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RELATIONSHIP OF ^{220}Rn AND ^{222}Rn PROGENY LEVELS IN CANADIAN UNDERGROUND U MINES

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RELATIONSHIP OF ^{220}RN AND ^{222}RN PROGENY LEVELS IN CANADIAN UNDERGROUND U MINES

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ABSTRACT

Radon-222 and ^{220}Rn progeny are found in some Canadian underground U mines. Because both can contribute to lung dose, their experimental determination is important. The relationship between ^{222}Rn progeny Working Level, WL(Rn), and thoron progeny Working Level, WL(Tn), has been investigated in U mines. Experimental measurements extended over the period 1981 to 1986, and consisted of about 700 measurements of each WL(Rn) and WL(Tn). The data were analyzed by standard linear and power function regression analysis. A power function relationship between WL(Rn) and WL(Tn) seemed to fit the experimental data best. The relationship obtained is of practical interest as it permits the calculation of WL(Tn) from experimental values of WL(Rn). The relationship is useful for lung dose calculation purposes and in mine ventilation engineering calculations.

INTRODUCTION

Radon-222 gas and its short-lived progeny are found in relatively high concentrations in operating underground U mines throughout the world. Radon-220 gas and its short-lived decay products, a less common occurrence than ^{222}Rn gas and its progeny, are also found in some underground uranium mines in Canada and other countries.

It is a commonly accepted practice in Canada, as well as in other nations, to estimate ^{222}Rn progeny concentration alone for dose calculation purposes. However, because ^{220}Rn progeny, as well as ^{222}Rn progeny, are found in many U mines, it is important to estimate both concentrations for the purpose of controlling and estimating exposures and doses.

The accurate measurement of ^{222}Rn and ^{220}Rn progeny in ^{222}Rn/^{220}Rn mine atmospheres is not straightforward. It is a lengthy and time consuming task that severely limits the number of samples that can be used for personal dosimetry and ventilation engineering purposes. Because of this it is important to determine whether there is a relationship between ^{222}Rn progeny and ^{220}Rn progeny that can be used to derive one variable from the other with reasonable accuracy.

Because of the short half-life of the ^{222}Rn progeny compared with the relatively long half-life of the ^{220}Rn progeny, the ^{222}Rn progeny are estimated first. Hence, the question is whether ^{222}Rn progeny measurements allow a reliable estimation of ^{220}Rn progeny under average operating field conditions.

This paper presents data on the relationship between ^{222}Rn progeny and ^{220}Rn progeny taken over an extended period of time at several locations in a Canadian underground U mine or U mines with similar mass ratio ^{238}U/^{232}Th and ventilation characteristics.

THEORETICAL BACKGROUND

Measurements of ^{222}Rn progeny and ^{220}Rn progeny in ^{222}Rn/^{220}Rn underground U mine atmospheres vary widely in complexity according to the accuracy required and the radiation quantities of interest. A precise, simultaneous, activity concentration measurement of the short-lived decay products of ^{222}Rn (^{218}Po, ^{214}Pb and ^{214}Po) and the short-lived decay products of ^{220}Rn (^{212}Pb and ^{212}Bi) can be carried out using five gross α counts of a single filter sample (Zh83). This technique also enables the estimation of another useful variable: the Working Level (WL), a variable related to the potential alpha energy concentration (PAEC) in air. The PAEC is the potential α energy concentration corresponding to any mixture of the short-lived decay products of ^{222}Rn or ^{220}Rn present per unit volume of air. The special unit 1 WL corresponds to the ultimate release of α energy of 1.3 MeV L^{-1}. The relationship between 1 WL unit and the PAEC in SI units is: 1 WL = 20.8 μJ m^{-3} (ICRP81).

If the progeny concentrations are not necessary and only the Working Level is the variable of interest, simplified counting procedures can be used. Routine measurement of ^{222}Rn progeny Working Level, WL(Rn), and thoron progeny Working Level, WL(Tn), can be conducted using two gross α count methods. The methods essentially consist of the following steps.

a) a sampling period. For moderate activity concentrations, the sampling period is in the range 3-10 min;

b) a waiting period of 40-90 min after the end of sampling followed by an α counting period of 1-10 min; and

c) a waiting period of 5-11 h after the end of sampling followed by an α counting period of 5-10 min.

The sampling and α counting periods suggested above depend mainly on the ^{222}Rn progeny and ^{220}Rn progeny concentrations. For low and very low concentrations, the sampling and counting periods could be substantially longer than those indicated above.

Steps (a) and (b) describe the Kusnetz method (Ku56) for determining WL(Rn). Steps (a) and (c) correspond to the Rock method (Ro70) for estimating WL(Tn). A more general and detailed method that can be used for both WL(Rn) and WL(Tn) has been developed by the author (Bi84).

The time delay between steps (b) and (c) is necessary to allow essentially complete decay of the ^{222}Rn progeny before counting the ^{220}Rn progeny. However, it should be noted that because of the relatively long half life of the ^{220}Rn progeny, the α count obtained in (b) requires correction to take into account the presence of ^{220}Rn progeny. Calculation of WL(Tn) was done assuming a ^{212}Bi to ^{212}Pb disequilibrium ratio of 0.5. This value is approximately representative of underground conditions in the mine locations where the samples were taken.

Knowledge of WL(Tn) and WL(Rn) is not only of interest for the estimation of radiation exposure and dose, but the ratio WL(Tn)/WL(Rn) represents a good indicator of air flow conditions, i.e., ventilation characteristics, in underground uranium mines (Bi85).

The values of WL(Tn) and WL(Rn), and hence their ratio, in partially enclosed radioactive environments such as those corresponding to underground uranium mines depend on various factors including:

a) the mass ratio ^{238}U and ^{232}Th;

b) air flow conditions at the location;

c) physical characteristics of the U and Th bearing ore;

d) environmental factors such as barometric pressure;

e) other factors, such as plate-out of progeny on mine walls and other surfaces; and

f) mining operations.

Items (a), (b), (c) and (f) depend on the location in the mine. Because of this, the ratio WL(Tn)/WL(Rn) should be expected to vary from one mine location to another. In general, however, reasonably representative average values for the mass ratio $^{238}U/^{232}Tn$ and the physical characteristics of the ore can be assumed. Furthermore, if the ventilation characteristics of the mine are not drastically changed over time, one might anticipate the ratio WL(Tn)/WL(Rn) to either remain approximately constant or to vary within the limits of experimental error in some predictable fashion, at least within the acceptable error for the estimation and control of radiation exposures.

Because different underground U mines may have significantly different ore grades, and hence different mass ratios $^{238}U/^{232}Th$ and ventilation characteristics, the relationship between WL(Rn) and WL(Tn) could vary substantially from mine to mine. For example, measurements in Ontario (Canada) uranium mines show a ratio of WL(Tn)/WL(Rn) of about 0.5 to 0.8, and sometimes higher, whereas mines in Saskatchewan (Canada) indicate WL(Tn)/WL(Rn) to be approximately 0. Hence, the conclusions derived from experimental data from one mine would not be readily applicable to other mines.

A theoretical derivation of the relationship between WL(Rn) and WL(Tn) under field conditions is beyond the scope of this paper. The complexity of diffusion and transport mechanisms of radioactive gases through inhomogeneous and anisotropic media under varying air flow and barometric pressure conditions precludes the treatment of the problem here. Hence, only a useful empirical relationship is sought. However, for a relationship between two variables to be of practical interest and use, the relationship should be preferably a simple one. Two kinds of relationships have been investigated here, namely a power function and a linear function.

The linear function can be represented as follows:

$$WL(Tn) = m\ WL(Rn) + b \tag{1}$$

The power function is represented by equation (2):

$$WL(Tn) = k[WL(Rn)]^{\alpha} \tag{2}$$

The power function of equation (2) can be transformed as follows:

$$\log WL(Tn) = \alpha \log WL(Rn) + \log k\ , \tag{3}$$

which is a linear function that should plot as a straight line for log WL(Tn) versus log WL(Rn).

In this paper, experimental data on WL(Rn) and WL(Tn) have been collected and analyzed by regression analysis techniques to determine the best data fit according to equations (1) and (2). Furthermore, a comparison has been made between the two relationships to ascertain the best theoretical fit to the experimental data.

EXPERIMENTAL PROCEDURE

Most of the ^{222}Rn and ^{220}Rn progeny WL data presented in this paper was obtained at several locations within the same mine. However, data taken at other local underground U mines with similar ratios of $^{238}U/^{232}Th$ and ventilation characteristics gave similar results. The U and Th mass concentrations in the geological formation where the U mines investigated were located, were as follows: 0.05% Th in the form of ThO_2, and from less than 0.05% to about 0.35%, with an average value of 0.1%, for U in the form of U_3O_8. Hence the average mass ratio of $^{238}U/^{232}Th$ is approximately 2.

Measurements extended mainly over the period 1981 to 1986. The discussion presented in this paper is based on approximately 700 independent measurements (samples) of both WL(Rn) and WL(Tn).

Although, in general, use was made of the Kusnetz (Ku56), Rock (Ro75), and Bigu (Bi84) methods to determine WL(Rn) and WL(Tn), other more elaborate methods also were employed concurrently such as the Thomas-Tsivoglou method (Th72), 5 gross α count methods (Zh83), and α spectroscopic techniques. These methods enabled determination of the ^{222}Rn progeny and/or ^{220}Rn progeny concentration and hence their state of disequilibrium, which is a measure of the ventilation characteristics (e.g., air residence time) of the locations where measurements were conducted. The average disequilibrium ratios for the ^{222}Rn progeny concentrations were approximately 0.6 for the ratio ^{214}Pb/^{218}Po and about 0.4 for the ratio ^{214}Po/^{218}Po. The ratio ^{212}Bi/^{212}Pb was in the range 0.4 to 0.6. A value for the ^{220}Rn progeny disequilibrium ratio of 0.5 was taken. It was not possible to calculate reliable values for the residence time from the above disequilibrium ratios because different mine models predict different residence times for the same disequilibrium data (Bi85). Direct mine air residence time measurements varied according to the volume of the section of the mine under consideration and air flow conditions through the volume. However, on the average, residence times in the range 10 to 60 min were measured.

After calculation of WL(Rn) and WL(Tn), frequency distribution graphs of WL(Rn) and WL(Tn) and of the ratio WL(Tn)/WL(Rn) were plotted. The correlation of WL(Tn) to WL(Rn) was determined by regression analysis using least squares techniques and equations 1 to 3 as shown in the next section.

RESULTS AND DISCUSSION

Figures 1 and 2 show the frequency distributions corresponding to WL(Rn) and WL(Tn), respectively. Figure 3 shows the frequency distribution of the ratio WL(Tn)/WL(Rn). The data of Fig 1 to 3 show that most values for WL(Rn) and WL(Tn) were below 0.3 and that the ratio WL(Tn)/WL(Rn) was relatively high, i.e., between 0.4 and 1.4. Linear and power function regression analysis (by the least squares method) of WL(Tn) versus WL(Rn) experimental data gave the following relationships:

$$WL(Tn) = 0.47\ WL(Rn) + 0.074\ ;\ R = 0.835\ ;\ SEE = 9.0 \times 10^{-2} \quad (4)$$

and $$WL(Tn) = 0.557\ (WL(Rn))^{0.727}\ ;\ R = 0.936\ ;\ SEE = 0.16 \quad (5)$$

where, R and SEE stand, respectively, for the linear correlation coefficient and the standard error of the estimate (i.e., WL(Tn)) for either the linear function of the log transformed power function. The equations used to determine the statistical variables of interest, namely, the slope, the y intercept, the correlation coefficient and the standard error of the y estimate (i.e., WL(Tn)) are given in the Appendix. Some elementary statistical analysis of the data has been summarized in Table 1.

Table 1 shows a broad range of radioactivity concentration values found in the field survey. This is indicative of the wide range of background conditions encountered in the many locations where measurements were taken, such as ventilation conditions and mining activity.

The above equations show that the linear function predicts WL(Tn) = 0.074 when WL(Rn) is 0. This corresponds to a very large value of the ratio WL(Tn)/WL(Rn), which can only occur under either extremely high ventilation conditions, i.e., very 'young' mine air, or for a very low mass ratio of $^{238}U/^{232}Th$, or both. This is a rather unlikely situation. The power function by its very nature predicts WL(Tn) = 0 for WL(Rn) = 0, a seemingly more 'reasonable' result. In addition, statistical correlation data indicate that at least in terms of the correlation coefficient, the power function better describes the experimental data. This is partly offset, however, by the higher standard error estimate of the power function compared to the linear function.

Data predicted by equations 4 and 5 for a range of values of WL(Rn) have been tabulated for comparison purposes and presented in Table 2. It can be seen that agreement of WL(Tn) calculated according to equations 4 and 5 is within approximately 20% for WL(Rn) $\gtrsim 0.12$. For low values of WL(Rn), i.e., <0.07, the linear function predicts significantly higher values than the power function.

Because of the discrepancy noted above, the data were divided into two main groups, namely WL(Rn) <0.07 and WL(Rn) >0.07. Those two groups of data were analyzed independently using the statistical techniques discussed above. The results are summarized in Table 3 and equations (6) to (9):

for WL(Rn) ≥ 0.07:

$$\mathrm{WL(Tn)} = 1.921\ \mathrm{WL(Rn)} - 0.004\ ;\ R = 0.79\ ;\ \mathrm{SEE} = 3.0 \times 10^{-2} \qquad (6)$$

$$\mathrm{WL(Tn)} = 0.65\ \mathrm{WL(Rn)}^{0.767}\ ;\ R = 0.82\ ;\ \mathrm{SEE} = 0.21 \qquad (7)$$

for WL(Rn) >0.07:

$$\mathrm{WL(Tn)} = 0.434\ \mathrm{WL(Rn)} + 0.096\ ;\ R = 0.83\ ;\ \mathrm{SEE} = 8.2 \times 10^{-2} \qquad (8)$$

$$\mathrm{WL(Tn)} = 0.54\ \mathrm{WL(Rn)}^{0.7}\ ;\ R = 0.86\ ;\ \mathrm{SEE} = 0.14 \qquad (9)$$

Comparisons of equations (7), (9) and (5) show that the power function for the three groups of data remains essentially the same and that the correlation coefficient and standard error of the estimate vary, respectively, within the following values: $0.82 < R < 0.94$ and $0.14 < \mathrm{SEE} < 0.21$. However, the linear function for WL(Rn) $\lesssim 0.07$ differs substantially from the data for which WL(Rn) >0.07,as the standard error of the estimate (SEE) in equations (6) and (8) shows. Graphical data are shown in Figures 4 and 5. Figure 4 shows the linear function and power function, i.e., equations 4 and 5, respectively, corresponding to all experimental data. Figure 5 shows the power function (equation 5) in long-log coordinates for all experimental data.

The analysis above suggests that because of:

a) The differences in slope, intercept and standard error estimates between equations 6 and 8 for the two different ^{222}Rn progeny Working Level ranges chosen, and between equations 4 and 6 representing, respectively, the entire data set and the low data range, i.e., WL(Rn) $\lesssim 0.07$;

b) The relative similarity in numerical values of the coefficients k and α (see equation 2) representing the power function corresponding to the low, high and entire range of WL(Rn); and

c) The higher correlation coefficient (although higher standard error also) corresponding to the power function as compared with the linear function, the power representation is slightly favoured over the linear function as it is more readily applicable to any range of ^{222}Rn progeny Working Level values measured in the field.

CONCLUSION

The experimental data presented here show that for the underground uranium mine locations investigated there is a relationship between ^{220}Rn progeny Working Level and ^{222}Rn progeny Working Level. This relationship can either be expressed with adequate approximation by a linear function or a power function. In general, however, the power function is preferred because it is more readily applicable to the entire range of progeny data found under field conditions. From the above it may be surmised that the ^{220}Rn progeny Working Level can easily be estimated from measurements of ^{222}Rn progeny Working Level with adequate accuracy for most practical purposes, including the estimation of exposures and doses to miners and for ventilation engineering purposes.

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Table 1 - Elementary statistical analysis of ^{222}Rn and ^{220}Rn progeny data.

Variable	Mean Value	Standard Deviation (SD)	Standard Error (SE)*	Minimum Value	Maximum Value	Range
WL(Rn)	0.256	0.289	0.011	0.001	2.347	2.346
WL(Tn)	0.194	0.163	0.006	0.004	1.250	1.246
log WL(Rn)	-0.885	0.590	0.023	-3.000	0.370	3.370
log WL(Tn)	-0.897	0.458	0.018	-2.398	0.097	2.495

Note: The total number (n) of air samples taken was 675. Each sample was used to determine both WL(Rn) and WL(Tn).

* Defined as: $SE = SD/n^{1/2}$

Table 2 - Theoretical data calculated with linear and power functions

WL(Rn)	$WL(Tn)_{LF}$* (Linear function)	$WL(Tn)_{PF}$** (Power function)	$\frac{WL(Tn)_{PF}}{WL(Tn)_{LF}}$
0	0.074	0.0	0.0
0.01	0.079	0.020	0.25
0.02	0.083	0.032	0.38
0.03	0.088	0.043	0.49
0.04	0.093	0.054	0.58
0.05	0.097	0.063	0.65
0.075	0.109	0.085	0.78
0.10	0.121	0.104	0.86
0.12	0.130	0.119	0.91
0.15	0.144	0.140	0.97
0.17	0.154	0.154	1.00
0.20	0.168	0.173	1.03
0.25	0.191	0.203	1.06
0.30	0.215	0.232	1.08
0.35	0.238	0.260	1.09
0.40	0.262	0.286	1.09
0.50	0.309	0.336	1.09
0.60	0.356	0.384	1.08
1.00	0.544	0.557	1.02

Note: The indices LF and PF indicate linear function and power function, respectively.

* Calculated according to Equation 4.

** Calculated according to Equation 5.

Table 3 - Elementary statistical analysis of ^{222}Rn and ^{220}Rn progeny data.

Variable	Mean Value	Standard Deviation (SD)	Standard Error (SE)*	Minimum Value	Maximum Value	Range	WL(Rn) Range
WL(Rn)	0.025	0.020	0.002	0.001	0.068	0.067	0 to 0.07
WL(Tn)	0.044	0.049	0.004	0.004	0.250	0.246	"
log WL(Rn)	-1.767	0.400	0.033	-3.000	-1.168	1.832	"
log WL(Tn)	-1.541	0.374	0.031	-2.398	-0.602	1.796	"
WL(Rn)	0.316	0.279	0.012	0.071	2.281	2.210	>0.07
WL(Tn)	0.233	0.146	0.006	0.010	0.948	0.938	"
log WL(Rn)	-0.635	0.336	0.015	-1.149	0.358	1.507	"
log WL(Tn)	-0.716	0.273	0.012	-2.000	-0.023	1.977	"

Note: The total number (n) of air samples taken was 150 for WL(Rn) = 0 to 0.07, and 525 for WL(Rn) >0.07. Each sample was used to determine both WL(Rn) and WL(Tn).

* Defined as: $SE = SD/n^{1/2}$.

LIST OF ILLUSTRATIONS

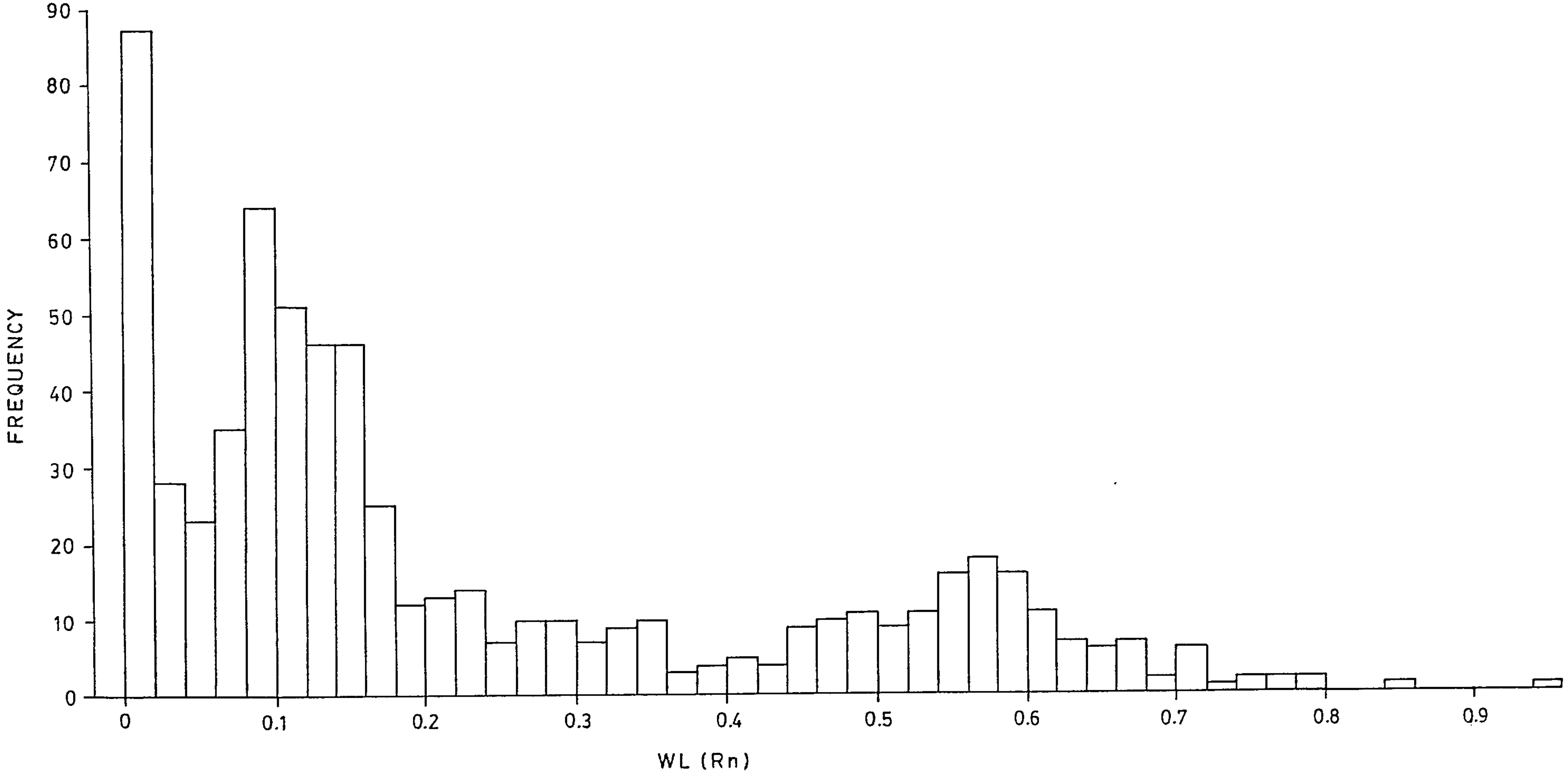
90
80
70
60
50
40
30
20
10
0
FREQUENCY
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
WL (Rn)

Figure 1

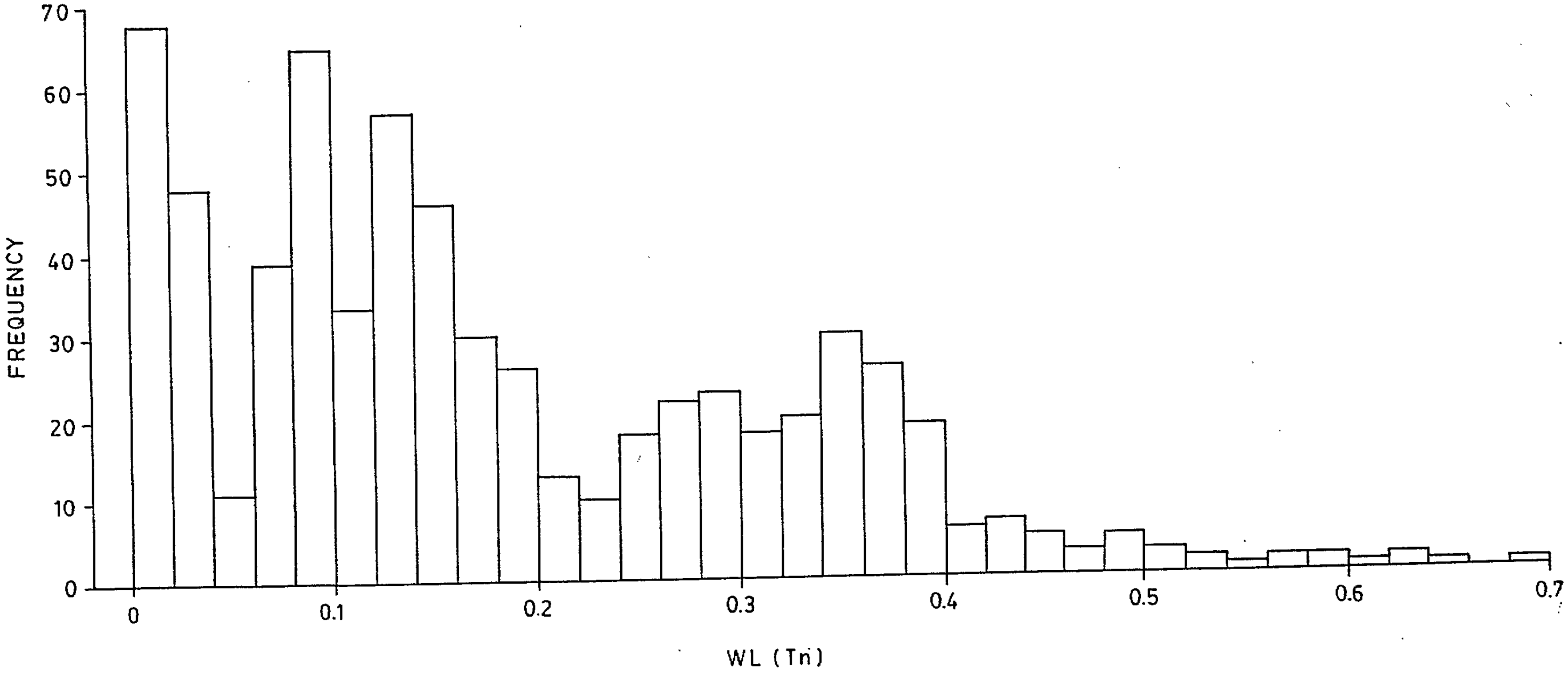

Figure 2

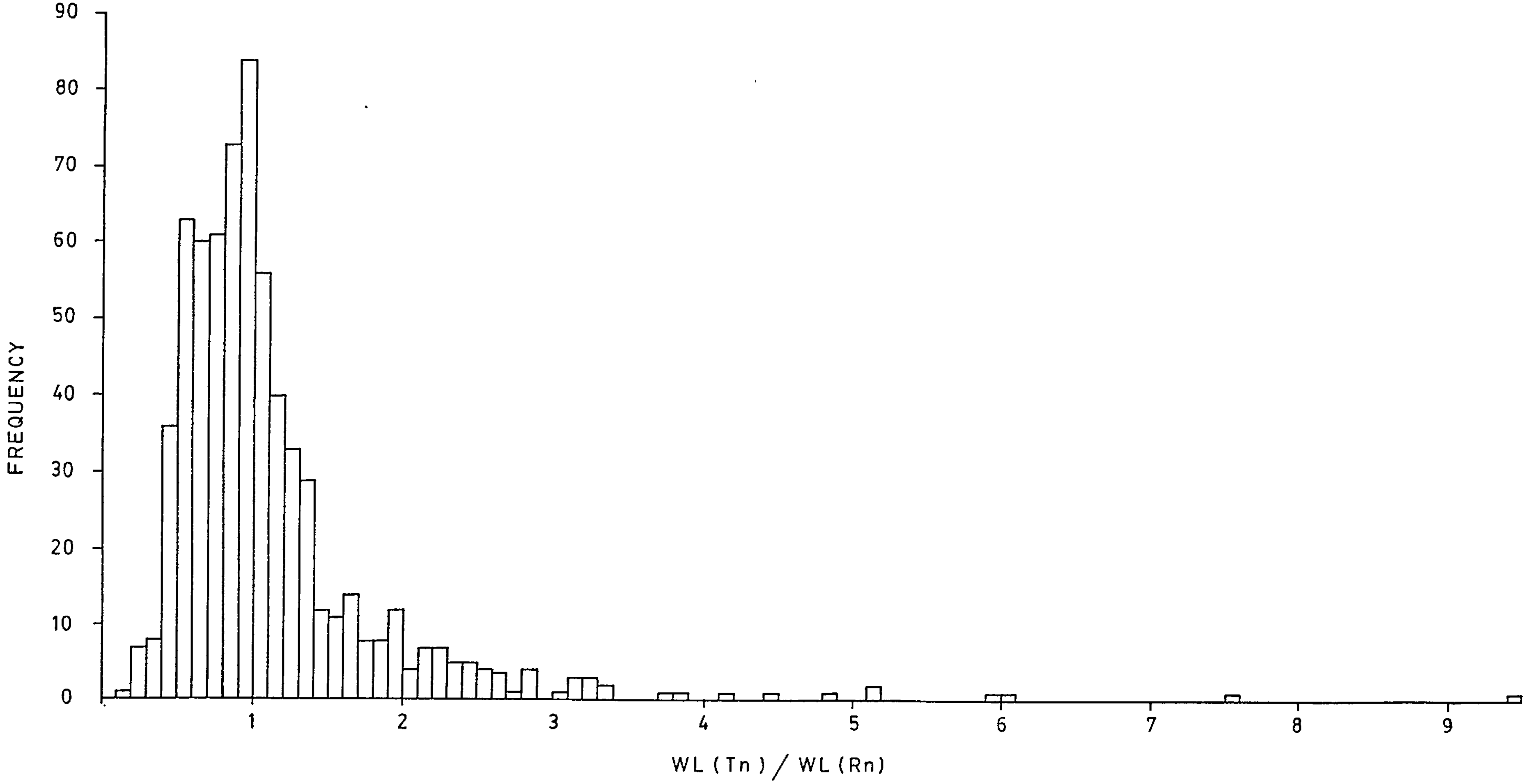

Figure 3

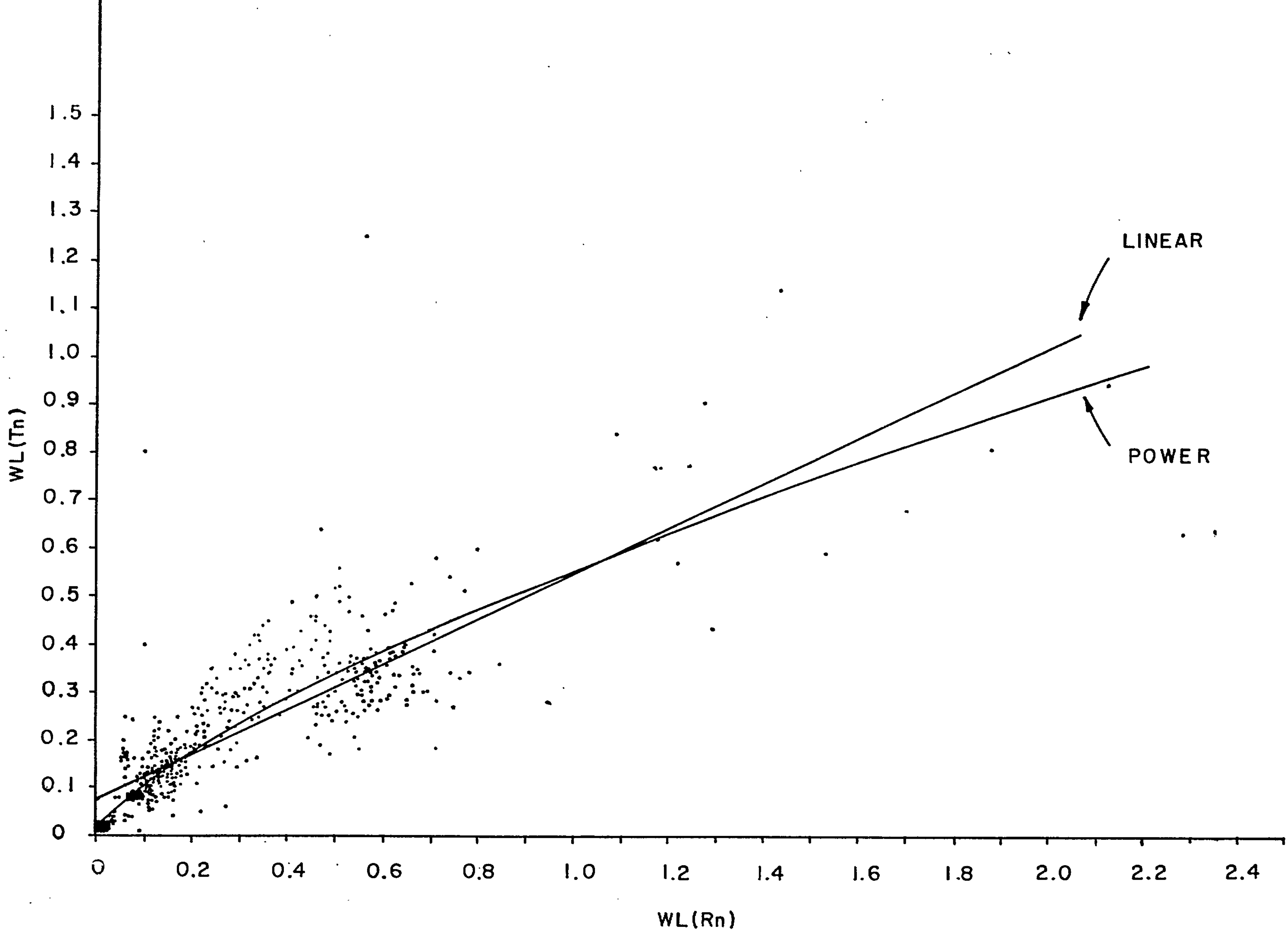
LINEAR
POWER
WL(Tn)
WL(Rn)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.1
1.2
1.3
1.4
1.5
0
0.2
0.4
0.6
0.8
1.0
1.2
1.4
1.6
1.8
2.0
2.2
2.4

Figure 4

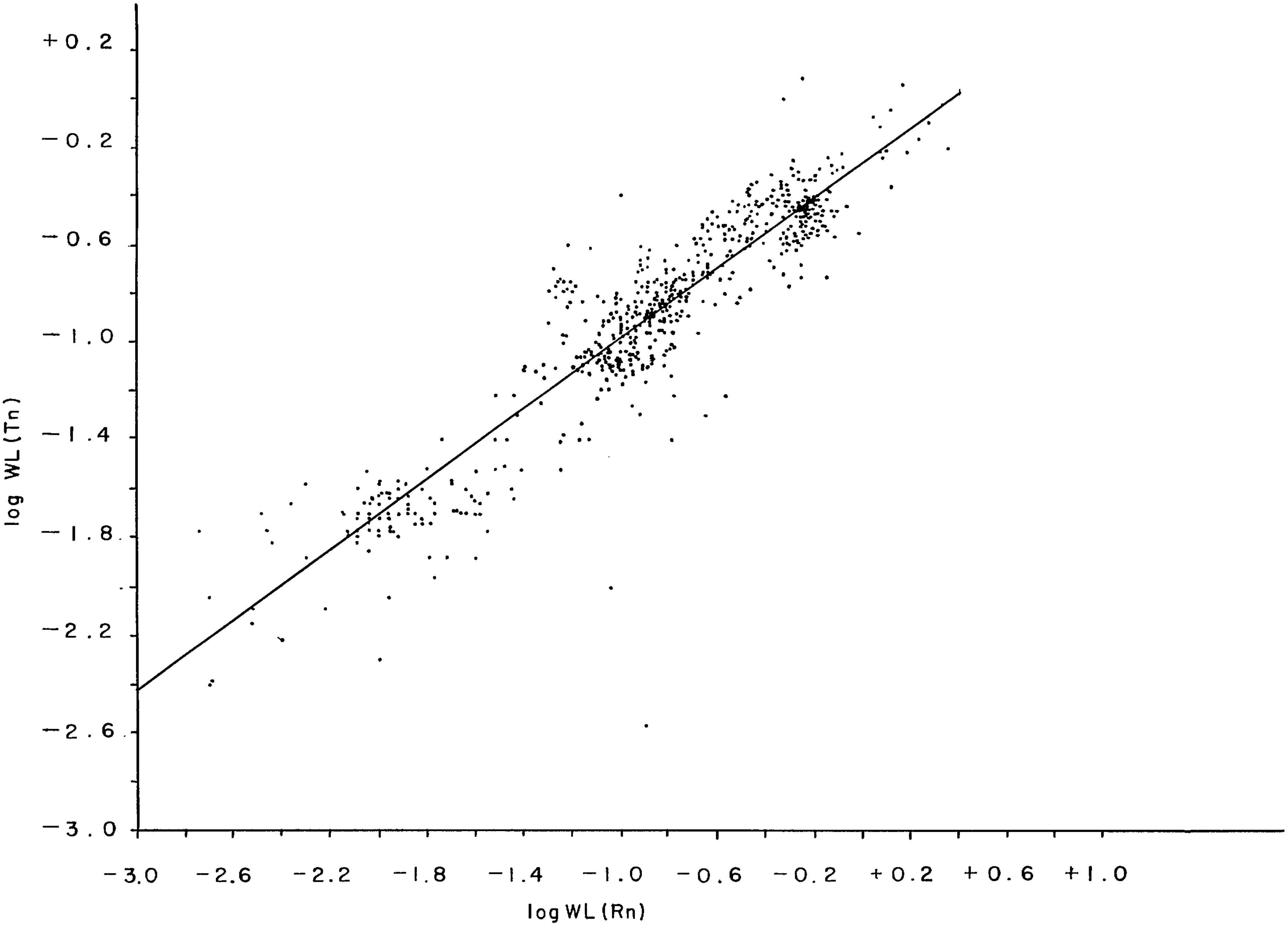
+0.2
−0.2
−0.6
−1.0
−1.4
−1.8
−2.2
−2.6
−3.0
log WL (Tn)
−3.0
−2.6
−2.2
−1.8
−1.4
−1.0
−0.6
−0.2
+0.2
+0.6
+1.0
log WL (Rn)

Figure 5

APPENDIX

Calling x and y the values of WL(Rn) and WL(Tn) measured on each air sample, respectively, and n the total number of air samples taken, the following relationships have been used in the statistical analysis of the data:

$$\text{slope} = \frac{n.\ \Sigma x.y - (\Sigma y).(\Sigma x)}{n.\Sigma y^2 - (\Sigma x)^2}$$

$$\text{y-intercept} = \frac{(\Sigma y).(\Sigma x^2) - (\Sigma x).(\Sigma x.y)}{n.\Sigma x^2 - (\Sigma x)^2}$$

$$\text{SEE} = \left(\frac{\Sigma y^2 - (\text{intercept}.\Sigma y) - (\text{slope}.\Sigma x.y)}{n}\right)^{1/2}$$

$$R = \left(1 - \frac{(\text{SEE})^2}{(\text{SD}(y))^2}\right)^{1/2}$$