimental errors, as errors of detection or measurement due to inadequate techniques. We believe this blemish could have been the case in the traditional measurements of the traditional disintegrations of Tc-99 (*Free dictionary* by Farlex, ND). Likewise, conferring to the organization *More Steam*, it gives additional information about the term "errors" which can result due to unexplained variation, situations that can obscure the desired results (*More Steam, 2019)*.

In addition, this information discusses uncontrollable factors that induce variation under normal operating conditions, which are referred to as "*Noise Factors*." Here, however, the term "error" is not a synonym with "mistakes". Error refers to all unexplained variation that is either within an experiment run or between experiments runs and associated with level settings changing. About the relation of so called experimental error, in experimental design, according to the *Minitab* computer program, Dr. Taguchi calls 'noise' the common cause of variation. In this stance, the relation of signal-to-noise provides a measurement of impact in the evaluation of the outcomes of an experiment to the ratio sound/noise. Here, the greater the relation of this signal to noise, the better off would be the results (Minitab computer program, 2019). Agreeing with the previous reasoning, we believe the classical values of the disintegration of Tc-99, have random experimental errors. From our stance, in this deference, the noise variation and the *Central Limit Theorem* (CLT) are closely related, in the sense that, as more variation of the data exists, the less the CLT is conformed. So, as explained previously, according to this Theorem, for a stochastic model to be good, there should not be variation and the sample size distribution of the means should be sufficiently large (n ≥ 30 random sample mean cases), provided the parental population is normal or nearly normal. If these conditions are not met, there will be experimental or noise errors. As asserted before, the CLT argues that the plotting the distribution of the mean of the means will resemble the normal distribution and the precepts of the CLT will be fulfilled. If, however, there is variation or a small mean sample size, the CLT principles will not be met and there will be experimental noise, thus inappropriate results. We contend, this is precisely the situation with the actual disintegrations of Tc-99. At any rate, from our perspective, our experimental design techniques pursue to identify and minimize the noise or experimental error, so the product will be more accurate. We did this, so to optimize the results in investigation, in academic pursuits or in industrial or medical applications. In this context, our research is focusing on the "experimental error," as a very important issue occurring in the measurements of the traditional disintegrations of the isotope Tc-99. About the notion of statistical regression analysis, extending a bit more the concept of what is called regression, from our view, there are applications of experimental design techniques involving linear, multiple and polinomial regression functions. In simple terms, experimental design is a technique whose objective is to make a series of tests aimed to analyze the cause and effect relationship in the pursuit of perfection of systems, scientific or industrial procedures or results. Our research applied regression techniques to the data values of the disintegration of Tecnetium-99 from chemistry books. To mention one is the text *Chemistry. The Central Science* of the authors Brown, LeMay and Bursten. For example, this book of chemistry cites an example of the disintegrations of technetium (page 838, of Chapter 21, exercise 121.69) and gives the table values of data collected on a sample of Tc-99. It was exactly the values of this table the ones this research used to process the data given hitherto. We did this, to investigate the factors that can influence the variable of interest (the disintegrations of Tc-99) aimed to identify hidden experimental errors. In fact, we applied experimental design techniques in time series regression analyses to the disintegrations of Tc-99, to identify the possible experimental errors that could have occurred in the calculated traditional values of the disintegrations of this radioisotope.

From our view, time series analysis is a statistical technique that covenants with time series data, or trend analysis. Time series data means that data is in a series of particular time periods or intervals. Here, our time series regression techniques made transformations of the disintegrations (Log_e) of the values of Technetium-99, as a function of time, to check on the type of reaction followed (first order) and the founding of the regression equation followed by the actual disintegrations of Tc-99. But most importantly, our study evaluated the utility of the regression model through diagnostic objectivistic and subjectivist residual graphs. and the use of the Durbin-Watson function, to test for first order autocorrelation or serial correlation to identify the possible random errors that could have arisen in the traditional or communal values of the disintegrations of Tc-99. In this instance, a residual plot refers to a graph that shows the residuals on the vertical axis and the independent variable on the horizontal axis. In this way, if the points in a residual subjectivist graph are randomly dispersed around the horizontal axis, a linear regression model is appropriate for the distribution data. Otherwise, a non-linear model is more appropriate. In this fashion, to identify the experimental errors of the established values of the disintegrations of Tc-99, our method used objectivistic diagnostics (direct perception of information utilizing statistical functions) that included the coefficient of determination (R^2), the standard error of estimate (s), the predicted sum of squares (PRESS) and the analysis of variance table (NOVA table). This approach was complemented by subjectivist diagnosis (indirect or complementary information of residual graphs) which included normal probability graphs, fitted values, histograms and observation order plots. Nevertheless, most importantly is the identification of autocorrelation and of experimental errors, through what is called the Durbin-Watson (D-W) diagnostic statistic used to detect autocorrelation problems (as what happened to the disintegrations values of Tc that have negative first order autocorrelation). With this Durbin-Watson statistic the rule of thumb is to have a value of 2.0 or very close to 2.0 for the time series model to be free of experimental errors owing to negative or positive autocorrelation faults. Generally, values of the D-W statistic greater than 2.0 indicate negative autocorrelation, whereas values smaller than 2.0, indict positive autocorrelation. In this respect, autocorrelation (correlation between the elements of a series and others from the same series separated from them by a given interval) problems in time series is intimately associated to experimental errors.

The presence of autocorrelation violates the supposition of independence and can mislead the validity of the objectivist results of R^2, s, PRESS, and ANOVA table. We contend, this is the case, when we analyzed the traditional values of the disintegrations of Tc-99. More succinctly, autocorrelation is a characteristic of data which occurs when there is a correlation between the values of the same variables based on related objects. This condition violates the assumption of independence. Such patterns tell us that something is wrong somewhere. To support the previous statements, according to *Nowosad, et al.* (2014) the structuration of correlograms (a visual way to show serial correlation in data that changes over time, i.e. time series data) is a valuable tool to determine the pattern of autocorrelation and the point where the autocorrelation ceases to be a problem. In this way, in applications of regression to time series, these models are susceptible to serial autocorrelation, circumstances that generate experimental errors that can compromise the desired results. What's more, *Penn State Eberly College of Science Stat*(ND) 501 discusses remedial measures for autocorrelation involving the Durbin-Watson test. They argue that the violation of independence of the error terms occurs because there is a known temporal component for how the observations were drawn. They say the easiest way to assess, if there is dependency, is by producing a scatterplot of the residuals versus the time measurement for that observation (assuming you have the data arranged according to a time sequence order). Agreeing with this assertion, if the data are independent, then the residuals should look randomly scattered about zero. However, if a noticeable pattern emerges (particularly one that is cyclical) then, dependency is likely a problem. In this instance, from the traditional point of view, the presence of first order time series autocorrelation can be detected by analyzing the normal probability residual plots, the fitted values and the histogram graphs and the analysis of the Durbin-Watson statistic. For example, in the normal probability plot, for the data to be free of experimental errors, all the data points must be placed on the least squares line or very close to it (a situation that did not occurred with the values of the Tc-99). Moreover, on the plot of the versus fits, there must be the same number of positive and negative values (a situation that did not occurred with the disintegration values of Tc-99, because there were more positive than negative values, a situation that flagged negative autocorrelation). Besides the histogram has to be bell-shaped (a situation that did not occurred with the values of Tc-99, because it was skewed to the right). Also the D-W statistic was higher than the accepted criterion of 2.0. This is because the resulting value of the Durbin–Watson statistic of 2.43167 was higher, than the standard value of 2.0. About the detection of first autocorrelation burdens, adding to the previous observations, our research came up with a possible innovative method, to detect instantaneous first order autocorrelation flaws. For example, by analyzing the first order reaction graph of Figure 2, that is, the plotting the ordinate transformed values versus time, if there are more distant point values below the regression line, than above this line, this observation indicates a negative autocorrelation (as it occurred here in the disintegrations of Tc-99.). If, however, there are more distant point values above the regression line, than below, this observation could flag a positive autocorrelation (a situation that was not the case, here). At any rate, convening to the previous arguments, autocorrelation has serious impacts, because it indicates the existence of experimental errors that can compromise the results (as it could have occurred in this study of the disintegrations of Tc-99). Furthermore, from our viewpoint, the presence of serial autocorrelation has very serious concerns. The main setback with autocorrelation is that it makes a model looks good, when in fact is not. For example, a negative autocorrelation occurs when an error of a given sign tends to be followed by an error of the opposite sign. In this stance, positive errors are usually followed by negative errors and negative errors are usually followed by positive errors. On the other hand, positive autocorrelation occurs when an error of a given sign tends to be followed by an error of the same sign. In this way, one consequence of autocorrelation is on the objectivist evaluation involving the regression equation, R^2, *s*, PRESS, mean square quadratic error (MS), etc.

Thus, under these circumstances, the interpretation of these diagnostics can be misleading, because the regression coefficients are impartial, but not efficient. Besides, in positive autocorrelation the mean quadratic error (MSe) can sub estimate the variance error (σ^2), whose value dictates the magnitude of the F value in the ANOVA table. The resulting unreal values of F will invalidate the test of hypothesis. Moreover, as the summations of the errors e^2 are affected, so will the value of R^2 which will be overestimated. All these pitfalls will make the objectivist statistical diagnostics of the model, questionable. We believe these kinds of drawbacks could be affecting the diagnostic statistics precision of the disintegrations of Tc-99. For the evaluation of the utility of the distribution of the disintegrations/min of the isotope Technetium-99 versus time, using the established values, our procedure applied an objectivistic assessment, which included the calculation of the regression equation, the coefficient of determination (R^2), the standard error of estimate (s), Predicted sum of squares (PRESS), variance inflation factors (VIFs), the D-W statistic and the Analysis of Variance (ANOVA) table. As mentioned before, R^2 is a statistic utilized to measure how good the regression model fits the data; it estimates the population coefficient ρ. In this context, as the R^2 value approaches 100%, the data fits the model (in our case R^2 turned to be 99.8%). Here, however, lower values of R^2, not necessarily mean poor fitted models. By the same token, high values of R^2 not necessarily mean good fits, if there are autocorrelation faults. For the model to be good, all other objectivist and subjective diagnostic functions and serial autocorrelation snags must be evaluated, accurately, before a final judgment is made on the accuracy of the best selected regression model. Similarly, the value of the standard error of estimate, *s* gives an indication of the amount of experimental error that can exist. About the PRESS value, this is a statistical function utilized to evaluate the usefulness of the selected regression model. Here, small values of PRESS produce good candidate models, with no variation; but also, the reverse is true. About the meaning of the R^2 statistic, even though this diagnostic is a popular measure of quality of fit in regression, this function, however, does not offer any significant insights into how well the regression model can predict future values. In its place, the PRESS statistic can be utilized, as a measure of predictive power of the regression equation. Furthermore, about the Variance Inflation Factors (VIFs), this statistic quantifies how much the variance is inflated, as compared to when the predictor variables are not linearly related.

In this deference, an acceptable value of VIF is about 5. Here, however, we will not be dealing with VIFs, because they only apply to multiple regression analyses. Further on, as shown before, in time series regression analysis, as exposed in Figures 3, the importance of the D-W statistic is a very important function that must be evaluated to test is a measure of the degree of first serial autocorrelation. That is, a characteristic of data in which the correlation between the values of the same variables is based on related objects. In this regard, this study used the D-W test, as a very important facet to show the disintegrations of Tc-99 are negatively autocorrelated. From this perspective, from the experimental design stand-point, first order autocorrelation indicates the presence of experimental errors, which compromise the anticipated results. As mentioned previously, the existence of autocorrelation in applications of time series analyses (as it occurred in the original values of this study) has many deleterious impacts that affect the regression prediction model and the veracity of the results. This is because the interpretation of a faulty objectivistic evaluation that involves the coefficient of determination R^2, the standard error of estimate s, the PRESS and the mean square error (MSE in ANOVA), can be deceptive. As mentioned before, this drawback of autocorrelation violates the assumption of independence, which underlies most of the conventional models in residuals from regression analysis. This hitch can underestimate the *s* (or the statistical term that measures the accuracy with which a sample represents a population) that can look as if the predictors were significant, when in fact they are not. Still further, reviewing the D-W statistic, as stated before, the ideal value of the D-W is 2.0. Here, however, if the value of the D-W statistic is greater or less than the preferred value of 2.0, this situation flags autocorrelation problems and asymmetry of the data.

In this instance, the established data of the disintegrations of Technetium had a value greater than 2.0, e. g., 2.43167; a situation that flagged negative autocorrelation. In any event, the utility of the D-W statistic is very important in the assessments of time series autocorrelation problems. The D-W statistic function is defined below:

$$D = \frac{\sum_{i=2}^{n} (e_i - e_{i-1})^2}{\sum_{i=2}^{n} e_i^2}$$

Where:

D is the D-W statistic
e_i, e^2……, en are sample residuals ordained, according to time
n is the number of observations
The *D* stands for the D-W statistic, and e_i, e^2 are sample residuals ordained according to time

Furthermore, to complement the utility of the regression model, our methodology utilized the analysis of variance (ANOVA) table for regression, to test the null hypothesis that the coefficient of the regression equation is equal to zero. In this instance, the components of the ANOVA table of regression are the Source of Variation, the sum of squares (SSR), error sum of squares (SSE) and the total sum of squares. This table also includes the degrees of freedom, the regression mean square (MSR), the mean square error (the MSE is a very important function that reflects the variability of the data and thus of the magnitude of the experimental error), the F value (the ratio of the mean regression sum of squares divided by the mean error sum of squares) and the *p* value (The *p* value is the probability of obtaining an F value as extreme or more extreme, as the one observed under the assumption that the null hypothesis is true). Here, the F statistics is used to test the null hypothesis that the slope of the regression is equal to zero, e. g. H_o: $\beta = 0$ (meaning there is no linear regression between *Y* and *X*) against the alternative hypothesis H_A:$\beta \neq 0$ (there is linear correlation between *Y* and *X*). Further, about the variables included in the ANOVA table, is the mean square error which is of fundamental importance because it is right there, where the experimental errors are reflected. The interpretation of the MS is that, as this value decreases, the F value increases and the *p* decreases, thus leading to the rejection of the null hypothesis (See Table 2).

MATERIALS AND METHODS

The methodology utilized in this research was focused on the study of the decaying established or original values of Technetium-99. The procedure included the following statements: (1). Usage of the established or conventional data of the disintegrations per minute of Technetium-99, as a function of time, given in books of chemistry (see Table 1 below). (2) Plotting of the decaying conventional disintegrations/min of Technetium-99 values, as a function of time, to show how much of the isotope is being left out, followed by the counterpart curve showing the emission of energy and/or the amount of the isotope oxidized. This situation is depicted in Figure 1. (3) Graphing of the logarithmic transformations (Log_e) of the dependent variable (Disintegrations/min), as a function of the independent variable (time, in hours) to check for the first order reaction law. This state of affairs is shown in Figure 2 below. (4) Description and discussion of the objective (unbiased or factual statistical calculations, as R^2, s, PRESS, ANOVA table) followed by the computing subjective plots (indirect perception of residual graphical schemes) and the calculation of the D-W statistic used to evaluate the established decaying values of technetium-99. This being so, the study utilized the existing or established or conventional data of the Technetium-99 disintegrations according to the table shown below. Next, the method schemed the decaying established disintegrations/min of technetium-99, as a function of time, and its opposite generated emission of energy (or the amount of the isotope oxidized) to analyze its configuration. Figure 1 below shows this situation. Then, the technique prepared a graph by making logarithmic transformations (base e) of the dependent variable (Disintegrations), as a function of the independent variable (time, in hours) to check for the first order reaction law and the identification of possible experimental errors. Figure 2 below shows this setup. For the evaluation of the utility of the distribution of the disintegrations/min of Tc-99, this situation is depicted in Table 2 below.

Table 1. Table showing the established disintegrations per minute of the isotope Technetium-99 data, as a function of time

Disintegrations/min of Technetium-99	Time (hours)
180	0
130	2.5
104	5.0
77	7.6
59	10.0
46	12.5
24	17.5

Source: Theodore L. Brown, H. Eugene LeMay, Jr., Bruce E. Bursten. Chemistry. The Central Science (2004). Page 863. (Published with permission). [13]

Table 2. Printed objectivistic statistical diagnostics of the original or traditional values of the disintegrations of Technetium-99, as a function of time

The regression equation is: Ln Disintegrations = 5.18975 - 0.112643 (Time)

Predictor	Coef	SE Coef	T	P	VIF
Constant	5.18975	0.02265	229.10	0.000	
Time	-0.112643	0.002351	-47.92	0.000	1.000

s	0.0346974
R-Sq	99.8%
R-Sq(adj)	99.7%
PRESS	0.0165970
R-Sq(pred)	99.40%

Source	DF	SS	MS	F	P
Regression	1	2.7642	2.7642	2296.06	0.000
Residual Error	5	0.0060	0.0012		
Total	6	2.7703			

Durbin-Watson statistic = 2.43167

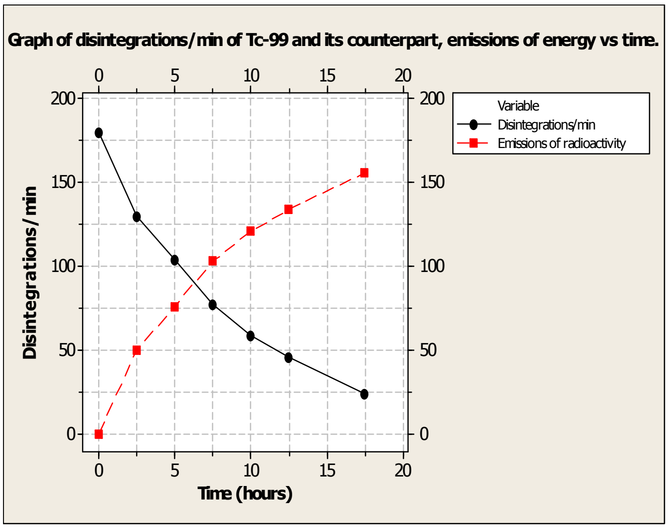

Figure 1.

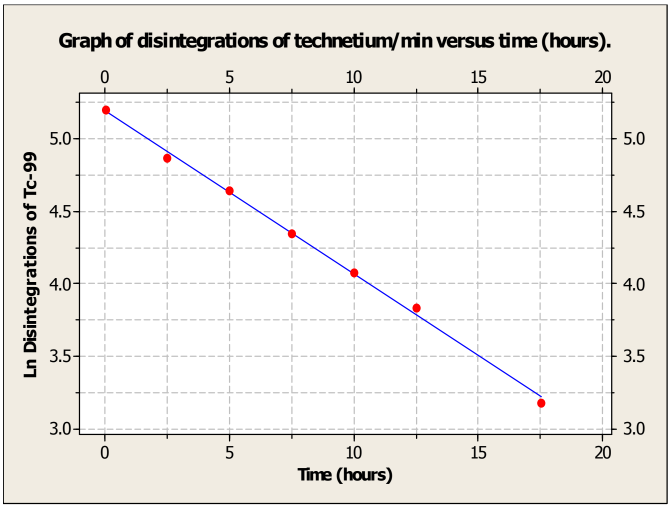

Figure 2. Graph of the Ln of the disintegrations/min of Technetium-99, as a function of time corresponding to the traditional values depicted in Table 1. In this graph, it looks that the biggest differences occurred at 17.5, 12.5 and 2.5 hours

RESULTS

The resulting graph utilizing the established values of the distribution of the disintegrations per hour of Technetium-99 is depicted in Figure 1. In this Figure, it is seen the classical exponential decay of this isotope, as a function of time. This Figure also shows the opposite curve denoting the emission of radioactivity and/or the amount of the isotope oxidized. Likewise, Figure 2 shows the follow-on graph by using the natural logarithmic transformations of the disintegrations of Tc-99 data. We did this task to check for the first order reaction that the data should follow. Here, however, a close scrutiny of Figure 2, clearly shows that in the graph of the logarithmic transformation of the data values of the disintegrations/hr of Technetium-99, as a function of time of the established values, many points are not exactly placed on the regression line, as they should be, since the decaying process of this isotope is a first order reaction. This observation is more notorious in times of 12.5, 17.5 and 2.5 hours. This situation is flagging experimental errors, because there seems to be variations in the measurements of the established or traditional values of the disintegrations of Technetium-99, as described in Table 1. From the experimental design standpoint, this state of affairs suggests there are experimental burdens in the traditional calculations of the disintegrations of Technetium-99. This finding could question the preciseness of the traditional clinical applications of the isotope Technetium-99. This is why medical investigators should revise the traditional values of the disintegrations of Tc-99 aimed at clinical applications of Technetium-99m, so to make the data more precise (if they have not done so). Furthermore, as shown in Figure 3 these printouts show the results of the objectivist results as the coefficient of determination, R^2 = 99.8%, the standard error of estimate, s = 0.0346974 and the statistic, PRESS = 0.0165970. In this instance the ideal value of R^2 is 100% and the ideal values of *s* and PRESS are very close to zero. Moreover, this figure shows the results of the ANOVA table. Here however, we contend the diagnostic objectivist resultant values, could be questionable, because the data values are negatively auto correlated. But most importantly, Figure 3 also shows the resultant value of the Durbin-Watson statistic, which turned out to be 2.43167.

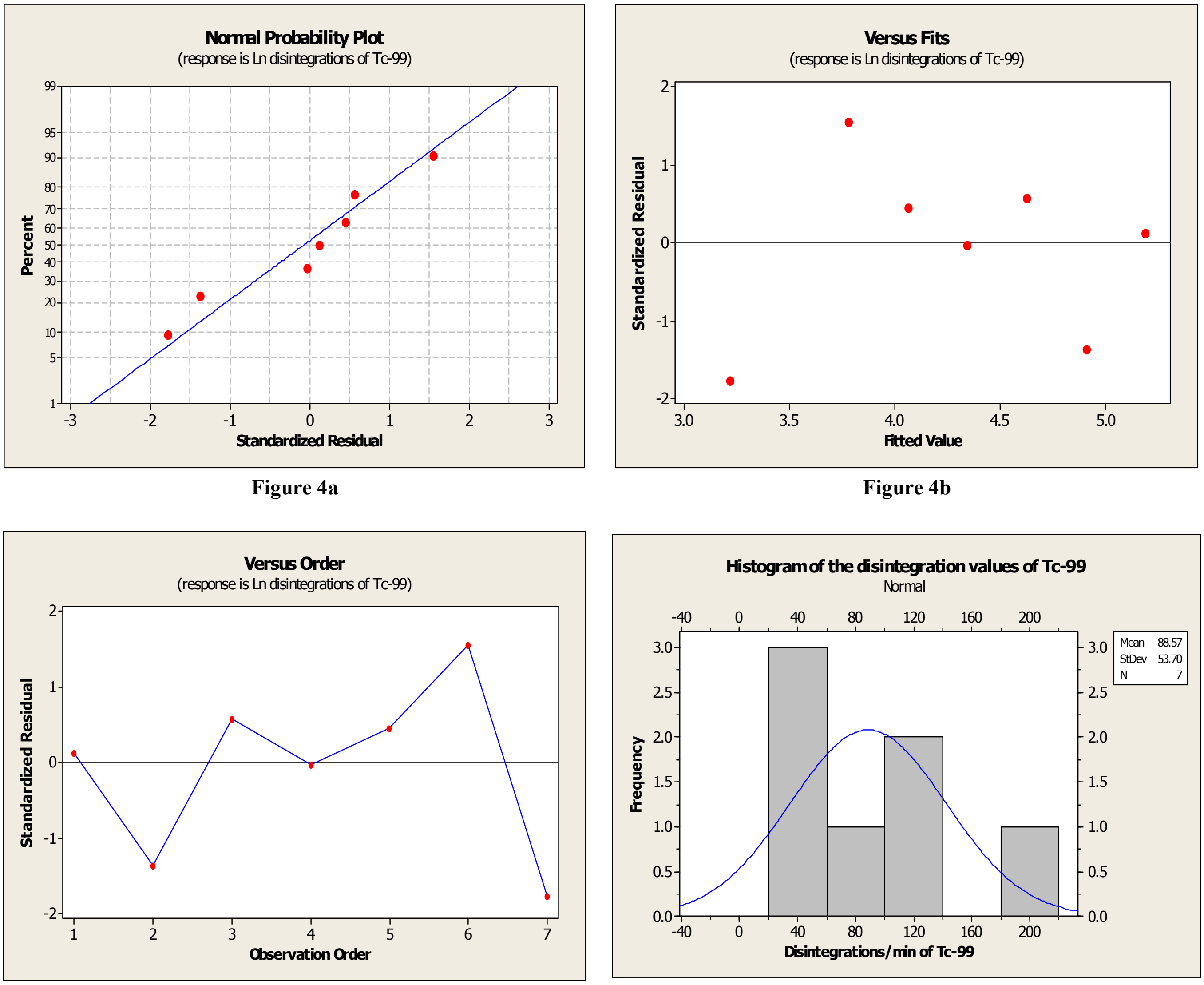

Figure 4a

Figure 4b

Figure 4c

Figure 4d

Figure 4. Figures 4a, 4b, 4c, 4d showing the subjectivist residual graphs of the normal probability plot (Figure 4a), Fitted value (Figure 4b), Observation order (Figure 4c) and Histogram (Figure 4d)

In this stance, the resultant value of the D-W statistic was greater than the ideal value of 2.0. In this instance, according to *Investopedia*, the Durbin-Watson (1919) statistic (DW or d) is a test for autocorrelation in the residuals from a statistical regression analysis. The D-W statistic will always have a value between 0 and 4. A value of 2.0 means that there is no autocorrelation detected in the sample. Values from 0 to less than 2 indicate positive autocorrelation and values from from 2 to 4 indicate negative autocorrelation (*Investopedia*, 2019). From our view, the D-W is a commonly used diagnostic used to test for the presence of first-order serial autocorrelation in the error of a time-series regression model. If its value is less than 2, it means the data is negatively autocorrelated. (A situation that occurred in this research). Otherwise, if its value is greater than 2, it means the data in positively autocorrelated. So according to our reasoning, the disintegrations of Tc-99 are negatively autocorrelated. Moreover, bestowing to the Twain Sun Yat-Sen University, this source furnishes a detailed description of tests of hypothesis considered in the Durbin-Watson Statistics. In addition it gives the table of the critical values of the D-W statistics. Added to this logical reasoning, this research went on step ahead by introducing a possible advanced resulting observation to detect first order serial autocorrelation. This contention is explained by the fact that, if one analyzes the configuration of Figure 2 of the graph of the Ln of the disintegrations/hr of Technetium-99, as a function of time corresponding to the traditional values depicted in Table 1, not all the points are placed exactly on the regression line, as it should be. This is right, because the disintegrations of Tc-99 follow a first order reaction. In this connection, this research contends that first order autocorrelation faults can be detected instantly by observing the number and position and distance of the point values below and above the regression line. Thus, if there are more point values below the regression line, this results in a first order negative autocorrelation (as it occurred here). The opposite is also true which would result in a positive autocorrelation. In this manner, the results showed that the biggest differences occurred at 17.5, 12.5 and 2.5 hours. Indeed, there were more distant point values below the regression line, than above, which resulted in a first order negative autocorrelation. Contrariwise, Figure 4, which includes figures 4a, 4b, 4c and 4d show the resulting corresponding evaluation of the original values of the disintegrations of Technetium-99, utilizing subjectivist residual plots. By analyzing these residual plots, we observe that, in the normal probability plot (Figure 4a) many points are not close to the least square line and most of them are below this line. This situation suggests the data of disintegrations is not normally distributed. Analogously, by analyzing the fitted value plot (Figure 4b), it is seen that there are not the same number of positive and negative standardized residuals. In fact, there are more positive than negative residuals. This situation, again, suggests a negative autocorrelation. Moreover, by analyzing the histogram (Figure 4d), it is seen the distribution data is skewed to the right, a situation that further flags a negative autocorrelation, thus, inaccurate results. Furthermore, in the plotting of the logarithmic transformed values, to witness the first order reaction (Figure 2), it is observed that there are three noticeable values (two below the regression line and one above; a situation that forecasts a negative skewness of the distribution (Mean > Median > Mode) of the disintegrations values of Tc-99, as seen in Figure 4d.

Conclusions and recommendations

This study used experimental design statistical arguments in an effort to show the actual calculations of the disintegrations of Technetium-99 are not very accurate. To substantiate this finding, a close examination of the plotting of the logarithmic transformed values, to attest the first order reaction of the data of Figure 2, suggests the data is not following a strict first order reaction, as it should be, because all the point values are not placed exactly on the least square line. A close scrutiny of Figure 2 shows that the biggest flaws occurred at 17.5, 12.5 and at 2.5 hours. This situation suggests the existence of experimental errors. Moreover, the value of the Durbin-Watson diagnostic equal to 2.43 is far from the ideal value of 2.0, a state of affairs dwindling experimental errors. Besides the complementary graphical diagnostics of the residuals, cast some doubts about the perfectness of the model of the disintegrations of Tc-99. This is especially true with the histogram which is skewed to the right, meaning the distribution data is negatively autocorrelated. Besides, in the normal probability graph, many points are not close to the regression line, a situation flagging asymmetry of the data. In addition, the plot of the residual fits is not randomly distributed and with more positive values than negative values, situations that preclude negative autocorrelation. Moreover, the resulting objectivist diagnostics of the R^2, s, PRESS, and ANOVA table may not be truly reliable, because of the autocorrelation problems discussed hitherto, that could make them ambiguous. In this connection, bestowing to the organization *Statistiska of Stockholms Universitet*, this source of information discusses the consequences of autocorrelation, by affirming that, when the disturbance term is seriously correlated, then the least square estimation is unbiased. This source adds that, under these circumstances, the variance of random terms may be seriously underestimated. However, in our research, even though the autocorrelation of the disintegrations of Tc-99 is not posing a very serious problem, we cannot afford to take any chances, when we speak about the clinical applications of Technetium-99m. This is the reason why this study recommends a strict revision of the actual model of the disintegrations of Technetium-99 aimed to improve the medical applications of this isotope. Thus, to improve the actual calculations of Tc-99 disintegrations, this study recommends an increase of the mean sample size of the disintegrations of this isotope, so to thwart the existing experimental noise. In this way, by increasing the random sample size, of the sample means of the disintegrations of Tc-99, the results will be in compliance with the principles of the Central Limit Theorem. As mentioned previously, according to this Theorem, for a stochastic model to be good, the sample size distribution of the means must be sufficiently large ($n \geq 30$ random sample mean cases), provided the parental population is normal or nearly normal. In this regard, the CLT argues that the plotting the distribution of the mean of the means will resemble the normal distribution and the precepts of the CLT will be fulfilled, with no experimental noise. If, however, there is variation or a small mean sample size (as the small sample distribution of the disintegrations of Tc-99 of this study), the CLT principles will not be met and there will be experimental errors. Thus, this circumstance will yield inappropriate results and the clinical applications of this isotope will not be optimal. It is for this reasoning that this research recommends a drawing of a sufficiently mean sample size of the disintegrations of Tc-99, so to be in accord with the principles of the CLT, hence, to secure more exact uses in the clinical applications of this radioisotope.

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