Radon Daughters in Mine Atmospheres

—A FIELD METHOD FOR DETERMINING CONCENTRATIONS

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THE PHENOMENAL growth of the uranium mining industry in recent years has resulted in the exposure of large numbers of persons to high atmospheric concentrations of radon and its immediate degradation products. Uranium is also found associated with ores from many types of mines and may exist in sufficient quantity to make the hazard from radon and its daughters a real one. These circumstances necessitate the development of a method for the rapid determination of these radioactive daughters in the field in order to evaluate any hazards that might exist and to facilitate the control of such hazards. A rapid, on-the-spot, determination is also essential to check the efficacy of ventilation or other control methods by measuring "before" and "after" concentrations.

Two methods are currently used to measure radioactive contamination in mine air. The measurement of either radon alone or of the daughters alone may be made. It has been shown,[1,2] however, that given a mixture of radon and its daughters, the greater hazard by far is from the daughters. Therefore, use of the radon concentration as the only index of the hazard to an individual is poor practice. The daughter product concentration is much more significant and must itself be determined. It is conceivable that unusual equilibrium conditions may exist where the radon is virtually free of its daughters and would constitute the primary hazard. This is the rare case, however, and in almost all instances the daughters are the important hazardous contaminant.

The field determination of radon concentrations is at best cumbersome. The simplest equipment used by personnel of this station for radon collection and measurement includes a vacuum pump, scintillation cells, a photomultiplier tube and amplifier, a scaler, and a source of 110-volt 60-cycle A. C. power. Equipment for measuring daughter products, contrarily, comprises only a filter holder, a small hand-cranked pump, and a calibrated alpha survey meter, for example a Juno.*

Previous methods of measurement[3] utilize a technique of sampling for the daughters with a molecular membrane filter for a predetermined period of time, determining the activity on the filter and then relating this to atmospheric concentrations following, essentially, the method developed by Harley.[4] For any sampling time other than the predetermined period, an involved calculation has to be carried out by the field inspector. This technique assumes equilibrium conditions between the daughters. Where such conditions do exist, it is relatively simple to determine the concentration of the daughters in terms of micromicrocuries ($\mu\mu c$) per liter. Where non-equilibrium conditions exist, however, an error is introduced in calculating back to the original concentration in $\mu\mu c$/liter. The method presented here decreases the magnitude of that error.

One may consider the atmospheric concentration of radon daughters in terms of activity, expressed in micromicrocuries or disintegrations per minute (dpm) per unit volume. On the other hand, the level may also be considered in terms of the total alpha energy resulting from the ultimate decay of the daughter products in the body. There will be essentially no selective inhalation or selective retention of the radon daughters by the lungs as these elements will be carried on atmospheric dust, inhala-

tion, and retention being characteristic of the properties of the dust. Due to the short half-lives of the first four radon daughters, they will all decay to RaD in the lungs. Therefore, the total internal alpha-energy dose to the lung tissue bears a direct relationship to the total alpha energy which would result from the complete decay of a given concentration of radon daughters in air. It then becomes possible to express atmospheric concentrations in terms of the total alpha-energy dose without being immediately concerned with the specific amounts of the several isotopic species. This results in a considerable simplification of terminology and calculation.

If we accept as the tolerance level in the working atmosphere for an eight-hour day 100 $\mu\mu c$/liter each of RaA, RaB, and RaC, we can then determine the total alpha energy in Mev which would result from the complete decay of this concentration by the following method.

Each atom of the radon daughters, when it decays through RaC', will produce a given amount of alpha energy. It is this alpha energy, released internally, which causes damage to the lung tissues. RaA is itself an alpha emitter with an energy of 6.0 Mev per alpha particle. In decaying through RaC', the RaA then contributes another 7.7 Mev per atom (the alpha decay energy of RaC'). In a like, manner, RaB and RaC will each eventually contribute 7.7 Mev of alpha energy per atom. Because of the extremely short half life of RaC' (2.73 x 10⁻⁶ minutes), the internal hazard present from the atmospheric RaC' can be neglected. The total alpha energy from 100 $\mu\mu c$/liter of each of the first three radon daughters is then:

for RaA: $100 \ \mu\mu c \times \dfrac{2.22 \ \text{dpm}}{\text{liter}} \times \dfrac{1}{\mu\mu c} \times \dfrac{1}{.227^*} \times$

$\dfrac{13.7 \ \text{Mev}}{\text{atom}} = 13400 \ \text{Mev/liter}$

for RaB: $100 \ \mu\mu c \times \dfrac{2.22 \ \text{dpm}}{\text{liter}} \times \dfrac{1}{\mu\mu c} \times \dfrac{1}{.0259^*} \times$

$\dfrac{7.7 \ \text{Mev}}{\text{atom}} = 66000 \ \text{Mev/liter}$

for RaC: $100 \ \mu\mu c \times \dfrac{2.22 \ \text{dpm}}{\text{liter}} \times \dfrac{1}{\mu\mu c} \times \dfrac{1}{.0352^*} \times$

$\dfrac{7.7 \ \text{Mev}}{\text{atom}} = 48560 \ \text{Mev/liter}$

Total $= 127960 \ \text{Mev/liter}$

For practical calculations this can be considered as 1.3 x 10⁵ Mev/liter. Complete decay of 100 $\mu\mu c$/liter of RaC' would yield:

$100 \times \dfrac{2.22}{2.54 \times 10^5} \times 7.7 = 6.8 \times 10^{-3} \ \text{Mev/liter}.$

This is negligible compared to the values for RaA, RaB, and RaC.

Since the reading on the field instrument will be interpreted in terms of dpm, we must equate the activity on the filter at a given time after the end of sampling with the original atmospheric concentration. For convenience, throughout the remainder of this discussion, we shall assume that the count is made exactly 60 minutes after the end of sampling (T_{60}). We assume further that an "instantaneous" sample was taken. This eliminates the need for calculating, at present, the build-up and decay of the daughters on the paper during the sampling period.

It can be shown, by solving an equation of the Bateman type, that there will be 0.091 alpha disintegrations per minute expressed as RaC' at T_{60} from an initial amount of RaA by itself equivalent to 1 $\mu\mu c$ at T_0. From RaB, similarly, there will be 0.764 alpha disintegrations per minute at T_{60} from 1 $\mu\mu c$ at T_0. For 1 $\mu\mu c$ of RaC, the T_{60} figure is 0.271. From an initial amount of 100 $\mu\mu c$/liter of each of the daughters, there will be 112.6 dpm of alpha activity on the filter after 60 minutes. (This alpha activity, of course, is due to the decay of RaC'). Given an equilibrium condition at the tolerance level, a meter reading showing 113 dpm 60 minutes after the end of sampling indicates a potential total alpha energy of 1.3 x 10⁵ Mev/liter.

This relationship suggests using the alpha survey meter reading as an index describing the potential total alpha energy of the concentration of daughter products present in the atmosphere. A reading indicating five hundred seventy dpm, for example, would then show that a condition of five times the tolerance level, or a total alpha-energy release of 6.5 x 10⁵ Mev/liter existed. If we were to calculate the total alpha energy available from 500 $\mu\mu c$ of each of the daughters, we would find the result to be identical.

*This represents λ, or the number of disintegrations per unit time—one minute in this case.

pling was determined. The number of alpha dpm at various times after the end of sampling was calculated. When the factor was applied to these activity readings, a multiple of the tolerance level was determined. For both equilibrium and non-equilibrium concentrations, the maximum error was 12% on the safe side.

Calibration of Instruments.

THE ALPHA survey meter used must be calibrated in terms of dpm of RaC'. This may be accomplished by using a RaC' source and cross-checking the survey meter against a laboratory proportional counter. The RaC' source is prepared by the following procedure. Some high-grade uranium ore or a freely emanating radium solution is kept in a closed container to permit the build-up of radon and its daughter products. This radon daughter concentration is sampled through a filter such as Whatman 41 or Millipore.® This is then permitted to stand for 15-20 minutes to assure virtually complete decay of the RaA. The only alpha emitter remaining on the paper then is RaC'.

Field Operating Method

1. On a suitable filter, collect a sample of 100-250 liters within a five to ten minute period.

2. Take an alpha survey meter reading of the filter activity from 40 to 90 minutes after end of sampling; convert this to dpm.

3. Divide this reading by the number of liters sampled to give dpm/liter.

4. Divide the dpm/liter by the factor on the graph.

5. The resultant figure gives the multiple or fraction of the tolerance level (1.3 x 10^5 Mev/liter) existing in the air.

Summary

A NEW CONCEPT of describing the tolerance level of radon daughters has been presented. This concept is in terms of the alpha-energy dose resulting from the complete decay of the daughters. A rapid field method for determining atmospheric concentrations of radon daughters in terms of Mev of alpha energy/liter has been shown. No attempt has been made to set a maximum allowable concentration for radon and/ or its daughters. Should an MAC different from the tolerance level given in this paper be adopted, the factors in Fig. 1 would change by an appropriate constant. The method itself would remain the same.

Bibliography

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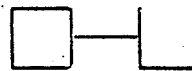6. TSIVOGLOU, E. C.: et al, Cited in reference 1.