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LONG-LIVED ALPHA-PARTICLE ACTIVITY IN DUST SAMPLES FROM HARD ROCK URANIUM MINES

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by

J. Bigu*

ABSTRACT

The long-lived α-particle activity in dusty air samples taken in hard rock underground uranium mines has been measured. Total respirable dust was estimated by conventional weighing techniques. Respirable quartz dust was measured using X-ray diffraction techniques. The long-lived α-particle activity in the samples was measured with α-particle counters (scalers) using a ZnS(Ag) screen optically coupled to a photomultiplier tube. and associated electronic circuitry, as the α-particle detector. Radioactivity measurements were made at least one month after collecting the dust samples to ensure complete radioactive decay of the airborne short-lived decay products of radon and thoron attached to dust particles. The long-lived α-particle activity was plotted against respirable quartz dust content in the samples. A linear relationship was obtained which can be expressed by the equation $A = mW_Q + b$, where A and W_Q represent the α-activity (mBq) and respirable quartz mass (mg) in the sample. respectively. Because measurements of airborne long-lived radioactivity are expensive and time consuming, the data presented here are relevant and useful as they enable the determination of airborne long-lived α-particle activity concentration. a potentially serious occupational hazard in uranium mines. from routine measurements of airborne respirable quartz dust concentration. Practical application of the results submitted here is limited to the mines studied. However. extension of these results to other mines is possible.

Key words: Long-lived radioactive dust: Uranium mines: Quartz dust.

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INTRODUCTION

Mining operations generate dust within a wide size range, namely from submicron size to well beyond the respirable size range (<1-10 μm) (1). Of particular interest, from the occupational health standpoint, is dust in the respirable range because once inhaled a fraction of it will be deposited in the respiratory system (2-5). Dust generated during underground uranium mine operations contains long-lived radionuclides from the natural radioactive decay chains of uranium (^{235}U and ^{238}U) and thorium (^{232}Th).

Although inhalation of dust poses a potential health hazard, inhalation of long-lived radioactive dust (i.e., dust containing long-lived radionuclides) poses an even greater health problem than inhalation of non-radioactive dust (6). Because of this, reliable and accurate methods of quantifying and identifying radioactive dust are of great practical interest. Several radionuclides have been identified in uranium mines dust by α-spectrometry, γ-spectrometry, neutron activation analysis, particle induced X-ray emission, fluorescence and spectrophotometry (7-10). The radionuclides so far identified include ^{235}U, ^{238}U, ^{232}Th and some of their short-lived (radon progeny and thoron progeny) and long-lived (e.g., ^{226}Ra, in mines) decay products (7-10).

The uranium deposits corresponding to the mines where dust sampling was conducted consist of layers and channels of Precambrian, pyrite rich, quartz-pebble conglomerate. The minerals within the conglomerates consist mainly of pyrite, brannerite (titanate of U) and uraninite (oxide of U) which are associated with varying amount of uranothorianite (silicate of U and Th), among other minerals. The U and Th minerals are mainly associated with sulphides (e.g., pyrites). The minerals occur interstitially to the quartz pebbles in the conglomerate matrix. Details on the geology, morphology and composition of the mining can be found in references 11-13.

This paper presents data on the long-lived radioactivity associated with dust in two (hard rock) Canadian underground uranium mines. The idea behind this work was to investigate whether a reasonable relationship could be established between long-lived radioactivity in dust samples and respirable dust mass reliable enough for α-particle dosimetry applications. This is important in view of possible new regulations by the responsible regulatory agencies which may require uranium mining operators to report airborne long-lived α-particle activity for radiation exposure estimation purposes. Because dust mass in air samples from local underground uranium mines is routinely assessed at our laboratory as part of a collaborative agreement, a reliable relationship between long-lived radioactivity and dust mass would greatly facilitate the determination of personnel exposure levels by measurement of dust mass alone. Furthermore, not only could this method be applied routinely in the future, but it could also be used to calculate earlier long-lived airborne radioactivity from past dust measurement records.

EXPERIMENTAL PROCEDURE

Dust samples were taken at two hard rock underground uranium mines at a variety of mine working locations. Dust samples were collected on silver membrane filters (5 μm mean pore size) using gravimetric dust samplers (14) and nylon cyclones. Total dust mass and quartz dust mass in the respirable range (<1-10 μm) were estimated. Total dust mass was measured by weighing the filters before and after the samples were collected. Quartz dust was measured by X-ray diffraction analysis (15).

Alpha-particle activity from long-lived radionuclides associated with dust samples were estimated by gross α-count using α-particle counters with a ZnS(Ag) screen optically coupled to a photomultiplier tube, and associated electronic circuitry, as the α-particle detector. Alpha-particle activity measurements were made not earlier than one month after the dust samples were taken. This procedure was followed to ensure, quite conservatively, complete decay of airborne short-lived decay products of radon and thoron collected in the filter during sampling. It should be noted that the short-lived decay products of radon and thoron arise from the decay of: a) long-lived radionuclides such as ^{226}Rn in dust; and b) radon (^{222}Rn) and thoron (^{220}Rn) in mine air. The short-lived radioactivity from item b) is referred to above as airborne activity, and although collected during sampling is not associated with dust, i.e., item a). Radioisotope identification was carried out by α-spectrometry and γ-spectrometry as discussed elsewhere (7,9,10).

Gross α-activity in each sample was measured as follows. Three sequential 10-min background α-counts were taken followed by three sequential 30-min α-counts with the sample in place. The average values for the background and sample α-count, in counts per second (cps), were calculated, and the difference in α-count, i.e., net α-count, was computed. The net α-count was converted into absolute disintegration rate, in disintegrations per second (dps), i.e., Becquerels (Bq), where, by definition, 1 Bq = 1 dps.

The airborne α-particle activity concentration, A_s, in Bq/m^3, is given by:

$$A_s = 10^3 \; A/QT_s \qquad \text{Eq 1}$$

where, Q is the air sampling flowrate in L/min, and T_s is the sampling time in min. The symbol A is the long-lived α-particle activity measured in Bq in the dust sample. This activity is given by $A = \bar{N}_\alpha/\varepsilon$, where $\bar{N}_\alpha$ is the average net α-particle count in cps, and ε is the α-particle counting efficiency of the α-particle counter used. The efficiency of the counter was determined using a standard ^{241}Am source of activity traceable to the U.S. National Bureau of Standards.

Because of man-power and time constraint considerations, 400 dust samples from two uranium mines (200 samples from each) were selected at random from over 1000 dust samples available taken during 1983-85. This number was reduced to 313, i.e., 76 samples (1983), 121 samples (1984), 116 samples (1985), for the following reasons: a) because the radioactivity associated with the dust collected in the samples was observed to be directly related to either the total respirable dust mass, or to the respirable quartz dust mass fraction, and b) because the grade of the uranium ore in the mines investigated was not particularly high, hence, the α-particle activity measured on the dust samples was rather low. Hence, in order to improve the statistics of α-particle counting, dust samples with total respirable dust mass greater than 0.02 mg were used for the purpose of the present analysis. This represented about 80% of the samples randomly selected for analysis.

The above procedure should not be viewed as introducing a bias into the measurements. The sole reason for the selection of dust mass is the limit of detectability for the variables measured by the particular experimental technique used, i.e., X-ray diffraction for quartz dust and gross α-particle count for long-lived radionuclides. It is our experience that the limit of detection for quartz dust determination by X-ray diffraction is substantially lower than the limit of detection for long-lived radionuclides by gross α-particle counting using a 25 mm diameter ZnS(Ag) screen optically coupled to a photomultiplier tube, and associated electronic circuitry. The counter/detector used here is Model Tri-Met 372A, manufactured by Tri-Met Instruments (Winnipeg, Canada). The α-particle 'background' of ZnS(Ag) detectors are relatively high because of the ever presence of airborne radon and thoron progeny in buildings. Other contributions to background include contamination by radioactive dust due to sample handling, cosmic background, and electronic noise. Other radioactivity techniques could have been used to determine the long-lived radionuclides associated with mine dust with sensitivity comparable to or higher than X-ray diffraction for quartz dust. e.g., neutron activation, particle X-ray emission, and other techniques (7-10). However. these techniques are much more time consuming (and prohibitively expensive) than the simple and straightforward gross α-particle count method.

Dust samples collected at each mine over the period 1983-85 were analyzed independently by linear regression analysis using the least squares method. In addition. dust size distribution measurements were carried out at several mine locations and during a number of mining operations (e.g.. ore transportation. rock crushing, drilling and mucking) using 8-stage cascade impactors, Sierra Model 210, manufactured by Sierra Instruments Inc., (now Anderson), operated at a flowrate of about 13 L/min. These measurements enabled the determination of the dust Mass Median Aerodynamic Diameter (MMAD) (16), and the Activity Median Aerodynamic Diameter (AMAD) (7,9,10). The MMAD and AMAD indicate, respectively. the median of dust mass distribution and the median of the radioactivity distribution associated with the dust. with respect to the aerodynamic diameter.

The accuracy of α-particle measurements depends on the Mass Median Aerodynamic Diameter (MMAD) of the dust cloud, and on the thickness of the dust sample. Large values for both cause appreciable α-particle self-absorption. However. thin samples suggest low α-particle counting, and hence, poor statistics of counting. The work presented here has assumed negligible α-particle self-absorption in the dust samples. This is approximately true as the MMAD was in the approximate range 2-8 μm. depending on the mining operation (7.10). and the thickness of the samples was less than 5×10^{-3} cm. In order to investigate α-particle self-absorption. dust samples were taken at a given underground location under constant air flow rate and different sampling times. The mass range of the samples were approximately the same as those collected during the 1983-85 period. It was found that, within experimental error, there was essentially a constant relationship between α-particle activity and dust mass indicating that self-absorption was not a serious consideration.

EXPERIMENTAL RESULTS AND DISCUSSION

Data collected in two hard rock underground uranium mines are shown in Figures 1 to 5. The two mines are located in the same general geographical area and are designated Mine A and Mine B for the purpose of this paper.

Figure 1 shows the percentage cumulative α-particle activity or dust mass versus the Equivalent Aerodynamic Diameter, EAD($D_{p,50}$), i.e., size of a spherical particle of density 1 g/cm^3 with the same terminal settling velocity as the sampled particle, for typical samples taken during rock crushing operations. The MMAD and AMAD can be obtained from Figure 1 by reading the values of EAD($D_{p,50}$) corresponding to 50% of the cumulative dust mass or α-particle activity, respectively, (7,10,16).

Figures 2 and 3 show, respectively, frequency histograms of airborne respirable quartz dust concentration (mg/m^3) and α-particle activity (i.e., long-lived radionuclides) concentration (mBq/m^3) for Mines A (top) and B (bottom). The graphs indicate that the frequency distributions for these sample populations are somewhat different for the two mines. This is to be expected and is attributed to different mining methods, tools and machinery used by the two mines. For example one mine is highly dieselized (trackless mining), i.e., extensive use of diesel equipment, whereas the other mine uses more conventional mining methods.

Figure 4 shows the long-lived radionuclides activity versus total respirable dust mass measured in the samples selected. The data correspond to samples collected in Mine A for the period 1983 to 1985. Also shown is the best fitted straight line from linear regression analysis. The slope and intercept values found were 18.34 and 12.21, repectively. The errors associated with these values (see Appendix) were, 2.32 and 20.12, respectively. The correlation coefficient found was rather low, i.e., 0.31, indicating a poor correlation between the above variables. Analysis of data from Mine B (not shown) shows essentially the same results. (It should be noted that an identical graph to Figure 3 is obtained if the α-particle activity (mBq) and total dust mass (mg) are substituted by their respective airborne concentrations, i.e., mBq/m^3 and mg/m^3, as the above pair of physical variables are measured for each sample taken.)

Figure 5 shows the long-lived radioactivity associated with the dust samples versus respirable quartz dust mass for the period 1983 to 1985 for Mine B. Also shown is the best-fitted straight line obtained by linear regression analysis. Figure 5 shows that the data can be approximated by the following linear relationship:

$$A(\mathrm{mBq}) = 156.92\ W_Q + 3.44 \qquad \text{Eq 2}$$

where W_Q represents the quartz dust mass in mg. The error associated with the slope m = 156.92 and intercept b = 3.44, were 8.00 and 12.23, respectively. The correlation coefficient (CC) was : CC = 0.756. Details on the statistical analysis are given in the Appendix.

A similar analysis for Mine A gives the following linear relationship:

$$A(\mathrm{mBq}) = 160.25\ W_Q + 7.87 \qquad \text{Eq 3}$$

The correlation coefficient corresponding to Equation 3 is 0.70, a slightly lower value than that obtained for Equation 2.

A simple calculation for $W_Q \simeq 0$ in Equations 2 and 3 shows that a 'residual' activity is obtained ranging from about 3 mBq (Equation 2) to approximately 8 mBq (Equation 3). Taking into account that 1 mBq = 10^{-3} Bq =

10^{-3} dps = 6 x 10^{-2} dpm, where dpm stands for disintegrations per minute, and that the counting rate in counts per minute (cpm) is given by cpm = ε dpm, the above Equations give values for the residual activity of 0.072 cpm and 0.192 cpm, respectively. In this calculation, the counting efficiency of the scalers used was $\varepsilon \sim 0.4$. The values obtained above for the residual activity are within the range of experimental values observed for the background of the (uncontaminated) scalers used. This shows that Equations 2 and 3 for low, or zero, value of W_Q are consistent with experimental background activity measurements.

It should be noted that analytical functions other than linear could be used to examine the data, in fact, other functions were used but with no better results than those given above. However, because of its simplicity, the linear approach is very attractive and clearly preferred from a practical standpoint for routine handling of data by mine personnel.

The data presented here show a much higher correlation ($\gtrsim$0.7) between long-lived radioactivity and respirable quartz dust than between long-lived radioactivity and total respirable dust (C.C. ~ 0.3). This is to be expected as the other major component of total respirable dust in these mines is (combustible) non-mineral dust, e.g., diesel particulates generated by tools, machinery and vehicles used in underground mining operations. However, it should be noted that the correlation found between long-lived radionuclides, i.e., α-particle activity, and respirable quartz dust content in the samples is not meant to suggest that the radionuclides are actually attached or associated with quartz per se because, as it was earlier pointed out, uranium and thorium minerals are in fact associated with other minerals such as pyrites, which in turn are interstitial to the quartz pebbles in the conglomerate matrix. Rather, it is meant that the α-particle activity is associated with the 'grain' matrix containing not only quartz but uranium and thorium minerals associated with pyrites.

CONCLUSIONS

A linear relationship has been found between long-lived radionuclides activity and quartz dust content in samples from two hard rock underground uranium mines. The slope of the correlations, i.e., the specific activity (mBq/mg) of the samples, was very similar for the two mines in spite of the different quartz dust and long-lived radionuclides activity frequency distributions of the samples examined. Although not particularly strongly correlated (C.C. $\gtrsim$0.7), this relationship is suitable for practical applications which do not require a high degree of accuracy. Further analysis of the data shows, however, a rather low correlation (C.C. ~0.3) between long-lived radionuclides activity and total respirable dust, which makes this relationship of limited practical value.

The above linear correlations (Equations 2 and 3) suggest that the airborne long-lived radiactivity associated with respirable dust can be estimated approximately if the quartz content in the sample is known. However, the reader should be aware that, strictly speaking, the above relationships apply only to specific cases, and mine locations, as the long-lived radioisotope content in the ore body depends on the mine location, and the mining operations, e.g., leaching.

Clearly the reader is aware that long-lived radioactivity can be best estimated by methods other than using a correlation between dust and radioactivity as given by Equations 2 and 3. One advantage offered by the method suggested here, however, is that quartz dust measurements are routinely conducted at our laboratory for the mining companies. Hence, it is a simple matter for mine personnel to calculate an approximate value for the long-lived radioactivity in the samples adequate for practical applications.

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LIST OF ILLUSTRATIONS

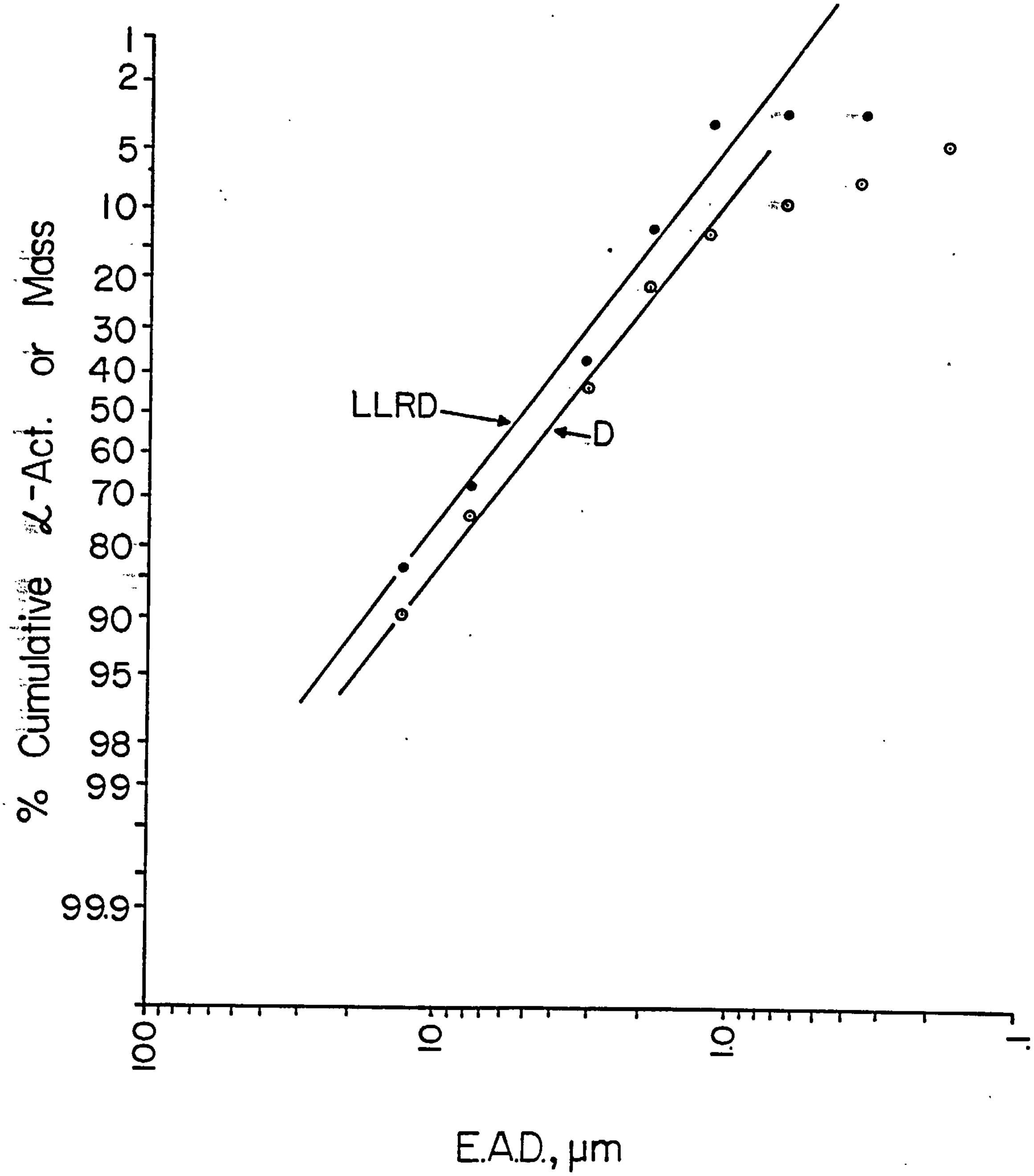

% Cumulative α-Act. or Mass
1
2
5
10
20
30
40
50
60
70
80
90
95
98
99
99.9
100
10
1.0
.1
E.A.D., μm
LLRD
D

Figure 1

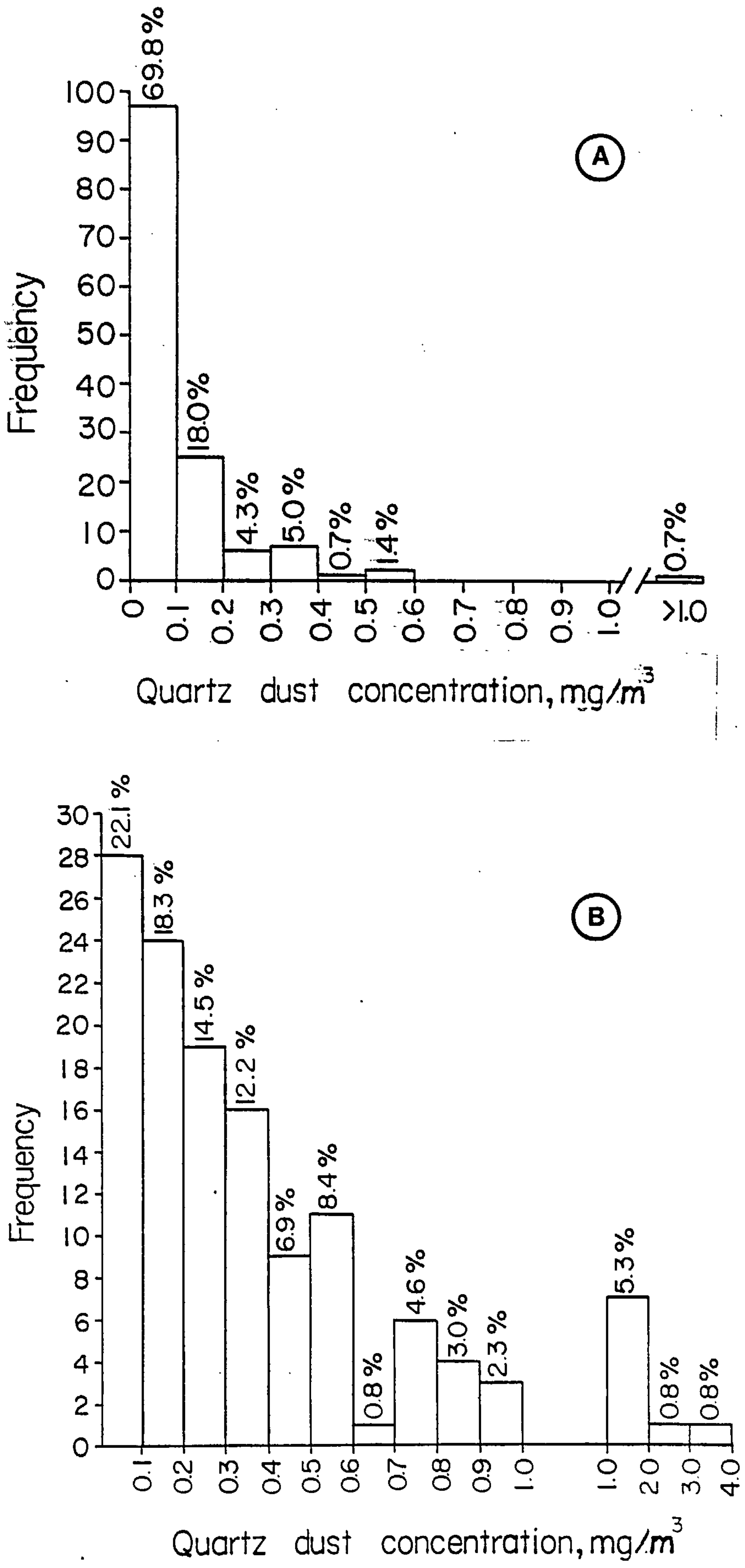
A
Frequency
100
90
80
70
60
50
40
30
20
10
0
69.8%
18.0%
4.3%
5.0%
0.7%
1.4%
0.7%
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
>1.0
Quartz dust concentration, mg/m3
B
Frequency
30
28
26
24
22
20
18
16
14
12
10
8
6
4
2
0
22.1 %
18.3 %
14.5 %
12.2 %
6.9%
8.4%
0.8%
4.6%
3.0%
2.3%
5.3%
0.8%
0.8%
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.0
2.0
3.0
4.0
Quartz dust concentration, mg/m3

Figure 2

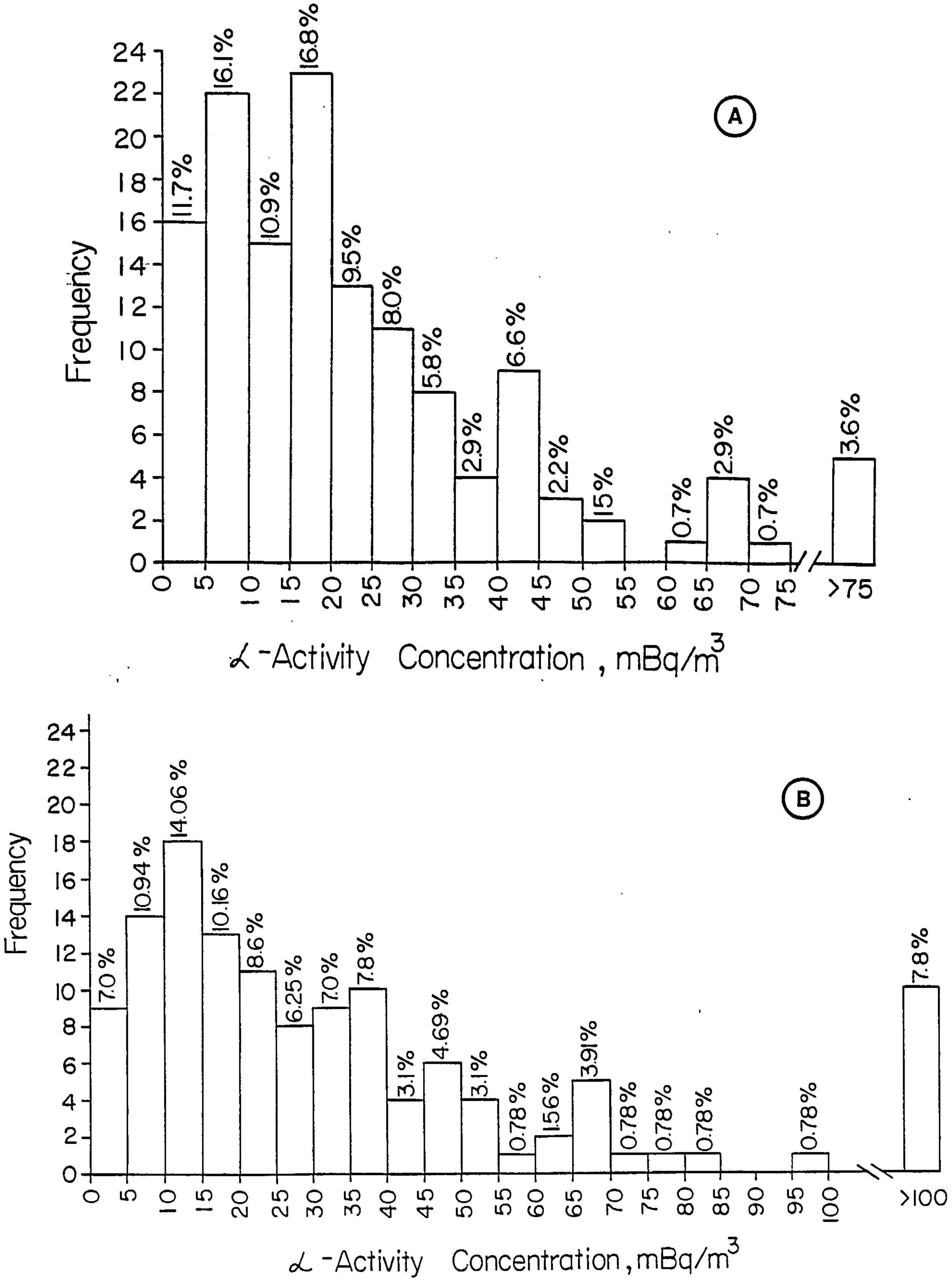
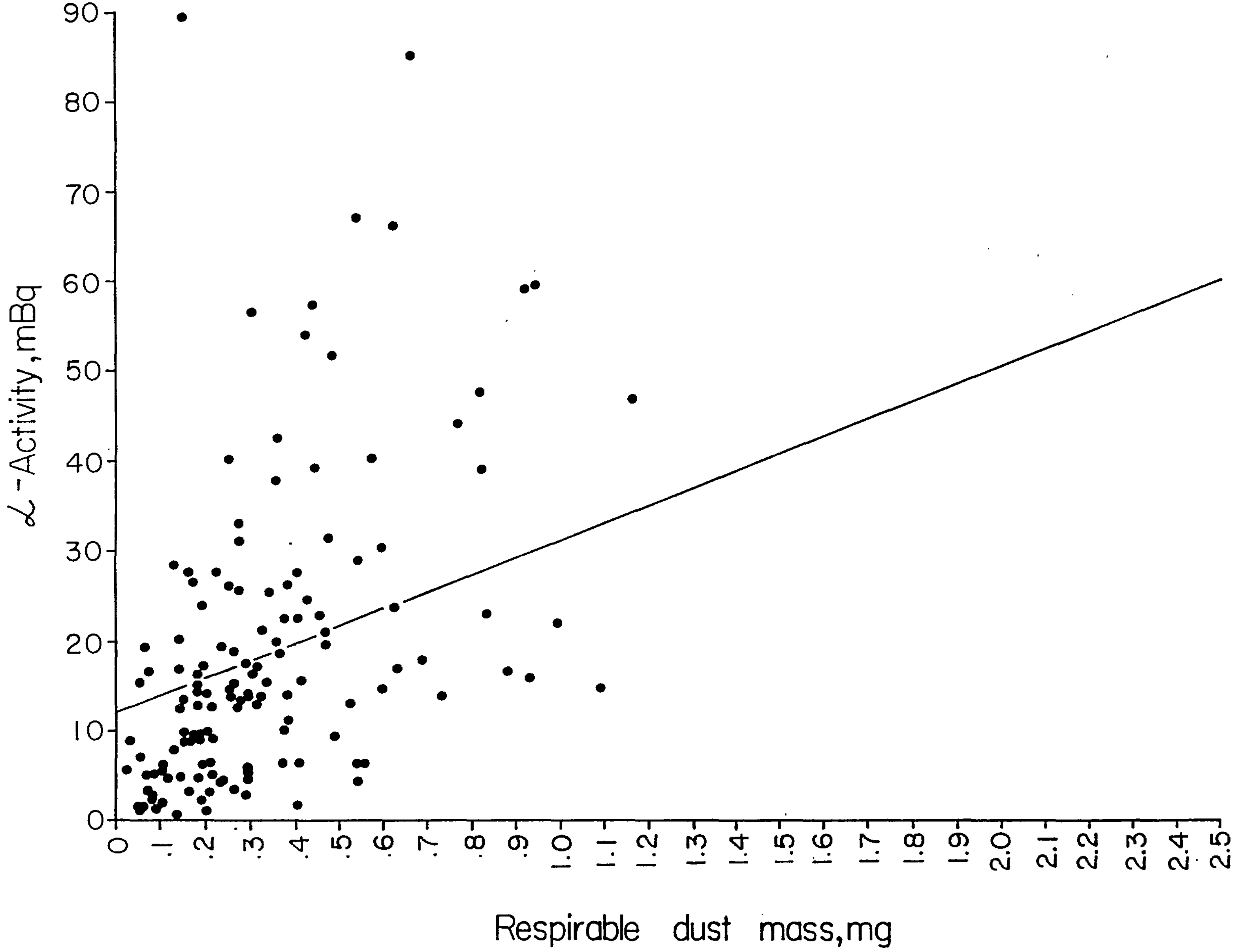

A
Frequency
24
22
20
18
16
14
12
10
8
6
4
2
0
11.7%
16.1%
10.9%
16.8%
9.5%
8.0%
5.8%
2.9%
6.6%
2.2%
15%
0.7%
2.9%
0.7%
3.6%
0
5
10
15
20
25
30
35
40
45
50
55
60
65
70
75
>75
α-Activity Concentration, mBq/m^3
B
Frequency
24
22
20
18
16
14
12
10
8
6
4
2
0
7.0%
10.94%
14.06%
10.16%
8.6%
6.25%
7.0%
7.8%
3.1%
4.69%
3.1%
0.78%
1.56%
3.91%
0.78%
0.78%
0.78%
0.78%
7.8%
0
5
10
15
20
25
30
35
40
45
50
55
60
65
70
75
80
85
90
95
100
>100
α-Activity Concentration, mBq/m^3

Figure 3



90
80
70
60
50
40
30
20
10
0
α-Activity, mBq
0
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
1.1
1.2
1.3
1.4
1.5
1.6
1.7
1.8
1.9
2.0
2.1
2.2
2.3
2.4
2.5
Respirable dust mass, mg

Figure 4

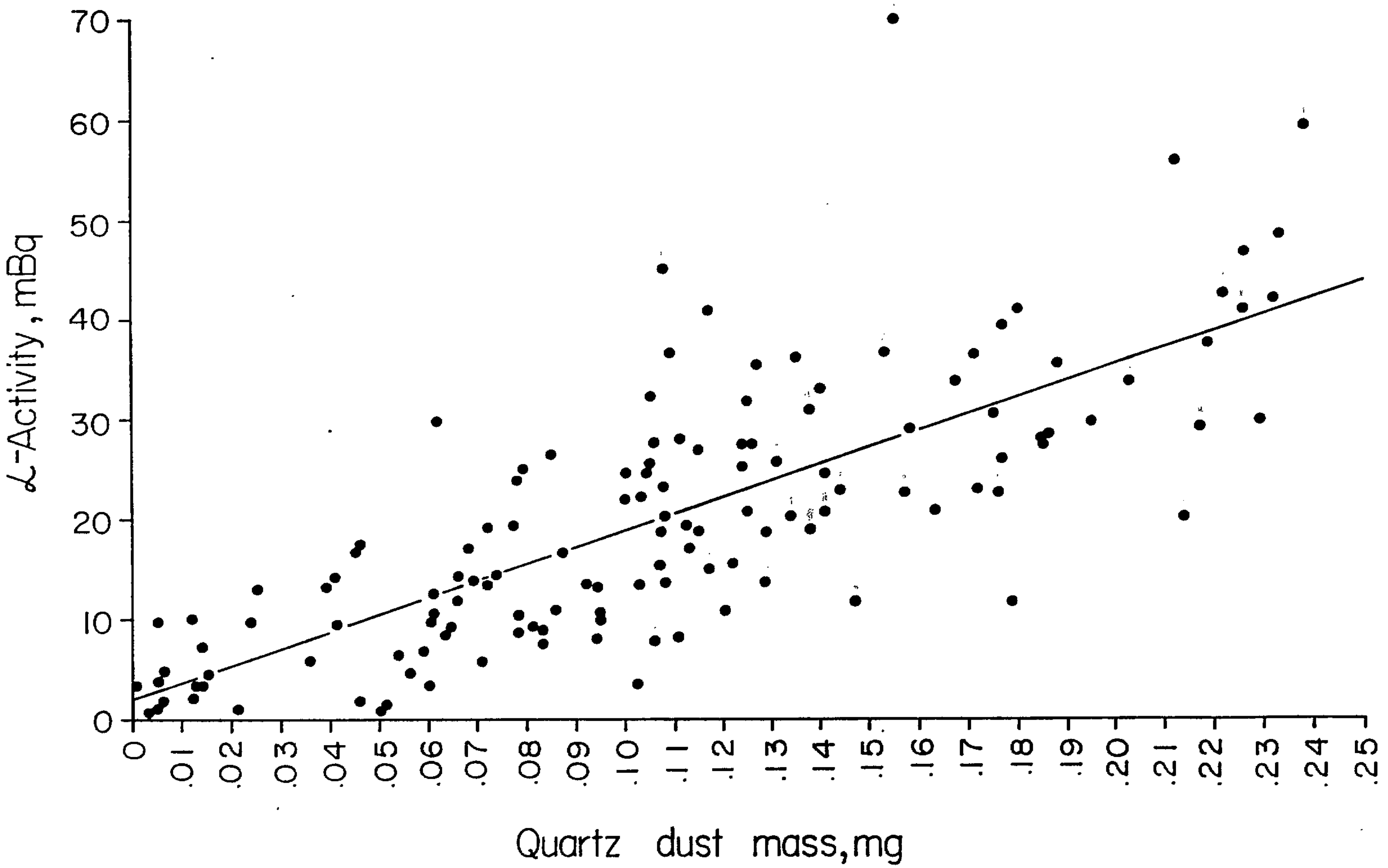

α-Activity, mBq
0
10
20
30
40
50
60
70
Quartz dust mass, mg
0
.01
.02
.03
.04
.05
.06
.07
.08
.09
.10
.11
.12
.13
.14
.15
.16
.17
.18
.19
.20
.21
.22
.23
.24
.25

Figure 5

APPENDIX

Calling x and y the values of W_Q and A 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:

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

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

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

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

where, CC, SD and SEE stand for correlation coefficient, standard deviation and standard error of the estimate, respectively.