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LONG-LIVED RADIONUCLIDES IN QUARTZ DUST SAMPLES FROM HARD ROCK URANIUM MINES

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by

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ABSTRACT

The long-lived radioactivity in dust 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 radioactivity in the samples was measured by conventional α-particle detection methods. The long-lived α-radioactivity was plotted against 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 quartz mass (mg) in the sample, respectively. The data presented are relevant from the occupational health viewpoint.

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

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RADIONUCLÉIDES À LONGUE PÉRIODE CONTENUES DANS DES ÉCHANTILLONS DE POUISSIÈRE DE QUARTZ PROVENANT DES MINES D'URANIUM EN ROCHE DURE

par

J. Bigu*

RÉSUMÉ

On a mesuré le rayonnement à longue période des échantillons de poussière prélevés dans des mines souterraines d'uranium en roche dure. La quantité totale de poussière respirable a été déterminée par des méthodes de pesage classiques. La poussière de quartz respirable a été mesurée à l'aide de techniques de diffraction des rayons X. Le rayonnment à longue période des échantillons a été mesuré par des méthodes classiques de détection de particules α. On a tracé une courbe du rayonnement α à longue période par rapport à la teneur en poussière de quartz des échantillons. Une relation linéaire a été obtenue et exprimée par l'équation $A = mW_Q + b$, où A et W_Q représente respectivement l'activité α (mBq) et la masse du quartz (mg) de l'échantillon. Les données présentées dans le présent rapport sont pertinentes au domaine de la santé au travail.

Mots-clés: Poussière radioactive à longue période; Mines d'uranium; Poussière de quartz.

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INTRODUCTION

Dust emissions from underground uranium mine operations contain long-lived radionuclides from the natural radioactive decay chains of uranium (^{235}U and ^{238}U) and thorium (^{232}Th).

Mining operations generate dust within a wide size range, namely from submicron size to well beyond the respirable size range (1-10 μm). Of particular interest, from the occupational health standpoint, is dust in the respirable range because once inhaled it is deposited in the respiratory system.

Although inhalation of dust poses a potential health hazard, inhalation of radioactive dust poses an even greater health problem. 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 mills and uranium mines dust by α-spectrometry, γ-spectrometry, neutron activation analysis, fluorescence and spectrophotometry. 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.

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 amounts 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.

This paper presents data on the long-lived radioactivity associated with dust in two (hard rock) Canadian underground uranium mines.

EXPERIMENTAL PROCEDURE

Dust grab samples were taken at two hard rock underground uranium mines at a variety of mine working locations. Dust samples were collected on silver membrane (5 μm) filters using gravimetric dust samplers (1) 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.

Alpha-particle activity from long-lived radionuclides associated with dust samples were estimated by gross α-count using standard techniques. Radioisotope identification was carried out by α-spectrometry and γ-spectrometry as discussed elsewhere (2).

Because, (a) the radioactivity associated with dust collected in the samples was observed to be related either to total dust mass, or to a particular fraction(s) of the total dust mass, and (b) the grade of the uranium ore in the mines investigated was not particularly high, the α-particle activity measured on the samples was rather low. Hence, in order to improve the statistics of α-particle counting, dust samples with total dust mass in the range ~0.02 mg to $\gtrsim$0.4 mg were selected for the purpose of the present analysis.

Radioactivity 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, i.e.,

radon daughters and thoron daughters, collected during the dust sampling period. (It should be noted that a contribution to the total α-count from the short-lived decay products of radon and thoron is always present due to the decay of ^{226}Ra and ^{228}Th in the ore.)

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 minute (cpm), were calculated, and the difference in α-count, i.e., net α-count, was computed.

Net α-count, in cpm, was converted into absolute disintegration rate, in disintegrations per minute (dpm). The disintegration rate was converted into activity in mBq, and activity concentration in air (mBq/m^3). The activity, A, is given by:

$$A(mBq) = \frac{16.67}{\varepsilon}\left(\frac{\bar{N}_\alpha}{T_\alpha} - \frac{\bar{B}}{T_B}\right) \quad (1)$$

The activity concentration level, A_S, is given by:

$$A_S = 10^3 A/QT_S \quad (2)$$

where Q is the sampling flow-rate in L/min, and T_S is the sampling time in min. As A is in mBq, A_S is given in mBq/m^3.

In the above expressions, $\bar{N}_\alpha$ and $\bar{B}$ stand for the average sample gross α-count, over time T_α, and average background α-count over the time T_B, respectively. The symbol ε represents the α-count efficiency of the instrument used to measure gross α-count. It should be noted that by definition 1 Bq = 1 dps, where dps = dpm/60, i.e., disintegrations per second.

A total of 313 samples was analyzed, i.e., 76 samples (1983), 121 samples (1984), and 116 samples (1985). The number of samples analyzed per year was decided according to the available range of dust mass suitable for gross α-count measurements, and radioisotope identification by α- and

γ-spectrometry.

Data were analyzed on a yearly basis for each mine. The three independent sets of data corresponding to 1983, 1984 and 1985, respectively, were compared in order to identify possible differences arising from changes that may have occurred in ore grade, mining operations or sampling locations and methods. Finally, the data collected over the 1983-85 period for each mine were analyzed as a single set of data. It should be noted that only part of all the data analyzed is presented here. Data have been selected for particular illustrative purposes.

EXPERIMENTAL RESULTS AND DISCUSSION

Some data collected in two hard rock underground uranium mines are shown in Figures 1 to 5. The two mines were located in the same general area and will be referred to in the future as mine A and mine B.

Figures 1 and 2 show, respectively, frequency histograms of airborne respirable dust concentration (mg/m^3) for mines A (top) and B (bottom). The data correspond to the samples selected for the present study. The graphs indicate that the frequency distributions for these two sample populations were significantly different for the two mines.

Figure 3 shows the long-lived radionuclides activity versus total respirable dust mass measured in the samples selected. The data correspond to selected samples collected in mine A for the period 1983 to 1985. Also shown is the best fitted straight line from linear regression analysis. The correlation coefficient found was rather low, i.e., about 0.45, indicating a poor correlation between the above variables. Analysis of data from mine B (not shown) shows essentially the same results.

Figure 4 shows the long-lived radioactivity associated with the dust samples, against respirable quartz dust mass for mine B. A set of three

graphs are shown in Figure 4 corresponding to 1983, 1984 and 1985, respectively. Also shown in Figure 4 are best-fitted lines by linear regression analysis. Table 1 shows the slopes and intercepts of these lines.

Table 1 shows that the slopes of the lines corresponding to the three periods of time investigated are not significantly different, i.e., about 8% difference between the maximum and minimum values for the slopes. The three linear correlations can, therefore, be considered identical for practical purposes.

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.

$$A(mBq) = 166.88\ W_Q + 2.22 \qquad (3)$$

where W_Q represents the quartz dust mass in mg. The slope m = 166.88 mBq/mg, and intercept b = 2.22 mBq corresponding to Equation 3 for the combined period 1983 to 1985 does not differ significantly from the values obtained for 1983, 1984 and 1985, as shown in Table 1. The correlation coefficient corresponding to Figure 5 was about 0.8.

A similar analysis for mine A shows the following linear relationship:

$$A(mBq) = 160.25\ W_Q + 7.87 \qquad (4)$$

with about the same correlation coefficient as for mine B.

Equations 3 and 4 for the two mines can be considered almost identical for all practical purposes.

A similar analysis to that shown above for total respirable dust, instead of respirable quartz dust, has shown a significantly lower correlation coefficient. It is, therefore, concluded that there is a reasonably good linear correlation between the long-lived radioactivity associated with

airborne dust in the respirable size range, and the quartz dust content in the dust sample. No such good correlation exists, however, for total respirable dust. This is to be expected as the other major component of total respirable dust, in the present case, is non-mineral dust, e.g., diesel particulates generated by tools, machinery and vehicles used in underground mining operations.

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 attached or associated with quartz 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.

The accuracy of α-particle measurements depend on the Mass Median Aerodynamic Diamater (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 thickness of the samples was less than about $1.0 \times 10^{-2} - 5.0 \times 10^{-3}$ cm, and the MMAD was in the approximate range 2-8 μm, depending on the mining operation.

CONCLUSIONS

A linear relationship has been found between long-lived radionuclides 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 significantly different quartz dust and long-lived radionuclides activity frequency

distribution selected for the analysis of the samples (see Figures 1 and 2).

The above linear correlations (Equations 3 and 4) suggest that the airborne long-lived radioactivity associated with respirable dust can be estimated approximately if the quartz content in the sample is known, and vice versa. 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 operation, e.g., leaching.

The data presented in this paper are of practical interest from the occupational health viewpoint.

ACKNOWLEDGEMENT

The author is grateful to the mining companies for their interest in this work and for providing air flow data and exposure (i.e., sampling) time data corresponding to the samples investigated here. The author would like to acknowledge the assistance of D. Irish (Co-op student, Waterloo University) in conducting some of the long-lived radioactivity measurements.

REFERENCES

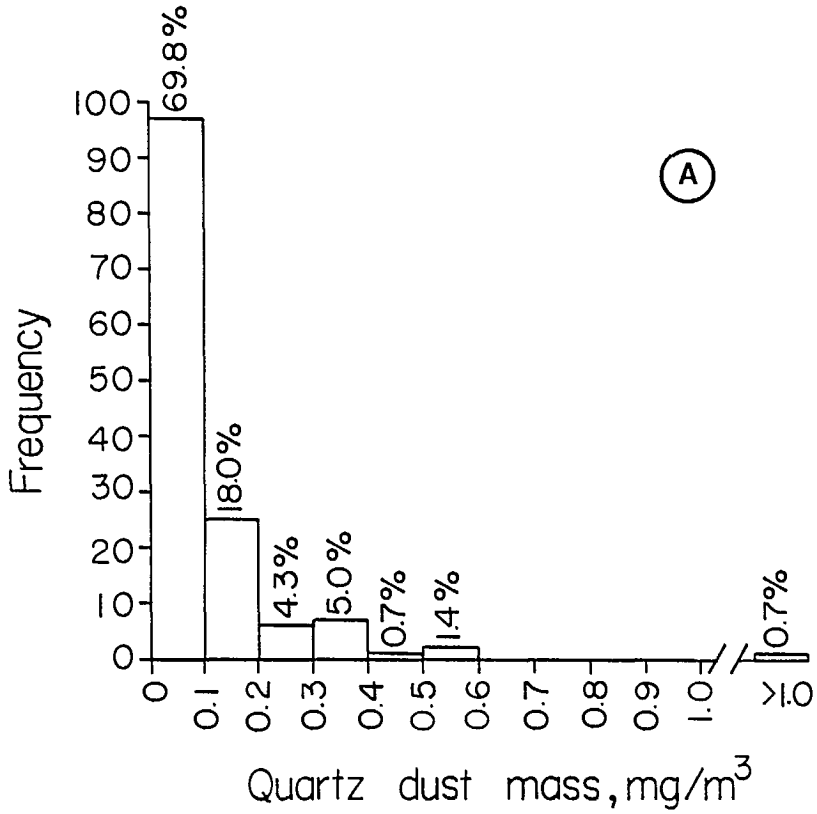
1. Knight, G., "Mine dust sampling system - CAMPEDS"; CANMET Report 78-7, CANMET, Energy, Mines and Resources Canada; 1978.
2. Bigu, J. and Grenier, M., "Studies of radioactive dust in Canadian uranium mines"; CIM Bull, vol. 77, pp. 62-68, 1981.

Table 1 - Data pertaining to lines of Figure 4 by linear regression analysis

Period	Slope (mBq/mg)	Intercept (mBq)	Remarks
1983	175.6	2.82	Fig. 4, upper curve
1984	162.0	1.41	Fig. 4, middle curve
1985	174.1	4.23	Fig. 4, lower curve
1983-85	166.88	2.22	Fig. 5
Ave.*	170.57	2.82	

*Average of 1983, 1984 and 1985.

LIST OF ILLUSTRATIONS


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 mass, mg/m3

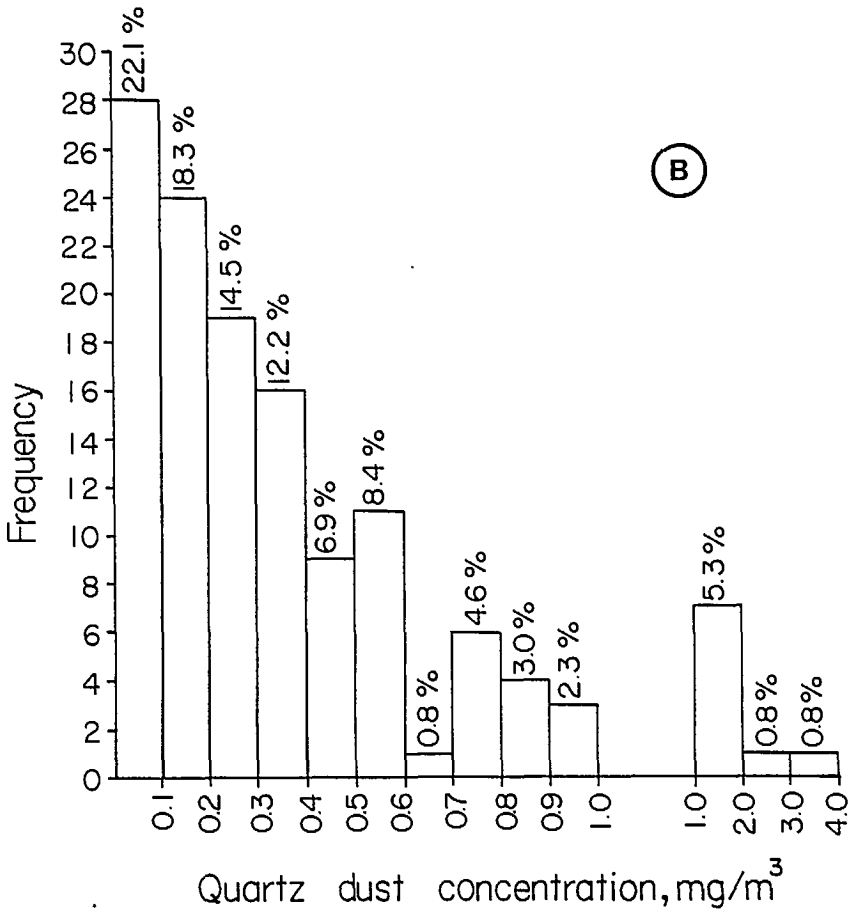
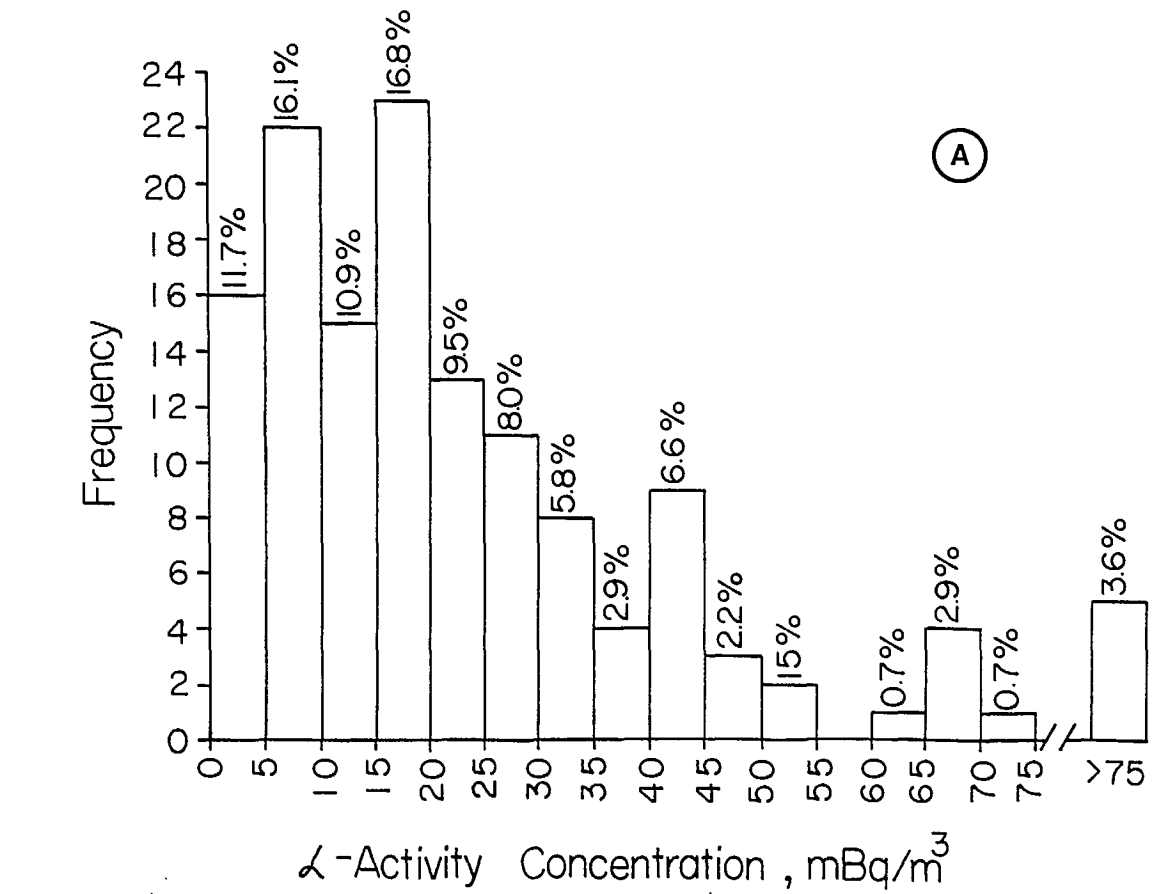

B
Frequency
30
28
26
24
22
20
18
16
14
12
10
8
6
4
2
0
22.1 %
18.3 %
14.5 %
12.2 %
6.9 %
8.4 %
0.8 %
4.6 %
3.0 %
2.3 %
5.3 %
0.8 %
0.8 %
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1.0
1.0
2.0
3.0
4.0
Quartz dust concentration, mg/m3

Figure 1


A
Frequency
11.7%
16.1%
10.9%
16.8%
9.5%
8.0%
5.8%
2.9%
6.6%
2.2%
1.5%
0.7%
2.9%
0.7%
3.6%
>75
α -Activity Concentration , mBq/m3

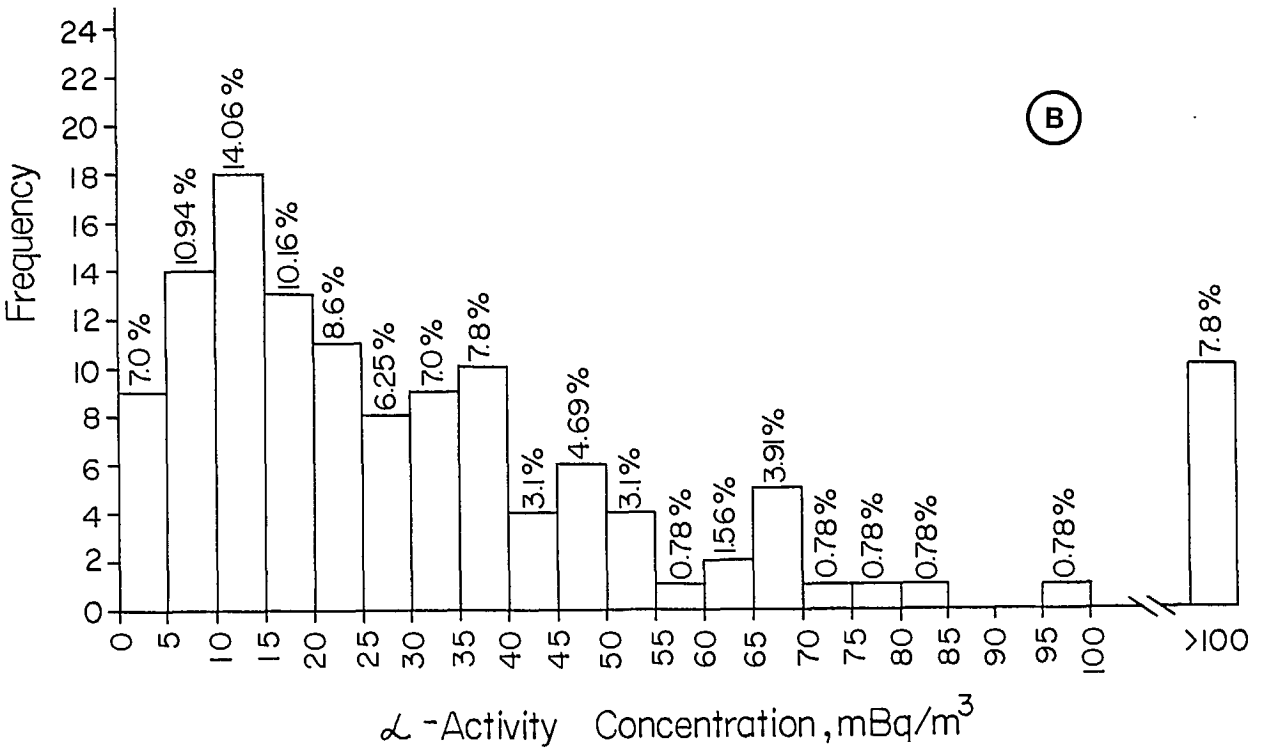

B
Frequency
7.0%
10.94%
14.06%
10.16%
8.6%
6.25%
7.0%
7.8%
3.1%
4.69%
3.1%
0.78%
1.56%
3.91%
0.78%
0.78%
0.78%
0.78%
7.8%
>100
α -Activity Concentration , mBq/m3

Figure 2

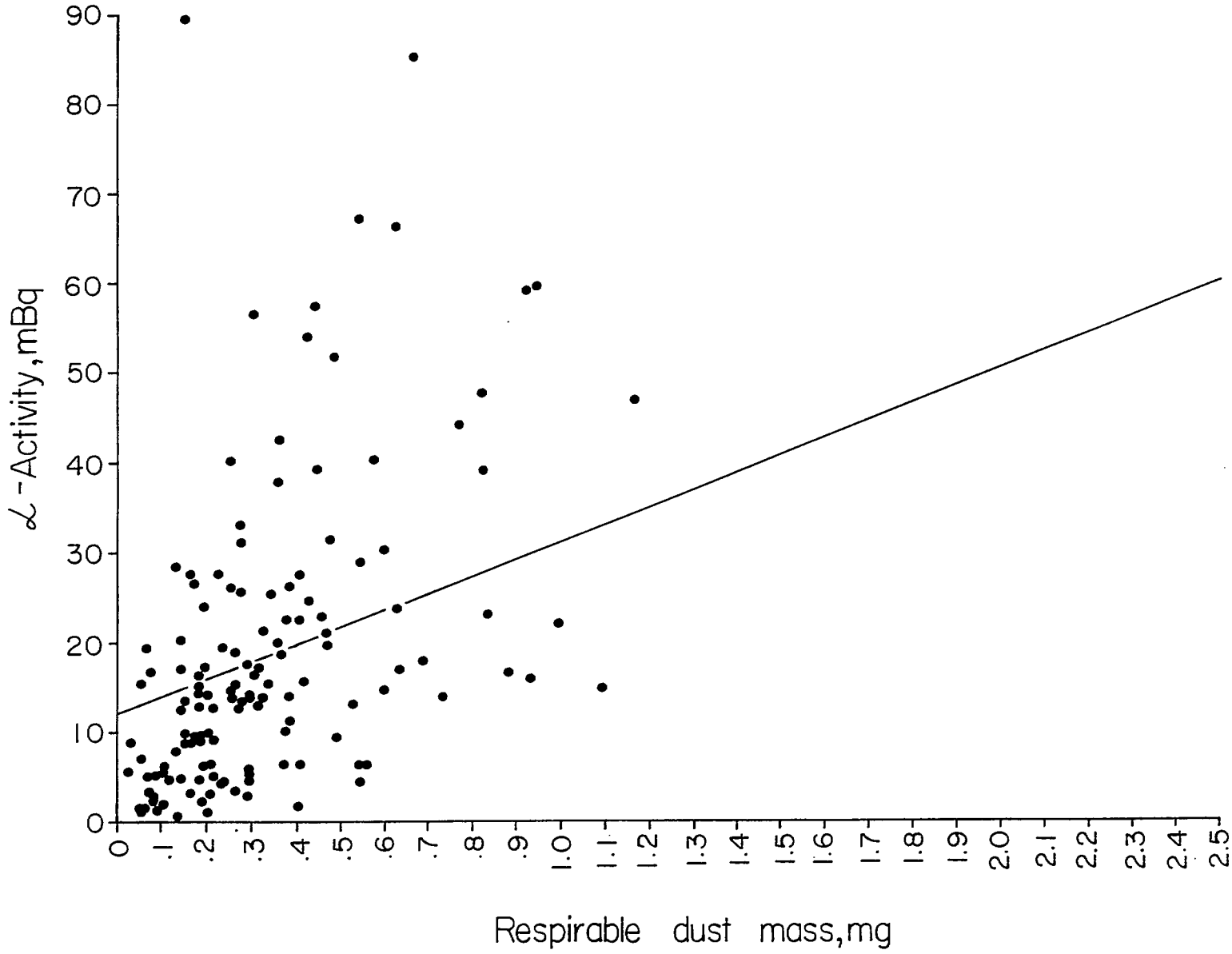

90
80
70
60
50
40
30
20
10
0
α - Activity, mBq
0
.1
.2
.3
.4
.5
.6
.7
.8
.9
1.0
1.1
1.2
1.3
1.4
1.5
1.6
1.7
1.8
1.9
2.0
2.1
2.2
2.3
2.4
2.5
Respirable dust mass, mg

Figure 3

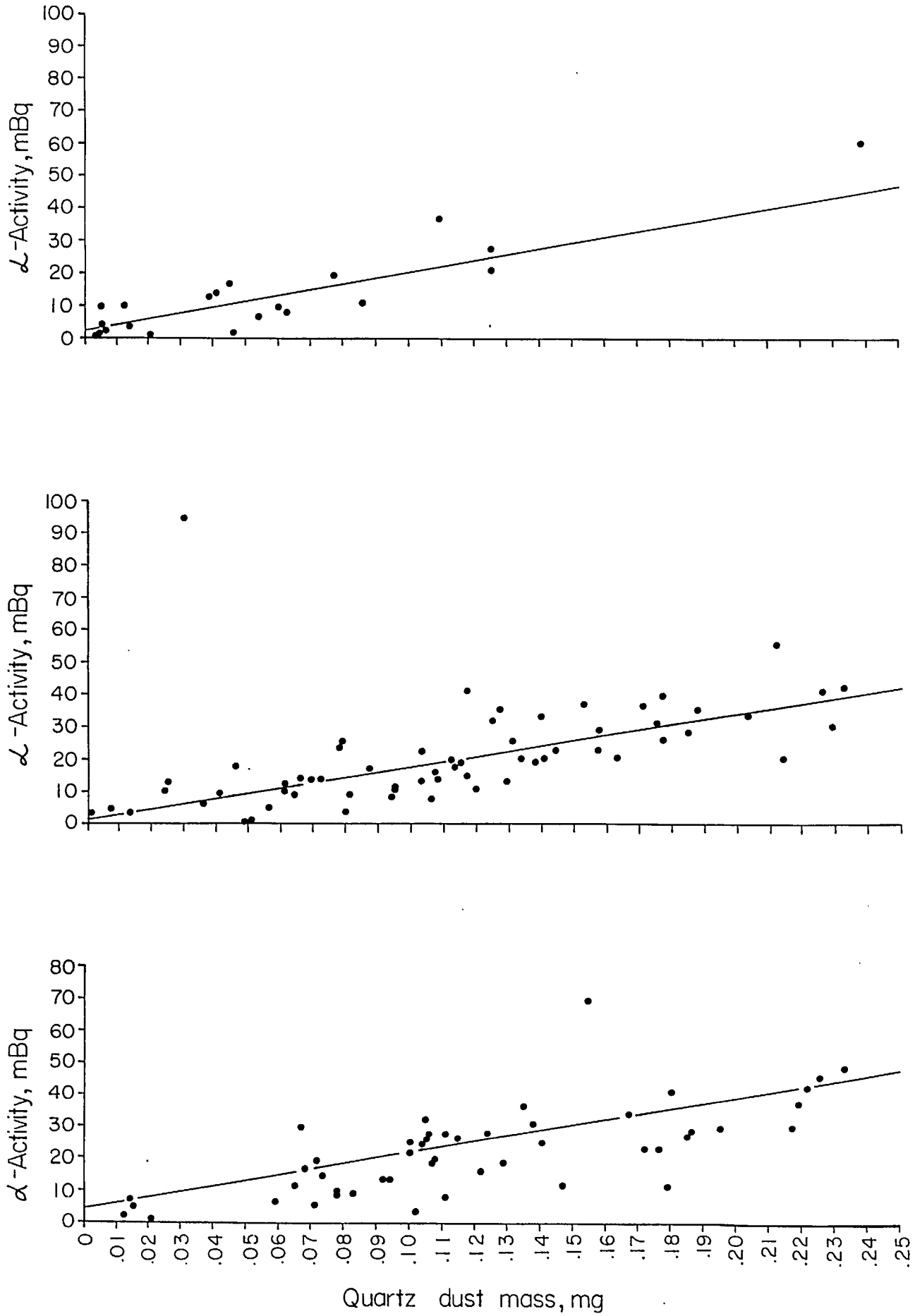

α-Activity, mBq
α-Activity, mBq
α-Activity, mBq
Quartz dust mass, mg

Figure 4

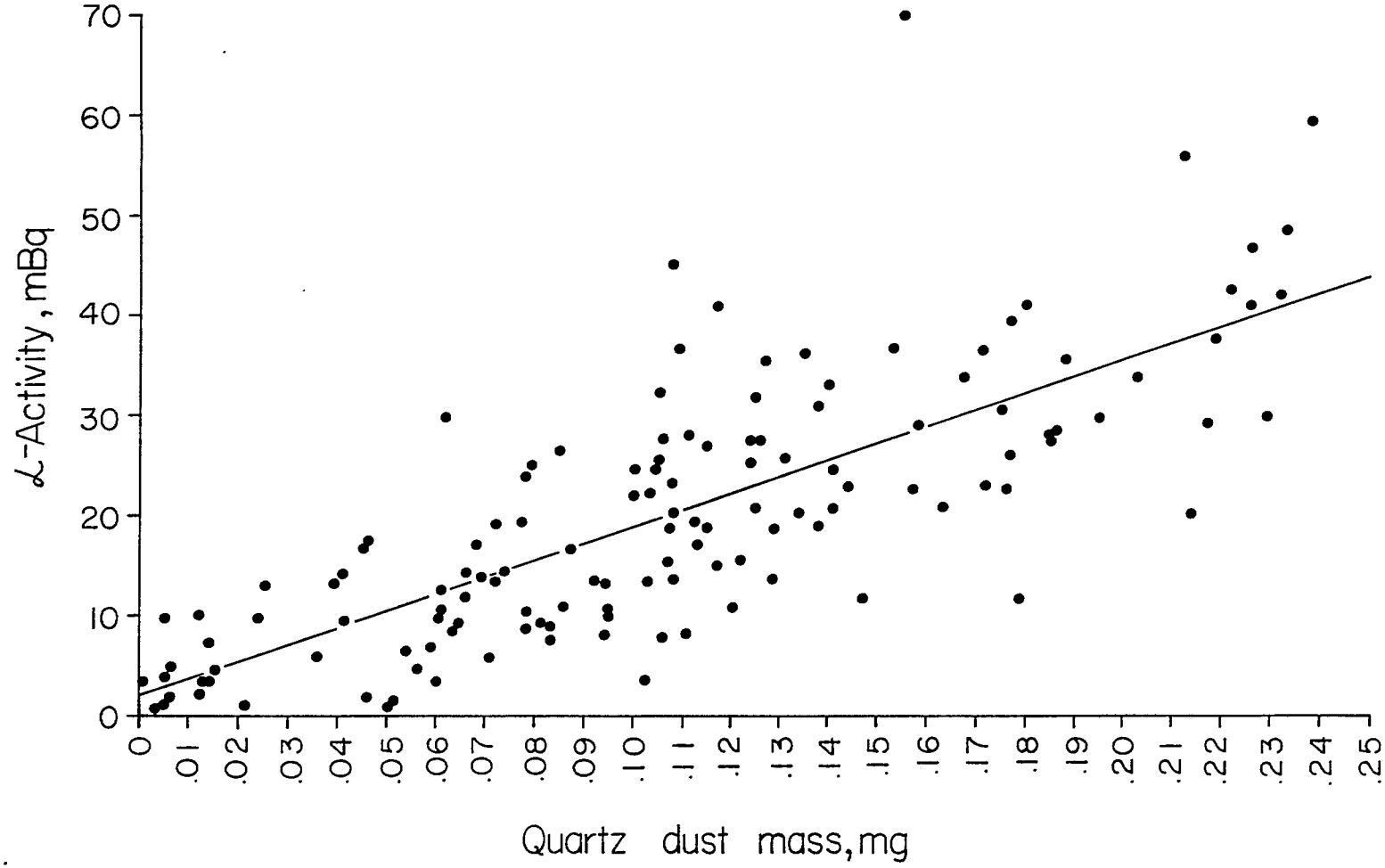

70
60
50
40
30
20
10
0
α-Activity, mBq
0
.01
.02
.03
.04
.05
.06
.07
.08
.09
.10
.11
.12
.13
.14
.15
.16
.17
.18
.19
.20
.21
.22
.23
.24
.25
Quartz dust mass, mg

Figure 5