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Grab Sampling and Continuous Monitoring Techniques for Radon Measurements at Controlled Environmental Conditions

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For this purpose, two devices RDA - 200 and TRI - MET 372 are used for active-type. Devices RGM I/I and CRM- 1027 are used for continuous monitoring. The experiments are carried out at two different environmental conditions, one by using high temperature (40°C) and relative humidity (100%), the other by using low temperature (16°C) and low relative humidity (50%).

Radon calibration chamber is a good tool in testing for radon measurements to different devices and techniques, a 65m^3 plexy glass chamber is constructed to compare different detecting devices with the change of environmental conditions (temperature – relative humidity – samples concentration) as well as the use of devices inside or outside the chamber. This chamber resemble to a great extend the mines atmosphere which help in using it as a pilot plant.

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GRAB SAMPLINGANDCONTINUOUSMONITORINGTECHNIQUESFORRADONMEASUREMENTSATCONTROLLEDENVIRONMENTALCONDITIONS

Strictly as per the compliance and regulations of :

Grab Sampling and Continuous Monitoring Techniques for Radon Measurements at Controlled Environmental Conditions

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The obtained results show that: Lower stability of measurements with the change in environmental conditions, more stability for the RDA - 200 and Tri - met devices at high temperature and high humidity. From the other hand a more stability for both RGM and CRM devices clearly appeared at low temperature and low humidity conditions. This may be attributed to that the measurements depend mainly on an electronic circuit which have a higher performance at these environmental conditions.

I. Introduction

There are two main categorized measuring techniques, active *technique and passive technique.*

The Grab Sampling Techniques (Active-type): In this case, air is sampled by drawing it through a filter material, housed in an adequate filter holder by means of air sampling pump. Radioactivity counting is carried out after the sampling period.

i) In case of Rn-gas measurements. The examined air sample is collected by filling a container, either previously evacuated or by a flow-through type in the sampling area and releasing the container. The radon from the sample is then later (may be days later) transferred to a measuring chamber, which may be ionization chamber or scintillation cell Lucas, (1957).

An integrated count method has been described (Jonassen, N. and Clements, W. E., (1974)., where the counting of radon activity may start at any time after filling the cell with daughter-free air. The accuracy of the method is shown to depend on the counting period rather than upon the time elapsed since filling. Another active method for measuring radon concentration using a metal cylinder and two filters (two filter method). In this case air is drawn through a cylinder fitted with a filter on either end. The front filter will remove all daughters in the air sampled, and the second filter will collect those daughters' products being produced by decaying of radon in the air, while it passes through the cylinder. The alpha activity from the filter at the end of sampling is then a measure of the radon concentration in the sampled air. (Fontan et al, (1962), Jacobi, (1964), Thomas and Le Clare, (1970)).

The measurement of radon daughters is often used not only for the determination of individual daughter concentrations but also for measurement (or estimation) of the potential alpha energy concentration of these daughters (WL). All methods used for both types of measurements are based on the analysis of the activity from a filter (or detector) on which the daughters have been deposited. The daughter's activity from the filter may be counted during and/or after sampling. Evans, (1969). If the air is drown through a filter, and the gross alpha activity is counted after a waiting a certain period for one time this called one count method [Kusntez, (1956)] and Rolle [Rolle, (1972)] methods, which differ only in the choice of sampling, waiting and counting times.

To overcome the low accuracy in the one count methods a lengthy operating procedure uses two uncorrelated counts. The most common two-count method is the Hill Method [Hill, (1975)]. The total alpha activity is integrated over two counting periods of two minutes after the end of sampling with the integrated counts I_0 and I_1 respectively. Using a special relation including this ratio and detector efficiency, the (PAEC) can be calculated. Methods for three uncorrected counts during and /or after sampling is called three-count methods, there are two techniques for the three count methods:

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a) Alpha gross count method, where the sum of RaA and RaĆ activities is measured. The most commonly used α-gross count method was developed by Tsivoglou [Tsivoglou, et al, (1953)] in which the activity from a filter is measured at three intervals after sampling. The amount of air sampled is about 50 liters. Because count rates are used instead of integrated counts, even at high concentrations (tens of thousands Bq/m^3), the reproducibility may only be good within 20-30 percent. By integrating the alpha activity over time intervals instead of measuring the rate at three times, it is possible to reduce the uncertainty of the Tsivoglou method considerably as proposed by Thomas [Thomas, (1972)].

The uncertainty is usually high for RaA, especially if the atmosphere is near radioactive equilibrium. From three different gross-alpha count rates it is possible to calculate the concentrations of RaA (Po-218), RaB (Pb-214) and RaC (Bi-214), by solving a system of three Bateman linear differential equations with three unknowns [Evans, (1969)].

b) Alpha spectroscopy methods, where RaA and RaC activities are measured individually. In these methods, the alpha particles from radon daughters can be counted separately and the daughter concentration can be obtained by distinguishing between the 6-MeV and 7.69 MeV alpha particles energy groups. A method employing only alpha radiation has been developed by Martz et al [Martz et al, (1969)].

In case of Continuous Monitoring Techniques: air samples are taken on a continuous basis lasting typically from several days to several weeks. Counting of radioactivity is conducted by the same instrument and concurrently with air sampling. Hence, the radioactivity count, or activity is being "integrated" by the instrument. Also, the continuous monitoring of radon concentrations is the most useful method for studying the effects of ventilation on radon concentration levels and determination of average radon concentration.

In continuous monitoring of radon daughters, the air is sampled continuously and the alpha and/or beta activities are integrated over intervals ranging from minutes to hours. [Dalu, G. A. and Dalu, G., (1971), Thomas, (1977), McVey et al, (1977) and Wrenn et al, (1975)].

In radon daughter measurement systems, the daughters are collected on a filter, which is placed in front of a detector. A common calibration procedure uses an essentially mono-energetic alpha emitter, americium-241, uniformly deposited on a metal disc as pseudo radon daughter calibration source. An absolute method for calibration of radon daughter system has been developed by Falk [Falk, (1982)]. Beckman [Beckman, (1975)] described calibration of air sampling equipment and counting equipment including field checks, calibration equipment and data treatment. For radon daughter calibration, Thomas (Modified Tsivoglou) and alpha spectrometric methods have been suggested as the most suitable primary counting methods [Lawrence Berkeley Laboratory, (1981)].

II. Experimental Techniques

a) Calibration chamber and samples

The chamber is made of (0.5 cm thick) plexy glass sheets. It is of cubic shape with volume of 0.65 m^3. On top, a removable ceiling contains four wholes of (1 cm in diameter) each. Two wholes are used for the intake of samples from the chamber, the other two are used for the electricity cables needed for the devices inside the chamber. It has also the possibility for use active techniques for radon and its daughter's detection (Tawfik, M. N 2011).

Figure 1 : Radon calibration chamber

Five different samples were selected from Gattar Mountain (Hurghada, Red Sea, Egypt). The uranium content of these samples was experimentally measured in the NMA. Laboratories (Salem, 2008). The chosen samples are nearly equal weight, size, and physical state (solid rocks) with no fractures or any crushing to smaller grains. The samples are labelled from one to five according to their uranium ppm measurements as follows: Sample 1 (35 ppm), sample 2 (200 ppm), sample 3 (330 ppm), sample 4 (470 ppm), sample 5 (850 ppm).

b) Devices and methods

A total of four devices are used, two of them (RDA - 200 and TRI - MET 372 for the grab sampling (active) techniques

i. *RDA – 200:* It is designed to measure alpha activity originated from radon and its daughters. The alpha particles register on the ZnS (Ag) phosphor coating of the scintillator cell or tray in the form of light flashes. Each flash of light, as seen by the high gain photomultiplier tube, is transformed into an electrical impulse. These impulses are accumulated, counted and then digitally displayed on a five digit, seven segments L.E.D. digital display, after a preset counting time has been completed. The device weight is about 3 kg. (EDA Instruments INC).

ii. *TRI-MET 372:* It is designed as an alpha counter, equipped with a foil type open Zinc Sulphide detector. The instrument is manufactured by (Tri-Met Instruments LTD). This type of detector is inexpensive, readily obtainable and is easily replaced. The sample holders can accommodate membrane filter samples of 25 mm, 37 mm. and 47 mm diameter. The device weight is about 2.5 kg, the Tri-Met was used for measuring (WL) by Roll – method. (Tri-Met instruments LTD).

The second category is for continuous radon gas monitoring

i. Portable Radioactive Gas Monitor type RGM l/l is a self-contained instrument for measuring the concentration of radioactive gases in air, it is calibrated for Tritiated Water Vapour (HTO) in air, for direct radon gas determination; a single portable, battery operated device is used to monitor radon gas. The device is of weight (14.5Kg). (Nuclear Enterprises Ltd) .

ii. Professional continuous radon monitor model 1027 is a patented electronic detecting device using a diffused-Junction photodiode sensor to measure the concentration of radon gas. The unit is operated from standard line power and includes a 9-volt battery which provides backup power. For continuous radon monitoring; a single fixed, electricity operated device is used for monitoring of radon gas. It has an internal detector for measuring radon gas and it is equipped with a digital screen for displaying of results. It has a weight of (1 Kg). (Sun Nuclear Corporation).

c) Measuring procedures

The experiment is done according to the variable factors, the plan can be divided into two parts: one using high temperature (40°C) and relative humidity (100%) conditions and the other by using low temperature (16°C) and low relative humidity (50%).

Each sample is put separately inside the radon chamber and the measurements started from zero time till 168 hours (one week), the measurements are taken for all devices at the same time every four hours resulting in 42 measurements for each device. The temperature is adjusted to reach 40 °C and a by using a water bath as water source to ensure a relative humidity level of 100 % (Tawfik, M. N 2011).

The procedures are done as follows:

i. For the total potential alpha energy concentration expressed as working level PAEC. Rolle method is used, using a portable air sampler to withdraw up to 5 l/min of air through a filter paper.

Radon daughters are collected on a high-efficiency filter paper for 10 minutes and then alpha-counted with either a rate meter or scalar after a prescribed delay of about 5 to 10 minutes. (Rolle, 1968).

The working level is calculated by the following formula (Safety Series No. 43, (1979)]:

$$WL = \frac{R - B}{EvtF} \quad (1)$$

Where:
R = Count rate at T minutes from the end of sampling (in counts/min)
B = Count rate due to background measurement
E = Counting efficiency, decimal fraction
v = Volumetric sampling rate in L/min
t = Sampling time in minutes
F = Conversion factor =212 for sampling periods of from 1 to 20 min

ii. For direct radon gas measurement using RGM l/l: The technique starts by withdrawing a sample of air by the air pump followed by a direct measurement by the ionization chamber detector to obtain the result of the measurements in a few seconds from sampling. (Grossi, 2001).

iii. For continuous radon monitoring: A sensitive semiconductor detector is exposed to radon to start the measuring processes, by this way the radon can be continuously monitored. These values are stored using a small internal memory for future revision.

III. The Annual Dose Conversion Factors were Done as Follows

1) For the data information from (RDA - 200 and TRI-MET 372) instruments .The total working hours per one year will be two thousand (2000) hours per year. The ICRP-31 (1980) (International Commission for Radiation Protection) submits a conversion factor (CF)) for the calculation of annual dose from the WLM to be five times greater, so according to the previous information the final formula can be written as:

$$\text{Annual dose (mSv/y)} = \frac{(WL)X(2000h)X(5CF)}{(170h)} \quad (2)$$

2) The calculation of annual dose using the data information from the RGM 1/1 (Radioactive Gas Monitor) which gives the data in μCi/m^3.

$$\text{Annual dose (mSv/y)} = \frac{(\mu Ci/m^3)X(1000UC)X(0.2331CF)}{(1000DUC)} \quad (3)$$

3) The calculation of annual dose using the data information from the CRM -1027 (Continuous Radon Monitoring) which gives the data in pCi/l.

$$\text{Annual dose (mSv/y)} = (pCi/l)X(0.2331CF) \quad (4)$$

Where :

UC: is the unit conversion from μCi/m^3 to pCi/l.

DUC: is the device conversion factor from Tritiated Water Vapour (HTO) in air to μCi/m^3.

IV. Results and Discussion

a) Active techniques (grab sampling) one count method

i. *At High Temperature and Humidity*

For the Tri-Met 372, figure (1) shows the experimental results of the calculated annual doses using equation (2) with the five samples, a low annual doses (mSv/y) are reported for low sample concentrations and minimum time period (4-20 h) while high values for annual doses (mSv/y) are reported for maximum sample concentrations and maximum time periods (140-164 h)

At low samples concentrations a smoothly increasing in the calculated values are observed which may indicate that the instrument was able to discriminate between small concentrations at the harsh environmental conditions. At a high samples concentrations a high ability for the discrimination between high and low concentrations epically at the harsh environmental conditions was observed

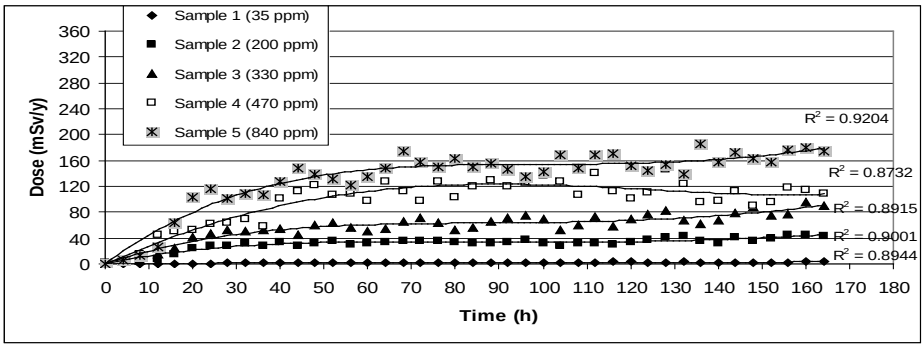

Figure 1 : Calculated annual dose (mSv/y) using the measurements of Tri-met device for all samples at high relative humidity and temperature versus time (h).

For RDA 200, figure (2) shows experimental results of the calculated annual doses using equation (2) with the five samples the time interval.

It is obvious, low annual doses (mSv/y) were reported for low sample concentrations and minimum time period (4 – 20 h) While high values for annual doses (mSv/y) was reported for maximum sample concentrations maximum time periods (140 – 163 h).

At low samples concentrations a smooth variation are observed which may indicate that the instrument is able to discriminate between small concentrations at the harsh environmental conditions. Also it is shown that at high samples concentrations a complete separation between the measured values, which means that the instrument has the ability to discriminate between high and low concentrations at harsh environmental conditions that may gives it advantages for using in such cases.

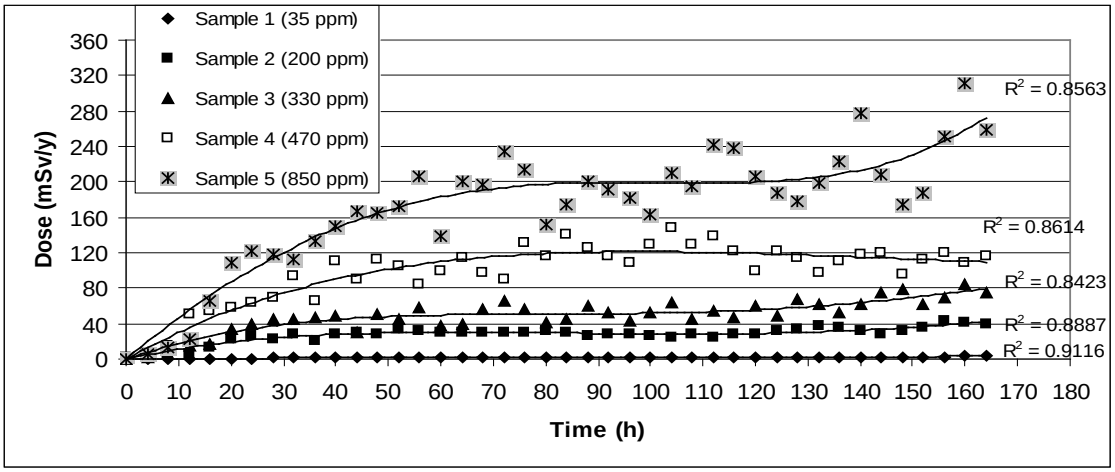

Figure 2 : Calculated annual dose (mSv/y) using the measurements of RDA 200 device for all samples at high relative humidity and temperature versus time (h).

ii. *At Low Temperature and Humidity*

For the Tri-Met 372, figure (3) shows that low annual doses (mSv/y) are reported for low sample concentrations and minimum time period (4 – 20 h), while high values for annual doses (mSv/y) are reported for maximum sample concentrations maximum period (140 – 164 h).

From the table also we can get that, an increase in the calculated annual doses with time when using different samples from sample (1) to sample (5). A nearly closed data appears for sample (2) and sample (3) also a slight overlap appears for sample (4) and sample (5).

There is some coincidence in some measured values in case of high and low samples concentrations where there is a complete difference between the two groups, which means that the instrument does not have the ability to discriminate between high and low samples concentrations at normal environmental conditions.

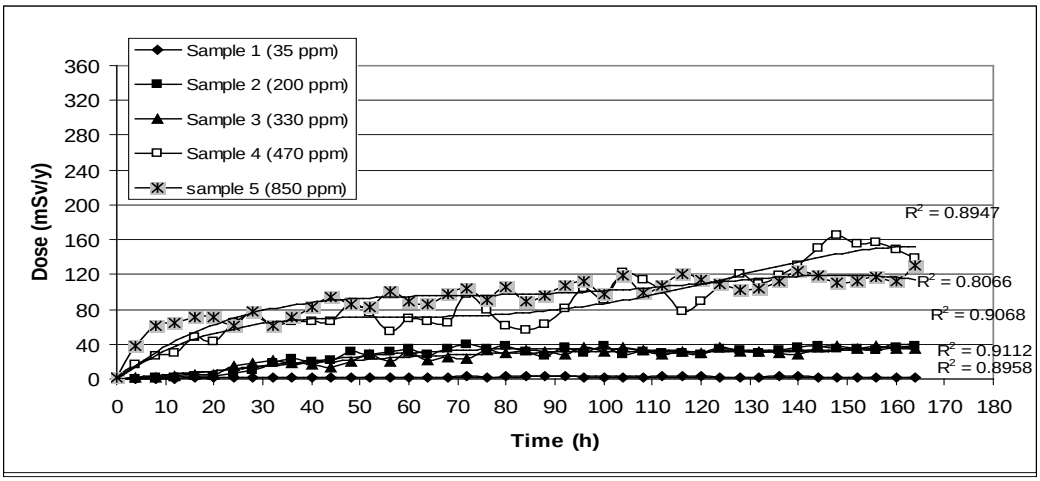

Figure 3 : Calculated annual dose (mSv/y) using the measurements of Trimet device for all samples at low relative humidity and temperature versus time (h).

For RDA 200, figure (4) shows that low annual doses (mSv/y) are reported for low sample concentrations and minimum time period (4 – 20 h). While high values for annual doses (mSv/y) was reported for maximum sample concentrations maximum period (140 – 164 h)

An increase in the calculated annual doses with time when using different samples, from sample (1) to sample (5). A slight overlap appears for sample (4) and sample (5).

At low samples concentrations a normally increases in the calculated annual dose with time.

There is an overlap in some measured values in case of high and low samples concentrations where there is a complete separation between the two groups, which means that the instrument does not have the ability to discriminate between high and low samples concentrations at normal environmental conditions.

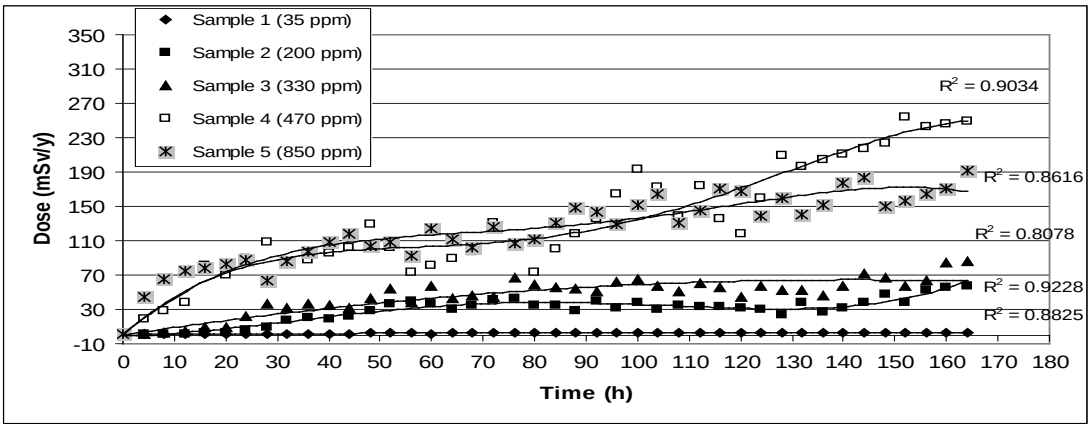

Figure 4 : Calculated annual dose (mSv/y) using the measurements of RDA 200 device for all samples at low relative humidity and temperature versus time (h).

b) Continuous radon monitoring

i. *At High Temperature and Humidity*

For the RGM 1/1, figure (5) shows experimental results of the calculated annual doses using equation (3) with the five samples

It is obvious that there is a gradually increase in the annual doses measured in (mSv/y) goes from low sample concentrations and minimum time period (4 – 20 h) to maximum sample concentrations maximum period (140 – 164 h).

At low samples concentrations a highly nearly closed values are observed while in the measured annual dose values for the most high two samples concentrations, there is a completely difference between them especially at high one (850ppm), which indicate a low performance for this instrument as it could not discriminate between high and low sample concentrations at harsh environmental conditions that may gives it disadvantages for using in such cases.

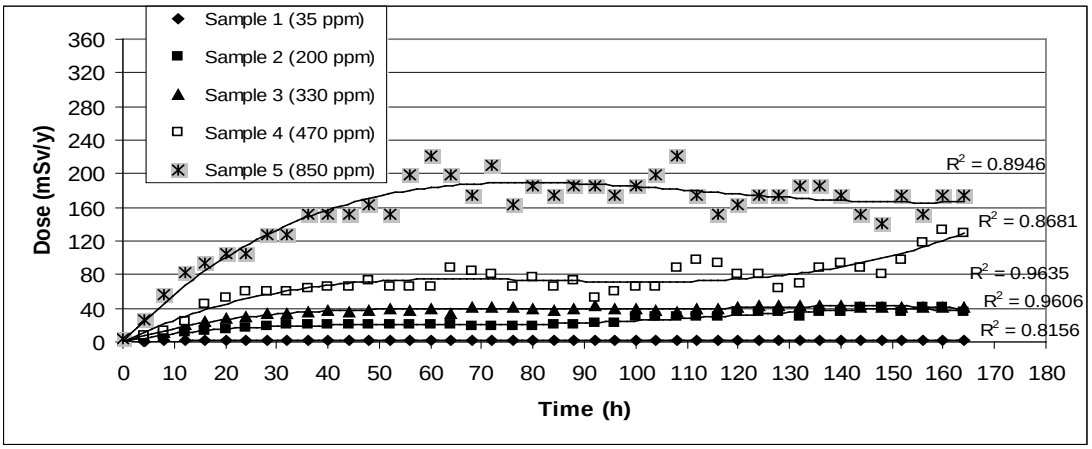

Figure 5 : Calculated annual dose (mSv/y) using the measurements of RGM device for all samples at high relative humidity and temperature versus time (h).

For the CRM- 1027, figure (6) shows experimental results of the calculated annual doses using equation (4) with the five samples

It is obvious that there is a gradually increase in the annual doses measured in (mSv/y) goes from low sample concentrations and minimum time period (4 – 20 h) to maximum sample concentrations maximum period (140 – 164 h).

From the data we can get that: At low samples concentrations a highly closed values with small resolution and discrimination at these low concentration values. While for the high samples concentrations there is a close separation with small over lapping at small time intervals, which indicate a slightly confusion in the performance of the instrument at high and low concentrations at harsh environmental conditions.

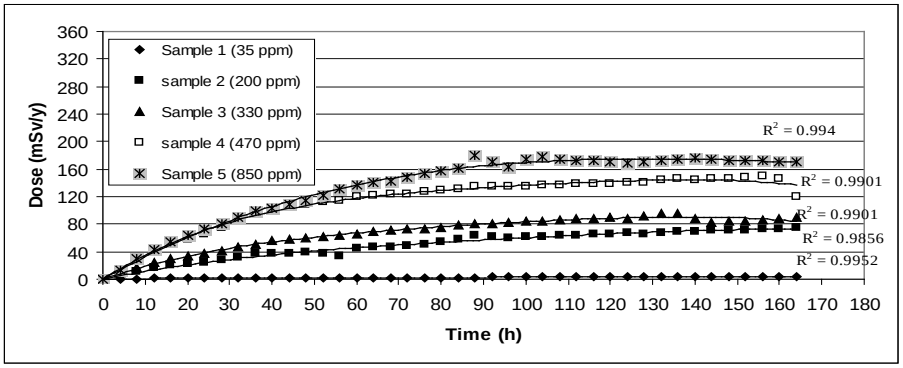

Figure 6 : Calculated annual dose (mSv/y) using the measurements of CRM device for all samples at high relative humidity and temperature versus time (h).

ii. *At Low Temperature and Humidity*

For the RGM 1/1, figure (7) shows experimental results of the calculated annual doses using equation (3) with the five samples. A gradually increase in the annual doses measured in (mSv/y) goes from low sample concentrations and minimum time period (4 – 20 h) to maximum sample concentrations maximum period (140 – 164 h)

At low samples concentrations, highly smoothed closed values are observed with minimum values.

A complete separation between the measured values for the annual dose of low and high samples concentrations and at low and high values for temperature and humidity, which indicate a high performance for this instrument at high and low samples concentrations and at normal environmental conditions that may gives it advantages for using in such cases.

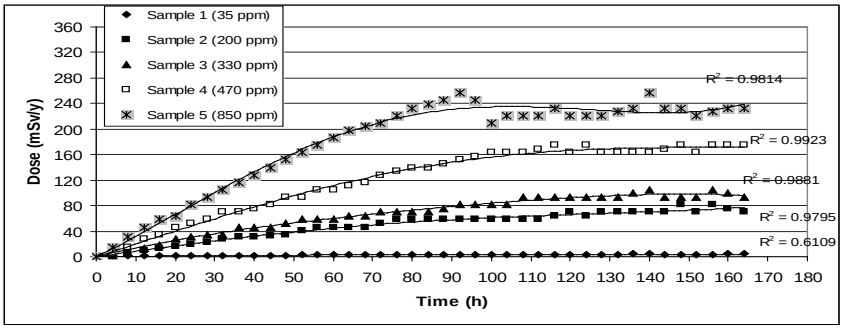

Figure 7 : Calculated annual dose (mSv/y) using the measurements of RGM device for all samples at low relative humidity and temperature versus time (h).

For the CRM- 1027, figure (8): At low and high samples concentrations, highly smoothed closed values with high resolution and discrimination at these concentrations values. This indicates high performance for this device at normal environmental conditions, giving it advantage for use in such cases. However at very high sample concentration it goes out of device range after 80 hours.

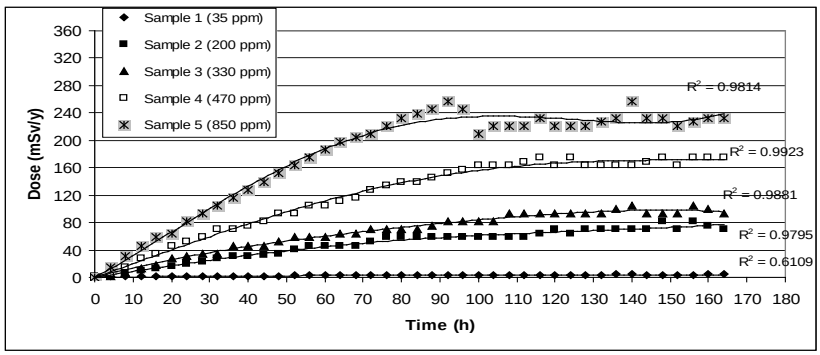

Figure 8 : Calculated annual dose (mSv/y) using the measurements of CRM device for all samples at low relative humidity and temperature versus time (h).

V. Conclusions

The comparison of used devices showed a higher time is required to complete the surveillance process (from the beginning till the end of measurements) is 13 min. for both Tri-met and RDA 200 devices while it needs from 1 to 3 min. for RGM device, the CRM device takes about 1 to 2 sec. for the measurement but it needs to run at least 1 h for before each measurement. That is based on the principals of the measuring devices and the theory of operation of used instruments.

Higher cost estimation to complete the surveillance process (from the beginning till the end of measurements) is 4.75 L.E. for the use of RDA - 200 device for both the cost of filter paper and required batteries, 4 L.E. for the use of Tri-Met device for both the cost of filter paper and required batteries, 1 L.E. for the use of CRM device for the cost of the required batteries while no cost is required for the use of RGM device.

Lower stability of measurements with the change in environmental conditions showed a more stability for the RDA - 200 and Tri - met devices at high temperature and high humidity due to the limited effect of high temperature and relative humidity on the sampling and counting instruments. From the other hand a more stability for both RGM and CRM devices clearly appeared at low temperature and low humidity conditions this may be attributed to that they depend mainly on an electronic circuits which have a higher performance at these environmental conditions.

No human mistakes are found when using CRM device but a small human mistakes can be found when using RGM device in the misreading of its analogue scale, while there are many expected human mistakes when using RDA - 200 and Tri - met device due to the use of stop watch for sampling time, the handling of the filter paper when removing from the sampling pump and applying to the RDA - 200 or Tri - Met devices.

Small maintenance operations can be done easily and in short time for both RDA - 200 and Tri - Met devices while it is difficult to do maintenance for both RGM and CRM device which requires a specialist to do them.

Large number of steps needed for the use of RDA - 200 and Tri - Met devices which are 7 steps. While RGM device needs 3 steps and CRM device requires only 1 step to complete the whole process from the beginning till the end of measurements.

Higher weight of RGM device is the larger one of 14.5 Kg followed by the use of RDA 200 device and its accessories of 3 Kg. the weight of Tri - met device and its accessories is 2.5 Kg, while a weight of only 1 Kg is for CRM device.

Table (1) shows the comparison between different used devices according to the comparison matrix (time, cost, efficiency and ease of use).

Table 1 : the inter-comparison between the used instruments

		Grab Sampling Technique		Continuous monitoring	
		Tri-met	RDA 200	RGM	CRM
Time		Medium	Medium	Low	High
Cost		High	High*	Low	Medium
Efficiency		Medium*	Medium*	Medium	Medium
Ease of use	No. of steps	High	High	Medium	Low
	Weight	Medium	Medium*	High	Low
	Training ease	High	High	Medium	Low

**is noted for the slight increase.*

This study recommends the application of RDA 200 and Tri- Met devices at mines and mills according to their high performance and limited requirements to work at these places.

The application of CRM and RGM devices at laboratories according to their high performance and limited requirements to work at these places.

It is very important to construct a complete comparison when choosing the set of devices and techniques for the use during the surveillance process.

Good preparation for surveillance and monitoring radiation programs including the best use of techniques and devices will achieve the goal of radiation protection which will help in decreasing the life-shortening percentage from(10%-15%) to less than that.

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