A Technique for Measurement of Xenon-133 Air Contamination

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A sensitive, inexpensive technique is described for measuring xenon-133 air contamination to levels below nonoccupational exposure limits. The method uses a column containing a measured amount of activated charcoal through which an air sample is passed. The relative effects of air flow rate, xenon concentration, air temperature, relative humidity, and charcoal weight on the amount of trapped xenon are described.

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Xenon-133 is used in nuclear medicine laboratories for measuring lung function. The low solubility of noble gases in blood and other tissues is well known (*1,2*). The leakage of xenon gas around a patient's mouthpiece, and from syringes or from other equipment used to hold or dispense xenon, has been described (*3*). In order to document that laboratory airborne xenon-133 does not exceed occupational limits (10^{-5} μCi/cc) routine monitoring is required. In addition, monitoring should be performed in areas adjacent to the laboratory to ensure that nonradiation workers are not exposed to levels above their legal limits (3×10^{-7} μCi/cc). This is particularly important when the air-conditioning and heating systems are shared by several departments. If waste xenon is vented to the atmosphere, areas accessible to nonradiation workers or the public should be monitored periodically for xenon. A routine monitoring program should have a sensitivity capable of measuring concentrations down to 3×10^{-7} μCi/cc, the maximum permissible concentration for uncontrolled areas. Commercial devices that cost in the range of $2,000 are generally not suited to accurate measurements at concentrations of 3×10^{-7}. There are a number of published methods for laboratory monitoring of Xe-133 (*3–6*). A very sensitive technique is to use an unshielded or partially shielded sodium iodide or GM detector calibrated with a small source of Xe-133 (*3,4*). In nuclear medicine laboratories the disadvantage of this technique results from interference by scattered Tc-99m radiation. The equipment, moreover, is not readily portable. Counting collected air samples avoids this problem but the method's sensitivity is so low that detection at nonoccupational limits is difficult.

Activated charcoal has been used for a number of years by both the nuclear industry to trap exhaust gases and in nuclear medicine laboratories to trap expired Xe-133. This paper reports the results of our use of activated charcoal to provide a simple and inexpensive method to measure Xe-133 air concentrations to below nonoccupational limits. Variables investigated include the amount of trapped xenon as a function of Xe-133 concentration, charcoal weight and mesh size, air temperature, flow rate, and humidity.

METHODS

A 30-liter neoprene latex balloon was used for

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mixing known amounts of Xe-133 in calibrated air volumes. A variable-speed pump pulled air samples from the balloon through a column containing activated charcoal, and the pump's exhaust air was collected in another balloon for disposal. The speed adjustment for the pump was calibrated for air volume, and the flow rate was varied from 57 to 3800 cc/min.

The columns were made in 5-cc disposable plastic syringe barrels from which the flange at the end had been removed to permit entry into a standard well counter. Charcoal† of mesh sizes 8/16 (coarse) or 12/28 (fine) was used to build the column, and was retained in place by cotton plugs at each end of the column. At the completion of the trapping procedure the columns were closed with a stopper at one end and a stopcock at the other.

A one-g charcoal-column filter was placed on a 3-in. NaI detector connected to a 1024 multichannel analyzer operated in the multiscale mode. Time-activity curves were generated for the column's washin and washout. This procedure was used to determine the amount of xenon-air volume that must be passed through the column to reach equilibrium or saturation—i.e., the point at which passing more gas through the column would not result in increased trapping. To determine whether temperature affected the saturation point, curves were obtained a) at room temperature, b) after heating the xenon-air mixture and charcoal column to 39°C, and c) after cooling it to 4°C in a refrigerator. For all subsequent testing the amount of trapped xenon was determined at the saturation point.

Xenon-133 concentrations from 10^{-7} to 10^{-3} μCi/cc were produced in the source balloon. To produce a known concentration in the source balloon, Xe-133 was placed on a charcoal column which was then flushed by a measured amount of air into the balloon. The net amount of Xe-133 transferred to the balloon was determined by counting the charcoal column in a NaI well detector before and after flushing. The 3- by 3-inch well detector was calibrated with a Cd-109 standard. Branching ratios of 0.36 and 0.043 were used for the 81-keV gamma of Xe-133 and the 87.7 keV gamma of Cd-109, respectively. Cadmium-109 and Xe-133 activities were recorded using a 1024 multichannel analyzer, with the photopeak area integrated from 71–91 keV for Xe-133 and 78–98 keV for Cd-109. The amount of trapped xenon was normalized by dividing the cpm of trapped xenon by the concentration in μCi/cc of the xenon-air mixture in the source balloon.

The amount of trapped xenon as a function of charcoal weight was determined using columns containing 0.26, 0.45, 0.86, and 1.73 g of activated charcoal. The columns were counted with a 12- by 4-in. NaI detector at a distance of 2 in. The normalized count rate per gram of charcoal was calculated.

All other determinations were made using one-g charcoal columns, counted in a 3- by 3-in. NaI well detector. A column of this size extends up to 1 ⅛ in. above the bottom of the well, producing a geometric counting efficiency 4% less than a 1-cc source. This change in geometric counting efficiency was determined by comparing the count rate of a small amount of Tc-99m diluted in 1 cc of water with the count rate after an additional 2 cc was added. A 1-cc source was assumed to be equivalent in geometry to the commercial Cd-109 rod standard that was used for calibration of the Xe-133.

Measurements were made to estimate the effect of a possible decrease in the amount of trapped xenon resulting from sampling outside air at a higher temperature and relative humidity as compared with laboratory air. To alter temperatures, the source balloon and charcoal column were placed in an oven at 32°C or 44°C and allowed to equilibrate for 30 min, at which time the heated xenon–air mixture was pumped through the charcoal column. Measurements were made on the same column at 23°C, before and after the heated measurements, to document the temperature affect. The procedure was repeated using a second column, and the results averaged. To alter relative humidity, 10 cc of tap water was added to the source balloon and measurements were made after the water vapor was allowed to equilibrate for several hr with the balloon gas. It was assumed that the relative humidity within the balloon was close to 100%. Laboratory relative humidity was normally much less than this, with a range of 48 to 52%.

To determine whether the amount of trapped xenon would be significantly altered by the passage of large volumes of air, two identical columns were tested. After the initial measurements, both columns were flushed with 4 liters of laboratory air. One column was then attached to the pump, which then operated continuously at 3000 cc/min for 16 hr, thus providing an air volume equivalent to about 1000 individual tests. It and the control column were then tested the next day for differences in Xe-133 trapping. Comparison of the two columns permitted analysis of the effect of air volume on the amount of trapped xenon, independent of any changes in the xenon concentration in the source balloon that might occur over the 24-hr period required for the test.

Measurements were made to determine the amount of Xe-133 leakage from the source balloon and the charcoal columns. Sampling under identical

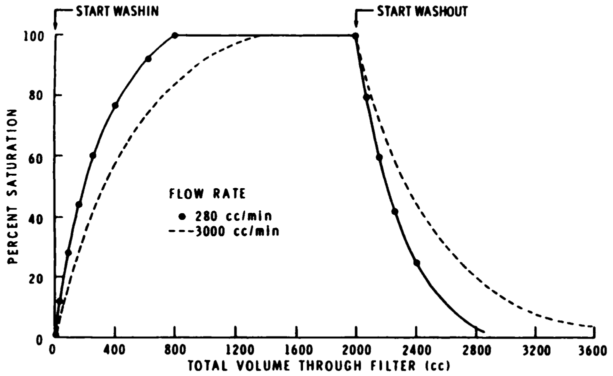

FIG. 1. Normalized buildup and washout of Xe-133 from a charcoal column using flow rates of 280 and 3000 cc/min.

conditions was repeated from the same balloon over a period of 3 days. Leakage was calculated from the percentage change in the trapped xenon after correcting for radioactive decay. Similarly, leakage from both stoppered and unstoppered columns containing trapped xenon was measured by repeated counting of the columns over a period of several days.

RESULTS

Leakage from the source balloon over a 3-day period was approximately 15%, indicating only minor loss from the balloon during the few hr required for these experiments. Leakage of trapped xenon occurred with a half-time of about 8 hr from an unstoppered charcoal column and about 40 hr from a stoppered column. No detectable amounts of Xe-133 leaked from the stoppered columns during the counting time required for each determination.

Figure 1 shows two normalized curves depicting the buildup and washout of xenon activity from a 1-g charcoal column as a function of the total volume of xenon-air mixture pumped through the column. Figure 1, which was obtained by fitting a smooth curve through the actual data points, is presented to demonstrate that although the volume required to reach equilibrium varies slightly with flow rate, equilibrium occurs after less than 2 liters have passed through the column at pump speeds of less than 3000 cc/min. Changing the temperature from 23°C to either 39°C or 4°C did not significantly affect the volume at which equilibrium was reached. Figure 1 also shows that xenon trapped in a column will be flushed from the column after passage of 2-3 liters of air. The significance of any residual activity will depend, of course, on the amount of xenon originally in the column.

TABLE 1. EFFECT OF CHARCOAL MESH SIZE ON Xe-133 TRAPPING, cpm/(μCi/cc), IN A COLUMN CONTAINING ONE g OF CHARCOAL*

Mesh size	Trapped xenon cpm/(μCi/cc)
Fine	2.2×10^8
Coarse	2.1×10^8

* Flow rate was 1660 cc/min and Xe-133 concentration was approximately 10^{-5} μCi/cc.

TABLE 2. EFFECT OF FLOW RATE ON XENON TRAPPING, cpm/(μCi/cc), BY ONE g OF CHARCOAL (FINE MESH)

Flowrate (cc/min)	Trapped Xe-133 cpm/(μCi/cc)
57	2.4×10^8
1660	2.2×10^8
3880	2.6×10^8

* Concentration of Xe-133 was approximately 10^{-5} μCi/cc.

TABLE 3. EFFECT OF LARGE QUANTITIES OF AIR PASSING THROUGH THE COLUMN ON XENON-133 TRAPPING

Liters of air		$\frac{\text{cpm in test column}}{\text{cpm in control column}}$
Control column*	Test column	
2	2	1.04
8	550	1.10
14	2350	1.07

* Two liters of air per determination; four liters to flush columns between determinations.

Altered conditions that did not effect the amount of trapped xenon at equilibrium are shown in Tables 1, 2, and 3. No significant difference was found for the two charcoal mesh sizes tested, or for changes in air flow rate. Flow rates of 57, 1660, and 3880 cc/min were tested. Passing large volumes of room air through a column resulted in no decrease in the amount of xenon trapped during subsequent testing, even when the amount of air passed through the column was the equivalent of about 1000 individual tests.

Intermittent exposure of the charcoal to xenon did not affect the amount of trapped xenon. The trapping was measured using a large syringe to pull the xenon-air mixture through the column in 200-cc increments. The values for two repeat measurements were 96% and 102% of the value obtained by the usual continuous-pumping method.

Shown in Table 4 are the equilibrium count rates per g of charcoal per unit concentration of the xenon-air mixture, as a function of the weight of

TABLE 4. EFFECT OF CHARCOAL WEIGHT ON XENON-133 TRAPPING*

Weight of charcoal	Trapped Xe-133 (cpm/g)/(μCi/cc)
0.26 g	2.7×10^8
0.45 g	2.5×10^8
0.86 g	2.6×10^8
1.73 g	2.6×10^8

* Cpm of trapped Xe-133 was divided by the grams of charcoal and the concentration of xenon-air mixture, (cpm/g)/(μCi/cc).

TABLE 5. EFFECT OF Xe-133 CONCENTRATION ON Xe-133 TRAPPING, cpm/(μCi/cc), BY ONE g OF CHARCOAL (FINE MESH).*

Concentration μCi/cc	Trapped Xe-133 cpm/(μCi/cc)
1.4×10^{-7}	2.4×10^8
1.4×10^{-6}	2.6×10^8
1.1×10^{-5}	2.4×10^8
2.9×10^{-3}	2.3×10^8

* Flow rate was 57 cc/min.

charcoal. This demonstrates the direct proportionality between the amount of gas trapped and the grams of charcoal. Table 5 shows the xenon trapping of 1 g of charcoal as a function of xenon concentration. In each case at least 1.5 liters of the xenon-air mixture was pumped through the column at a flow rate of 57 cc/min. This shows that the equilibrium amount of trapped xenon is a direct function of the concentration of xenon in the air.

The equilibrium trapping of xenon per g of charcoal was determined at room temperature (23°C) and after heating the trap and source balloon to 32°C or 44°C. The amount of trapped Xe-133 decreased to about 84% at 32°C and to 60% at 44°C. The trapped amount was decreased 8% by the increase in humidity produced by the addition of 10 cc of water to the source balloon.

In ten experiments, using the same column but including different xenon-air concentrations and pump speeds, the mean cpm per g of charcoal per unit concentration of the xenon-air mixture was 2.5×10^8 cpm/(μCi/cc) with a standard deviation of 0.31×10^8. To calculate Xe-133 concentration (μCi/cc) of an air sample with our detector and window settings, the net cpm of a sample are divided by 2.5×10^8. A net count rate of 2500 cpm is equivalent to an air concentration of 10^{-5} μCi/cc, the occupational exposure limit, and 75 net cpm corresponds to a concentration equal to the nonoccupational limit. To determine the concentration of Xe-133 in the air with a well detector having a detection efficiency different from ours, the following expression can be used.

$$\mu\text{Ci/cc} = \frac{A}{E} 2.5 \times 10^{-9} \qquad (1)$$

Where A is the net cpm of the xenon sample, E is the photopeak detection efficiency including any difference in geometry between the standard and samples, and 2.5×10^{-9} is a constant. This expression was derived from the normalized amount of trapped xenon described above, 2.5×10^8 cpm/(μCi/cc), and applies to 1 g of charcoal of the type used in this paper, at room temperature and under equilibrium conditions.

DISCUSSION

Our results indicate that Eq. 1 holds for the concentrations of xenon in air tested, representing partial pressures in the range of 10^{-6} mm Hg. This is not true, however, for all partial pressures and temperatures (*7,8*). The decrease in the equilibrium value to about 60% of initial, for a temperature increase from 23°C to 44°C, agrees with published absorption isotherms (*7*). Correction factors based on air temperature could be employed when a very hot environment is to be sampled, but the correction is small for laboratory sampling where the temperature is within a few degrees of 23°C. To alleviate the temperature problem one could collect the sample at the desired sampling point with a balloon, take the sample to the laboratory and allow it to reach laboratory temperature before drawing it through the charcoal column. Other sources of possible error are mistakes in weighing the charcoal (which should be small) and uncertainty in the absolute value of the Cd-109 standard. The latter may be as high as ± 10% because of uncertainties in the branching ratio (*9*). The equilibrium value was reproducible within ± 12% (one s.d., including counting statistics), even when obtained with varying pump speeds, xenon-air concentrations, and charcoal mesh size. Variation in trapping efficiency because of changes in relative humidity is not felt to be significant for routine monitoring in the laboratory. This problem could be obviated by inserting a moisture trap. The overall error for any one measurement should be within 30%, which is adequate for routine monitoring.

The technique described yields about 75 net cpm for an air concentration of 3×10^{-7} μCi/cc. Typical background counting rates are 20 cpm, therefore a 10-min count of sample and background yields about a 3% counting error. Any number of samples can be counted automatically using most commercial gamma counting equipment. Much higher sensitivities could be achieved using larger volumes of charcoal but with increasing geometric uncertainty for well counters. Some other type of detector and sample geometry could be used—e.g., a flat NaI detector with a large charcoal column on top. However, this would not lend itself to automatic counting.

The pump can be used to draw air samples directly through the column or to obtain xenon–air concentration averaged over a period of time. For this purpose the charcoal column is removed from the pump intake while the pump exhaust is collected during the desired time interval. The balloon air sample is then connected to the charcoal column and 2–3 liters of the sample air are drawn through the column at a rate of about 1–3 liter/min. The calculated concentration is then an average value over the period of time sampled.

We believe that weekly, possibly daily, monitoring is required of several representative areas within the nuclear medicine laboratory. If levels above those for nonoccupational exposure are found, i.e., 3×10^{-7} μCi/cc, monitoring should be extended beyond the nuclear medicine laboratory. If waste xenon gas is vented to outside air, a survey should be performed at least every six mo at several points near the point of release and at other points more distant.

FOOTNOTES

† North American Carbon (type G210), North American Carbon, Inc., Columbus, OH.

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