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PAPER 73-28

AN IONIZATION CHAMBER FOR CONTINUOUS MONITORING OF ATMOSPHERIC RADON: RADON-222 LEVELS IN OTTAWA AND GATINEAU HILLS, CANADA

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ABSTRACT

A 250-litre ionization chamber was constructed and tested and was found to be suitable for continuous monitoring of atmospheric radon. The ionization chamber consists essentially of a shielded 45 cm I.D. x 150 cm stainless steel cylinder, a 2 cm centre electrode and a filter and blower at the inlet. The negative ion current generated by ionizing radiation is collected from the centre electrode by a vibrating capacitor-type electrometer. To cancel the relatively large but constant background due to gamma-rays and junction potentials, only the fraction of the signal above this background level is amplified, smoothed and recorded on a chart recorder.

When stationary the chamber has a radon-222 sensitivity of 0.0045 pc/1/mpA and a detection limit of about 0.04 pc/1 when the signal is integrated hourly.

Continuous monitoring of atmospheric air from an elevation of 85 feet above ground level at the Geological Survey of Canada Building, Ottawa, revealed a diurnal cycle in the radon-222 content on quiet days with a maximum in the early morning hours. In general, higher radon levels corresponded with hazy quiet atmospheres and lower levels with clear and windy or rainy days.

Several tests in the Gatineau Hills, Quebec, showed that air from a 30-foot height contained about the same amount of radon at Fortune Lake where uranium mineralization is known to occur as at Pinks Lake where no uranium mineralization is evident.

By placing the chamber in a boat on a lake and passing compressed air, which had been used to agitate the water, through the chamber a reading much stronger than that from atmospheric air was obtained. Suitably modified, the ionization chamber could become useful in the search for uranium mineralization covered by water.

RÉSUMÉ

On a construit et essayé une chambre d'ionisation de 250 litres et on l'a trouvée convenable pour le contrôle permanent du radon dans l'atmosphère. La chambre d'ionisation est constituée essentiellement d'un cylindre blindé en acier inoxydable de 150 cm de longueur et de 45 cm de diamètre intérieur, d'une électrode de 2 cm au centre, d'un filtre et d'une soufflerie à l'orifice d'admission. Le courant d'ions négatifs produit par le rayonnement ionisant est tiré de l'électrode centrale à l'aide d'un électromètre de type à condensateur. Pour annuler l'arrière-plan relativement étendu mais constant dû aux rayons gamma et aux tensions de jonction, seule la fraction du signal qui s'élève au-dessus de ce niveau de fond est amplifiée, adoucie et enregistrée sur un enregistreur de courbe.

Lorsqu'elle est immobile, la chambre a une sensibilité au radon-222 de 0.0045 pCi/1/mpA et une limite de détection d'environ 0.04 pCi/1 lorsque le signal est complété à chaque heure.

Un contrôle permanent de l'air de l'atmosphère à une altitude de 85 pieds au-dessus du niveau du sol à l'édifice de la Commission géologique du Canada à Ottawa a révélé un cycle diurne dans la teneur en radon-222 pendant les jours calmes, le maximum se situant pendant les premières heures du matin. En général, les niveaux plus élévés de radon correspondent aux atmosphères brumeuses et calmes alors que les niveaux les plus bas correspondent aux journées claires et venteuses, ou pluvieuses.

Plusieurs essais dans les collines de la Gatineau (Québec) ont révélé que l'air à 30 pieds du sol contient environ la même quantité de radon au lac Fortune, où on sait qu'il y a une minéralisation d'uranium, qu'au lac Pink où il n'existe pas de minéralisation connue d'uranium.

En plaçant la chambre dans une embarcation sur un lac et en faisant passer l'air comprimé, préalablement utilisé pour agiter l'eau, à travers la chambre, on a obtenu une valeur beaucoup plus forte que celles obtenues à partir de l'air de l'atmosphère. Une fois modifiée convenablement, la chambre d'ionisation pourrait devenir utile pour la recherche des minéralisations d'uranium situées sous l'eau.

AN IONIZATION CHAMBER FOR CONTINUOUS MONITORING OF ATMOSPHERIC RADON: RADON-222 LEVELS IN OTTAWA AND GATINEAU HILLS, CANADA

INTRODUCTION

The successful application of radon-222 in surface waters and soils (Dyck, 1972) prompted the construction of a large ionization chamber in the hope that by continuously monitoring the radon content of the air in a region it would be possible to outline uranium-rich areas. A second possible application of such a device would be the more detailed investigation of lake and stream bottoms and shorelines by putting the chamber in a boat or on the float of a helicopter, moving along the surface of the water while agitating the water and passing the vapours through the chamber. Also, since the advent of high sensitivity airborne gamma-ray spectrometry it has become apparent that atmospheric radon can account for over half of the signal strength recorded by the detector in the uranium channel (Darnley, 1972). It would therefore be most helpful if this radon component could be corrected for continuously and in flight.

Because of the widespread occurrence of uranium and thorium on the earth's surface, the gaseous daughter products radon-222 and radon-220, also known as thoron, have been studied for a long time, particularly in connection with atmospheric turbulence and mass transfer studies near the ground (Israel and Horbert, 1970; Guedalia et al., 1970; Horbert, 1969; Pearson and Moses, 1966; Israel, 1965) and in the lower atmosphere (Lambert et al., 1970; Wilkening, 1970; Bradley and Pearson, 1970; Jonassen and Wilkening, 1970; Rama, 1970). Most workers use the batch-type method of sampling and measuring the radon content of the air. This involves the collection of the decay products of radon-222, which have attached themselves to aerosol particles, on filters and measuring the characteristic gamma-ray peaks of bismuth-214 and/or lead-214. Several such methods are described in the collection of papers in The Natural Radiation Environment (Adams and Lowder, 1964).

This paper describes results obtained with an ionization chamber similar to that described by Horbert (1965) and Israel (1965).

Description and Operation of the Ionization Chamber

A cross-sectional drawing of the ionization chamber is shown in Figure 1. For handling and transport the whole assembly is fixed inside a rectangular frame made of Dexion angle aluminum. To make the unit as light as possible the cylinder constituting the chamber proper and the outer ground shell were made from 0.8-mm-thick stainless steel. While this reduced the weight of the chamber it unfortunately resulted in somewhat flexible shells which caused spurious signals when readings were taken in a moving vehicle. The air filter is of the Absolute type guaranteed to remove 99.97 per cent of particles with diameter equal or greater than 0.3 micron. This insured that a negligible proportion of radon decay products attached to aerosols entered

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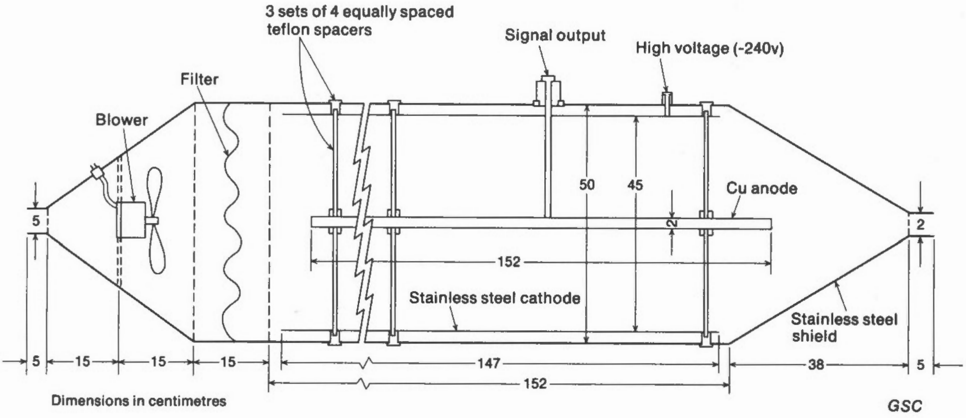

Figure 1. Ionization chamber cross-section.

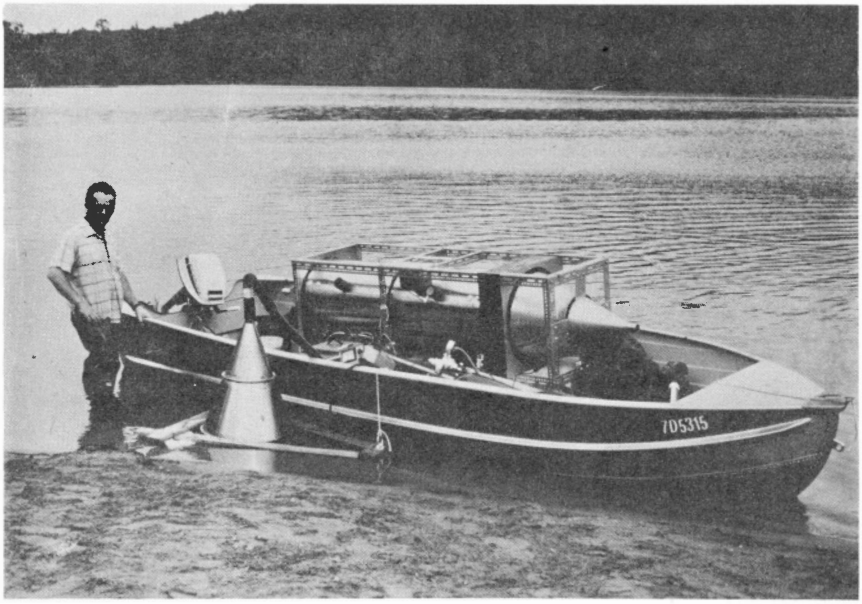

Figure 2. The ionization chamber is being readjusted for radon measurements over a lake.

the chamber. When in operation the air is forced through the chamber at a rate of about 400 litres per minute with a blower built into the intake port. A 240-volt photographic battery provides the accelerating voltage for the ions in the chamber. The negative terminal is connected to the outer electrode. The ion current is taken off the centre electrode by means of a vibrating capacitance electrometer across a 10^{12} ohm resistor and recorded on a chart recorder. In order to detect small variations in the current (of the order of 10^{-15}A) generated by the radon in the chamber in the presence of much larger background currents (of the order of 10^{-13}A) caused by gamma-rays in the vicinity and by junction potentials, a differential amplifier is used to amplify the fraction of the total electrometer output above the fairly constant background in any one location. This background is determined by passing radon-free air, i.e. air which has been stored for a month, through the chamber. The increased random fluctuations resulting from this amplification are damped with a 100-second time constant circuit described by Bristow (1972). With this time constant and the time constant built into the electrometer the unit has an overall time constant of about 3 minutes. Hence, in about six minutes the instrument reads about 97 per cent of the signal strength generated at the input. Since the radon content in air fluctuates only slowly this time constant is satisfactory.

In the field the electrical components are driven by two 12-volt 50-ampere-hour acid storage batteries via inverters. A field test in progress is shown in Figure 2. The inverted cone in the foreground is used to trap air which is forced into the water from a compressed air cylinder. The trapped air together with atmospheric air is then taken up by the blower and forced through the filter (rectangular box near the right end of the chamber) into the ionization chamber.

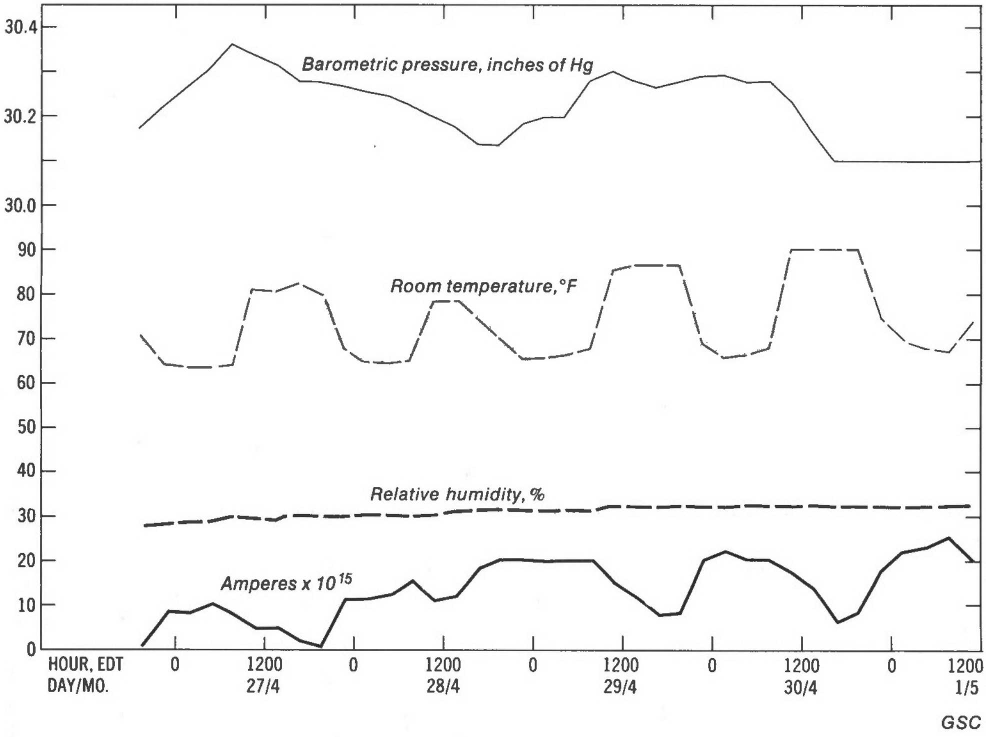

Figure 3. Effect of temperature on ionization chamber current output.

Calibration of the Ionization Chamber

The accuracy of the ionization chamber was determined in the laboratory by making a series of tests to determine the effect of temperature, humidity, and barometric pressure as well as radon content on the signal output using a closed loop airflow or no airflow at all. The effect of temperature on the current output is shown in Figure 3. This test was carried out in a room where the temperature cycled daily between 60 and 90°F. No air was passed through the chamber. It is obvious that an inverse correlation between temperature and electrometer current exists. Zero on the current scale in Figure 3 corresponds to approximately 160 to 180 mpA. This background current, which is quite constant, is due to gamma radiation in and adjacent to the chamber, beta and alpha decay of particles on the inside of the chamber, cosmic radiation, and junction potentials in the joints of the electrometer and chamber electrode assembly. Thermal noise does not account for the observed temperature-current correlation. This was ascertained by increasing the external gamma-ray intensity until the electrometer output had risen by a factor of ten and observing the current temperature cycles. Unexpectedly a tenfold increase in the change in current per degree of change in temperature accompanied the increase in gamma radiation level. It appears as though the ionization efficiency and/or the ion collection efficiency rather than thermal noise is responsible for the observed temperature-current correlation. In any event, for accurate measurements the temperature effect should be taken into account; after all a 30°F drop appears as a 0.08 pc/1 radon signal on the recorder at a background of 160 mpA.

The effect of humidity in the air on the ionization chamber current was determined by circulating air through the chamber in a closed loop in series with a box which contained a relative humidity meter and adding water vapour to the circulating air by forcing a small stream of air through boiling water into the closed loop. The results of three such tests are summarized in Table 1. During runs 1 and 3 the temperature of the circulating air remained fairly constant; during run 2 it dropped by 10°F cancelling the humidity effect. Humidity, therefore, has a small effect on the ion current in the range of 30 to 60 per cent relative humidity. Very humid air also leads to some instability in the functioning of the electrometer at the high input impedence of 10^{12} ohms used in the electrometer.

Table 1
Effect of humidity on ionization chamber current at constant temperature and pressure

Time, hr.	Test 1		Test 2		Test 3	
	ΔI, mpA	%, R.H.	ΔI, mpA	%, R.H.	ΔI, mpA	%, R.H.
0	0	31	0	28	0	34
1	- 4	36	1	38	-4	36
2	- 3	40	0	46	-4	43
3	- 5	45	0	50	-6	53
4	-10	53	0	55		
5	- 6	60				

Barometric pressure changes do not seem to affect the output appreciably. Small positive, negative, and zero correlations were observed during trial tests. One could argue that since a drop in barometric pressure decreases the amount of air in the chamber fewer collisions per cm of path of gamma-rays would occur resulting in lower ion current. But a corresponding drop in temperature which usually accompanies lows could compensate for the drop in pressure. Continuous monitoring of the two variables temperature and barometric pressure along with atmospheric radon over several months have not revealed noticeable correlations between radon and barometric pressure.

It should be noted that a good absolute filter is absolutely essential, for even smoke particles from combustion engines or cigarettes will cause a signal many times larger than that generated by the radon content in the air.

No accurate thorium standard was available to determine the thoron sensitivity of the chamber. However, one can arrive at an estimated sensitivity by considering the fate of the decay products of Tn and Rn. Considering alpha decay only (the energies of the beta and gamma decays are negligible in a rough estimate) we have the following decay schemes:

$$Rn^{220} \xrightarrow[54\text{ sec}]{6.3\text{ Mev}} Po^{216} \xrightarrow[0.16\text{ sec}]{6.8\text{ Mev}} Pb^{212} \xrightarrow[10\text{h}]{\text{beta}}$$

$$Rn^{222} \xrightarrow[3.8\text{d}]{5.5\text{ Mev}} Po^{218} \xrightarrow[3.1\text{ min}]{6.0\text{ Mev}} Pb^{214} \xrightarrow[26.8\text{ min}]{\text{beta}} Bi^{214} \xrightarrow[19.7\text{ min}]{\text{beta}}$$

$$Po^{214} \xrightarrow[10^{-4}\text{ sec}]{5.3\text{ Mev}} Pb^{210} \xrightarrow[22\text{ yrs.}]{\text{beta}}$$

In the case of thoron we get an energy expenditure of 6.3 Mev from the decay of Rn^{220} and 5.9 Mev from Po^{216} for a total of 12.2 Mev. The energy from the decay of Po^{216} is derived as follows: from the drift velocity of the Po^{216} ions, its half-life and average distance from the chamber wall one can show that about three quarters of the Po^{216} atoms decay before reaching the wall and one-quarter reach the wall before decaying. Hence, 75 per cent of the Po^{216} atoms will decay in the chamber air depositing 6.8 Mev of energy, and for the remaining Po^{216} atoms decaying in the chamber wall, 12 1/2 per cent will emit alpha particles into the chamber air depositing 6.8 Mev in the chamber, and 12 1/2 per cent will emit alpha particles into the chamber wall and not be detected. Thus, the energy deposited by Po^{216} in the chamber air is 87.5 per cent of 6.8 Mev, or 5.9 Mev/disintegration. For measurements lasting one hour or less the last 2 alpha emitters in the Rn^{220} chain, Bi^{212} and Po^{212}, do not contribute significantly to the energy expenditure in the chamber air due to the relatively long half-life of Pb^{212}.

In the case of radon and its decay products, the Rn^{222} expends 5.5 Mev in the chamber air, but Po^{218} will drift to the wall before decay, and thus its decay will deposit only 50 per cent of 6.0 Mev or 3.0 Mev in the chamber air, and Po^{214}, also deposited in the chamber wall will expend only 50 per cent of 5.3 Mev or 2.65 Mev. However, due to the half-lives of Pb^{214} and Bi^{214}, only about half of the equilibrium activity of Po^{214} will be attained in a one-hour measurement, and hence only 1.32 Mev will be deposited in the chamber air. Thus, a total energy of (5.5 + 3.0 + 1.3) = 9.8 Mev per decay of Rn^{222} plus daughter nuclides is realized. Hence, the relative sensitivities for thoron and radon should be about 1.2 to 1. From this it follows that the thoron sensitivity of the ionization chamber should be about 0.0037 pc/l/mpA for measurements which take less than an hour at a total current output of about 160 mpA. This value has been used in this report where thoron concentrations are given.

Results and Discussion of Field Tests

Essentially three kinds of field tests were carried out during the past summer; (1) spot checks of radon levels in air in the vicinity of and somewhat removed from uranium mineralization; (2) radon level measurements on the surface of lakes; and (3) continuous monitoring of radon in the atmosphere over an extended period of time in one location.

The spot checks were carried out on four different days at Pinks Lake and Fortune Lake in the Gatineau Hills, Quebec, and are summarized in Table 2. The air for these tests was obtained from a height of 30 feet. At this elevation measurable thoron levels were observed also. In view of the strong diurnal fluctuations in the radon content (see Fig. 4) the results in Table 2 are only indicative of the sort of concentrations one is apt to encounter in these areas. However, the average Fortune Lake readings are essentially the same as the Pinks Lake readings even though they were obtained after the Pinks Lake readings shortly after noon when the diurnal cycle reaches a minimum. One would not conclude from the results in Table 2 that the Fortune Lake area was uranium-bearing and the Pinks Lake area not. However, within two feet from the ground the concentrations of radon and thoron increased considerably; in fact, in the Fortune Lake area the radon levels were usually so high as to contaminate the ionization chamber.

Table 2
Spot checks on the radon and thoron concentrations in air at an elevation of 30 feet above ground in two locations in the Gatineau Hills, Quebec. Radon concentrations in pc/l

Date	Pinks Lake			Fortune Lake		
Da/Mo/Yr	Radon	Thoron	Wind	Radon	Thoron	Wind
19/05/72	0.050	0.018	E., light	0.072	0.012	E., light
07/06/72	0.015	0.033	E., moderate	0.086	0.026	E., moderate
20/07/72	0.170	0.046	none	0.094	0.052	S., light
27/07/72	0.075	0.028	none	0.072	0.059	W., light

To test the possibility of using the ionization chamber to make on-site radon measurements of lake waters, the assembly was put in a 16-foot boat and taken on a lake. A very crude arrangement permitted compressed air to

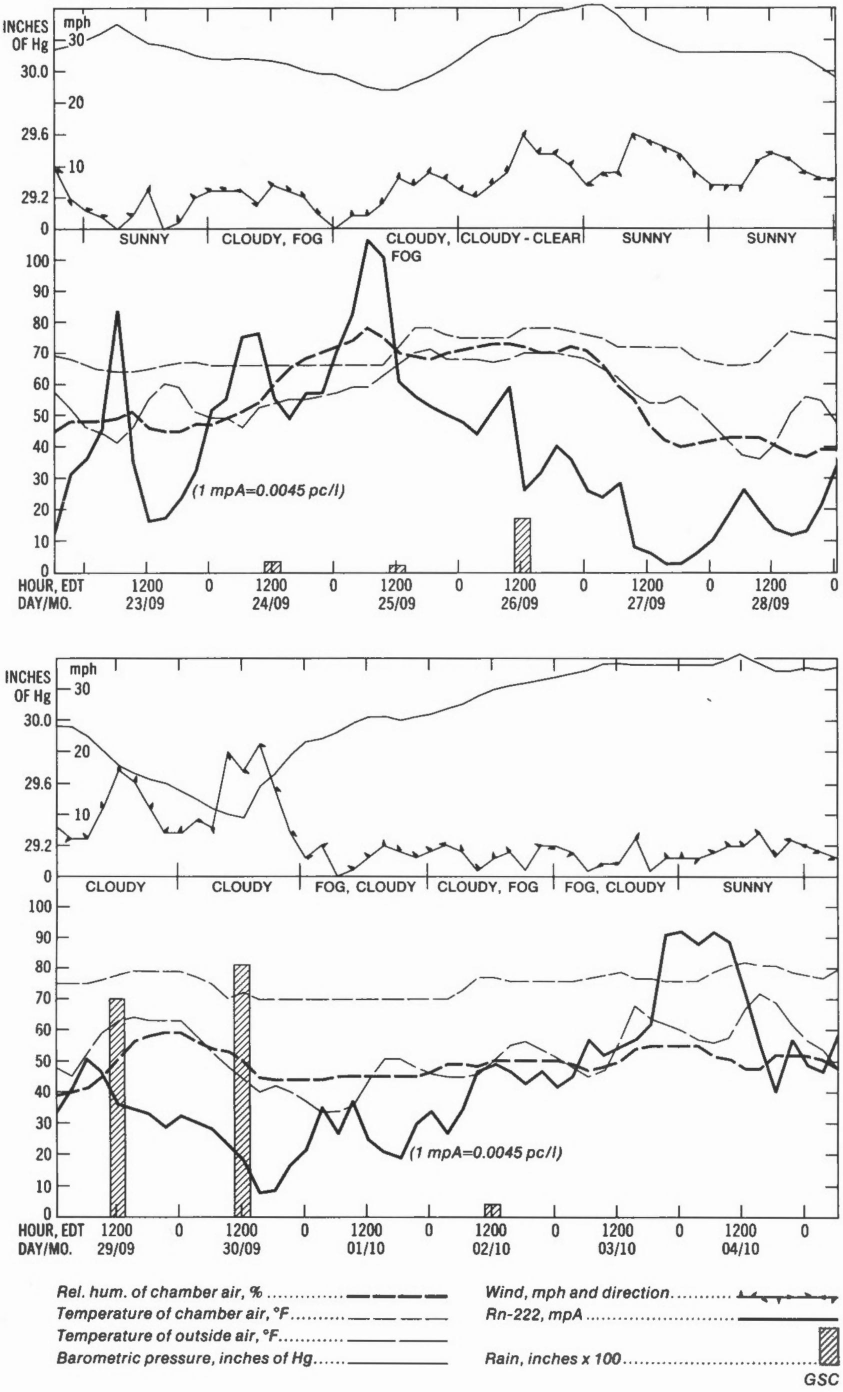

Figure 4. Radon-222 content of atmospheric air over Ottawa, Canada and meteorological variables during September 22-October 4, 1972.

be blown through the lake water and thence through the ionization chamber. For comparison several water samples were taken also and analyzed for radon by the conventional method. The results of two such tests are summarized in Table 3. The makeshift airflow arrangement permitted only qualitative tests and because of the slow response time of the instrument the readings were taken with the boat anchored. However, radon levels in the water were detectable with ease and were much higher than the radon levels in the air. Hence a smaller chamber of perhaps one-third the present size with a faster response time might prove useful in on-site radon measurements in lakes.

Table 3

Comparison radon contents in surface lake water and air in the Gatineau Park, Quebec. Radon concentrations in pc/l

Position of sample	Air		Water (Determined in Laboratory)	
	Kingsmere L.	Meach L.	Kingsmere L.	Meach L.
Three feet above lake surface	0.11	0.12		
At lake surface	0.15	0.18		
Air through water, near shore*	0.51	0.63	7.8	5.2
Air through water, near centre*	-	0.43	3.9	1.8

*Air was forced with 5 psig pressure into Kingsmere Lake and at 15 psig into Meach Lake; at 5 psig only a very small signal was recorded in Meach Lake.

Two selected time intervals during which radon was monitored continuously for a number of days, one portraying a relatively high and the other a relatively low radon activity in the air, are shown in Figures 4 and 5. The results were obtained with air taken from 85 feet above ground at the Geological Survey of Canada building in Ottawa. At this elevation no thoron activity was detected even on windy days so that the air could be admitted to the ionization chamber without having to go through a thoron decay chamber first. The long-term stability and ease of operation of the ionization chamber are best utilized in this type of measurement. The set-up can run for days or even weeks with a minimum of attendance. Only a brief daily check is required to see that neither the chart paper or the ink has run out. With the radon values are also plotted pertinent meteorological data* which show clearly the effect they have on the general radon content in the atmosphere. Warm, and particularly hazy days with light southerly winds coincide with high radon levels. Clear or rainy periods with northerly winds coincide with radon lows. Also clearly evident is a diurnal radon cycle peaking at about sunrise except on cloudy or windy days. No doubt, the quiet night followed by

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*Data obtained at Ottawa International Airport by the Department of the Environment, Atmospheric Environment Service.

the temperature inversion which takes place on clear days shortly after sunrise are responsible for this cycle. Similar atmospheric radon patterns have been reported by Gold _et al_. (1969).

It is clear from the complexity of the atmospheric radon pattern at any one location that one ionization chamber will not suffice for the outline of uranium-rich regions. However, a string of self-sustaining radon monitoring stations could eventually show the way to such regions.

The tests to date show that it is possible and convenient to monitor atmospheric radon continuously with a large ionization chamber. Technical difficulties prevent its use in a moving vehicle at this time. Perhaps a more rigid construction to prevent flexing of the chamber wall and an anticoincidence arrangement of two coaxial ionization chambers to cancel the varying gamma-ray background in a moving vehicle, will someday permit in-flight monitoring of radon.

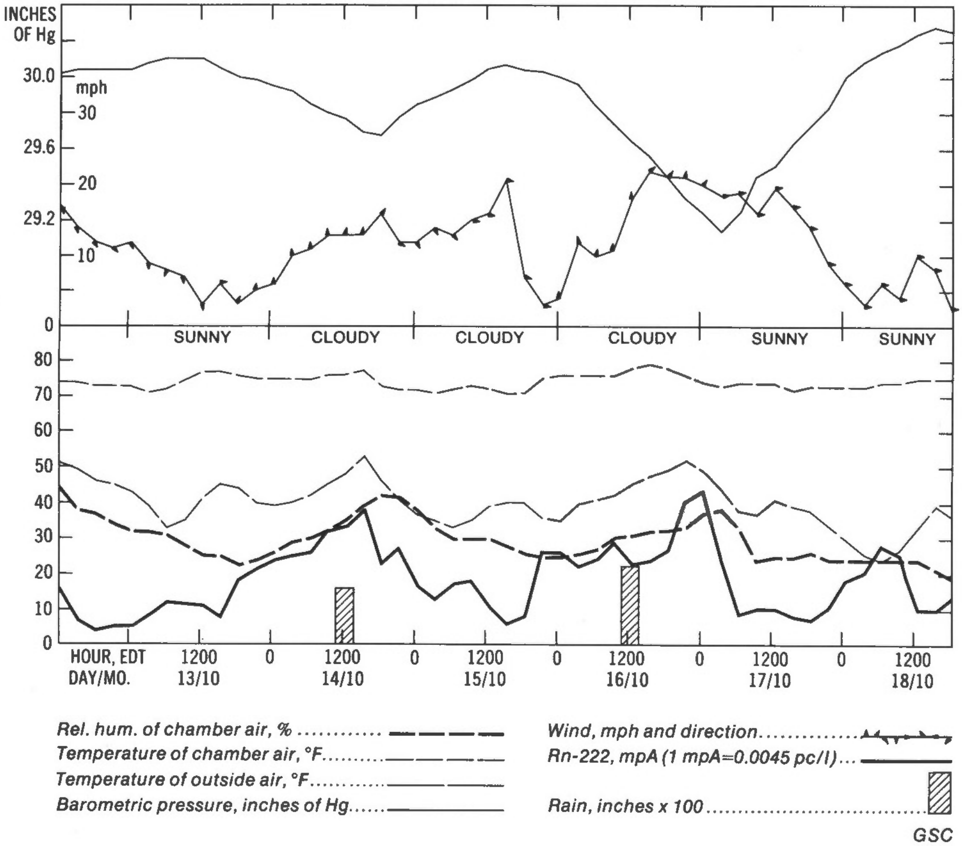

Figure 5. Radon-222 content of atmospheric air over Ottawa, Canada and meteorological variables during October 13-18, 1972.

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