

Two-Filter Radon Detectors With Aerosol Chambers: A Progress Report

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INTRODUCTION

The U.S. Bureau of Mines has developed a continuously measuring environmental radon monitor (ERM) that is about 50 times more sensitive than most commercially available instruments. The Bureau's monitor is a modification of the well-known, two-filter method (Thomas, 1972), and is based on the design reported by Whittlestone (1985). The main differences from the Whittlestone design are the way the aerosols are generated for the inside of the monitor and the size of the monitor. Identical to Whittlestone, the reason the aerosols are injected into the monitor is to reduce the radioactivity losses to the walls that, in turn, can increase the sensitivity more than 10 times compared to standard two-filter monitors.

Three main problems of these environmental radon monitors remain unsolved:

- (1) How to achieve a constant output aerosol generator - both in size and number.
- (2) How to find an aerosol that does not clog the second filter excessively so that it is not necessary, for instance, to change the second filter every day (Solomon, 1989), making the method impractical. Each change of the second filter, where the activity is measured, requires more than 1 hour waiting time to reach equilibrium before the instrument can start generating valid data.
- (3) How to understand and optimize the mechanism of losses of the radioactive aerosols to walls of the monitor as functions of turbulence, size of aerosols, and of ventilation rate.

EXPERIMENTAL

Figure 1 shows the schematic and figure 2 shows an overall view of the Bureau's environmental radon monitor. The total volume of the chamber is 400 l, and the monitor sensitivity for radon is about 300 cph/Bq/m³ (200 cpm/pCi/l).

The first filter is a 10.5 cm glass fiber filter, and the second a 2.5 cm glass fiber faced with a GM detector (marked D in figure 1). The key is the aerosol generator (atomizer) head described previously (Drouillard et al., 1984). The atomizer normally uses as an aerosol source a solution having a well-defined concentration of salts, usually fluorescein. A somewhat unexpected feature of the Bureau's ERM aerosol generator is the suitability of "distilled" water bought in grocery stores, which should contain no "salts," for aerosol generation. In figure 3, typical diffusion battery/condensation nuclei counter measured distributions of aerosols such generated are shown. Eighteen MΩ demineralized water used for the same purpose and filtered through 0.8μ filter gave similar results. A preliminary conclusion is that water (Bergh et al., 1989) and, according to our findings, even air, contain microorganisms (and their debris after passing through the atomizer head with a pressure drop of about 1.5 atmosphere) and that these are responsible for the aerosols

shown in figure 3. It has also been observed that the longer the source water stands, the greater the aerosol output from the generator (typically from $70 \times 10^3 \text{ cm}^{-3}$ to $110 \times 10^3 \text{ cm}^{-3}$). This consequent increase is presumably caused through microbial growth because, while in the reservoir, the water input into the atomizer is not filtered. A 1-gallon supply of distilled water lasts for a week or two, (depending on outside RH) providing sufficient time for the microorganism growth.

Gauge 1 measures detector filter differential pressure, and gauge 2 measures internal pressure in the chamber. This is achieved by means of the flow regulator (marked F in figure 1). To carry out the experiments to determine the aerosol wall losses for sizes around $D_p = 0.8 \text{ nm}$ (unattached radon daughters) two changes were made: (1) one two-way valve was added between the flow meter/regulator (F) and the second filter; (2) a filter/condenser and two filters, all in series, were inserted between the atomizer and the first pump (P1 on figure 1).

Sampling of unattached radioactive aerosol as well as of aerosols of sizes around $D_p = .018\mu$ was done through the sampling port shown in figure 1. The two-way valve was inserted so that no interference with the normal operation of the monitor took place during sampling; the outlet flow was just redirected through a sampling probe for 5 minutes and then returned to the detector (D in figure 1). The probe, when inserted inside the chamber, is only several centimeters away from the detector (D).

In order to achieve a condensation nuclei (CN) free atmosphere and thus pure unattached daughters inside the chamber (Holub, 1984), a filter/condenser and filters in two high-pressure holders had to be inserted between the pump and the atomizer (without the distilled water) to prevent any CN from entering the chamber. Consequent checks showed that CN levels were undetectable using a portable CN counter.

Po 218 radioactivity obtained using a modified Tsivoglou method (Thomas, 1978) was used to calculate the wall loss (plateout) rate, λ_p , using equation (1) (Holub, 1984):

$$\lambda_p = \left(\frac{Rn}{RaA} - 1 \right) \lambda_A - \lambda_v, \quad (1)$$

where RaA and Rn = concentration of 218 Po and Rn, respectively,

where λ_A = decay constant of 218 Po,

and λ_v = ventilation rate in the chamber.

Ten measurements were taken; activity levels of about $5,000 \text{ Bq/m}^3$ were obtained using a radon source of $22.3 \mu\text{Ci}$ in a flow-through mode. The error based on the 10 repeated measurements is about 7%, partly due to the temperature dependence of the radon source. These results are plotted as one point in figure 4, at size $D_p = 0.8 \text{ nm}$ and with appropriate error bars.

To determine the wall deposition (plateout) coefficient for aerosols about size $D_p = .018\mu$, the following procedure was used (Holub, 1984):

- (1) The monitor chamber was filled with the "pure" water aerosol to a level of about $4.5 \times 10^4 \text{ cm}^{-3}$ particles, after which the source of the aerosols was removed.

- (2) While all the parameters were held identical to normal operation of the monitor, condensation nuclei concentrations were measured every 3 to 4 minutes until all CN disappeared.

The results of these measurements are plotted in figure 5. Note that these aerosols (microorganism debris?) appear practically monodispersed in the range of the CN counter. Subtracting the ventilation rate, λ_v , equation (2) gives the wall loss coefficient

$$\lambda_p = \Lambda - \lambda_v, \quad (2)$$

where Λ = total disappearance rate of aerosols (s^{-1}).

The result of this experiment is also plotted in figure 4 at the appropriate size range, $D_p = .018\mu$ with estimated error bars. The curves plotted in figure 4 are based on the Crump and Seinfeld (1983) theory and are similar to ones used in our previous publication (Holub et al., 1989).

DISCUSSION

The curves and data in figure 4 show that for $n = 2.6$, a wide variety of turbulent systems behave in a similar way that can be described by a Crump and Seinfeld type of formula. Intuitively, it seems possible that $n = 2.6$ reflects the fractal nature of turbulence. It should be noted that the three systems shown in figure 4 are quite different in manners in which the kinetic energy rate per unit weight, ϵ , is delivered into the volumes in question: (1) glass sphere (Holub et al., 1989, Van Dingenen et al., 1989) where only the convection due to temperature differences between the wall and the inside air are present (total volume 0.2 m^3); (2) Bureau's radon chamber where the two fans provided almost four orders of magnitude more k_e (total volume 4 m^3); (3) Bureau's radon monitor chamber (total volume 0.4 m^3) where the jet from the first pump provides $k_e = 0.96 \text{ s}^{-1}$.

Unfortunately, the Crump and Seinfeld theory requires that $n = 2$. If not, the dimensional analysis shows that the eddy diffusion coefficient does not have the proper dimensions (m^2/s), rendering the theory meaningless. A major attempt to correct this situation by expressing the diffusion coefficient in terms of a dimensionless quantity reproduced the experimentally observed slopes; however, not the absolute values. A serious effort is under way at present to resolve this problem, and some results will be reported in the near future.

Controlling the number and the size of the condensation nuclei (CN), the flow rate, and the plateout rates of the unattached and attached radioactive aerosols allows one to optimize the system's performance, which are maximum sensitivity, convenient time resolution, and the stability to the usual and apparently inevitable ($\pm 30\%$) variations of the CN levels inside the chamber. Plans are being made to optimize the Bureau's detector in the near future. In the meantime, the environmental radon monitor was run at a flow of about 5 to 6 l/m, CN levels from 80 to 150 K/cm^3 with the resulting sensitivity of 300 cph/Bq/ m^3 (200 cpm/pCi/l). The duration of the experiment was about 2 years, during which the monitor performed without any serious problem at a site in downtown Denver (figure 6).

CONCLUSIONS

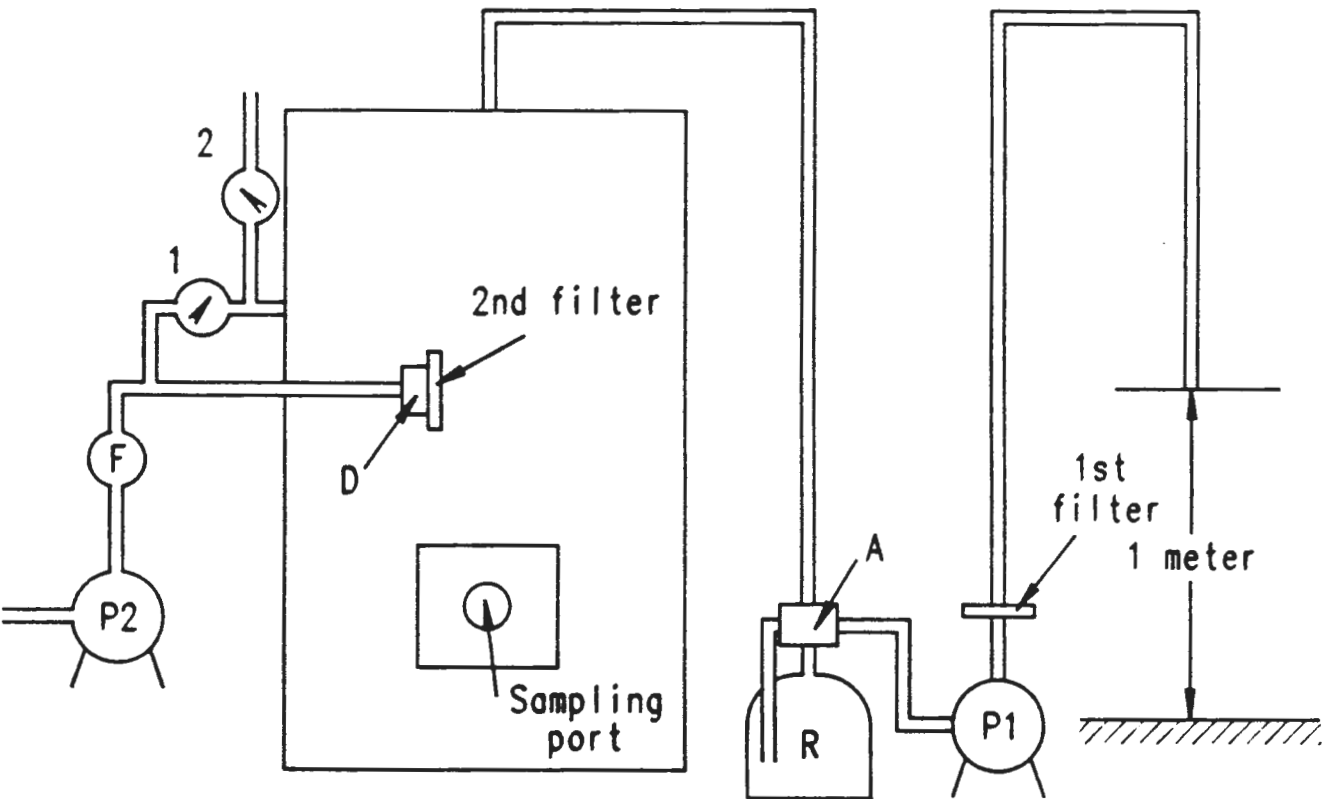
The Bureau's environmental radon monitor is suitable for measuring low levels of radon around uranium and other mines, as well as in any environment where radon levels are of concern.

Using standard aerosol science techniques and by means of two simple experiments, all relevant parameters necessary for a complete theoretical description were determined. This monitor had been performing successfully outdoors for more than 2 years.

REFERENCES

- Bergh, O., Borsheim, K.Y., Bratbak, G. and Heldol, M. (1989) Nature 340: 467-468.
- Crump, J.G., and Seinfeld, J.H. (1981) J. Aerosol Sci. 12: 405-415.
- Drouillard, R.F., T.H. Davis, E.E. Smith, and R.F. Holub, (1984), Bureau of Mines IC 8965.
- Gesell, T.F., (1983) "Background Atmospheric ^{222}Rn Concentrations Outdoors and Indoors: A Review," Health Physics 45, page 289, 1983.
- Holub, R.F., (1985) Rad. Prot. Dos 7: 155-158.
- Holub, R.R., Raes. F., Dingenen Van, R., and Vanmarcke, H. (1989) Rad. Prot Dos. Vol. 24: 217-110.
- Solomon, S.B. (1989) Private Communication
- Thomas, J.W. (1970) Health Physics 18: 113-122
- Van Dingenen, R., Raes, F., and Van Marcke, H. (1989) J. Aerosol Sci. 20: 113-122.
- Whittlestone, S., Health Phys. 69, (1985), 847-852

BUREAU'S ENVIRONMENTAL RADON MONITOR



KEY

A - Aerosol generator

R - Distilled water reservoir

P - Pumps, 1 and 2

F - Flow meter/regulator

D - Geiger Muller detector

⊗ - Gauges, 1 and 2

Figure 1. Schematic view of the Bureau's environmental radon monitor (2F-400).

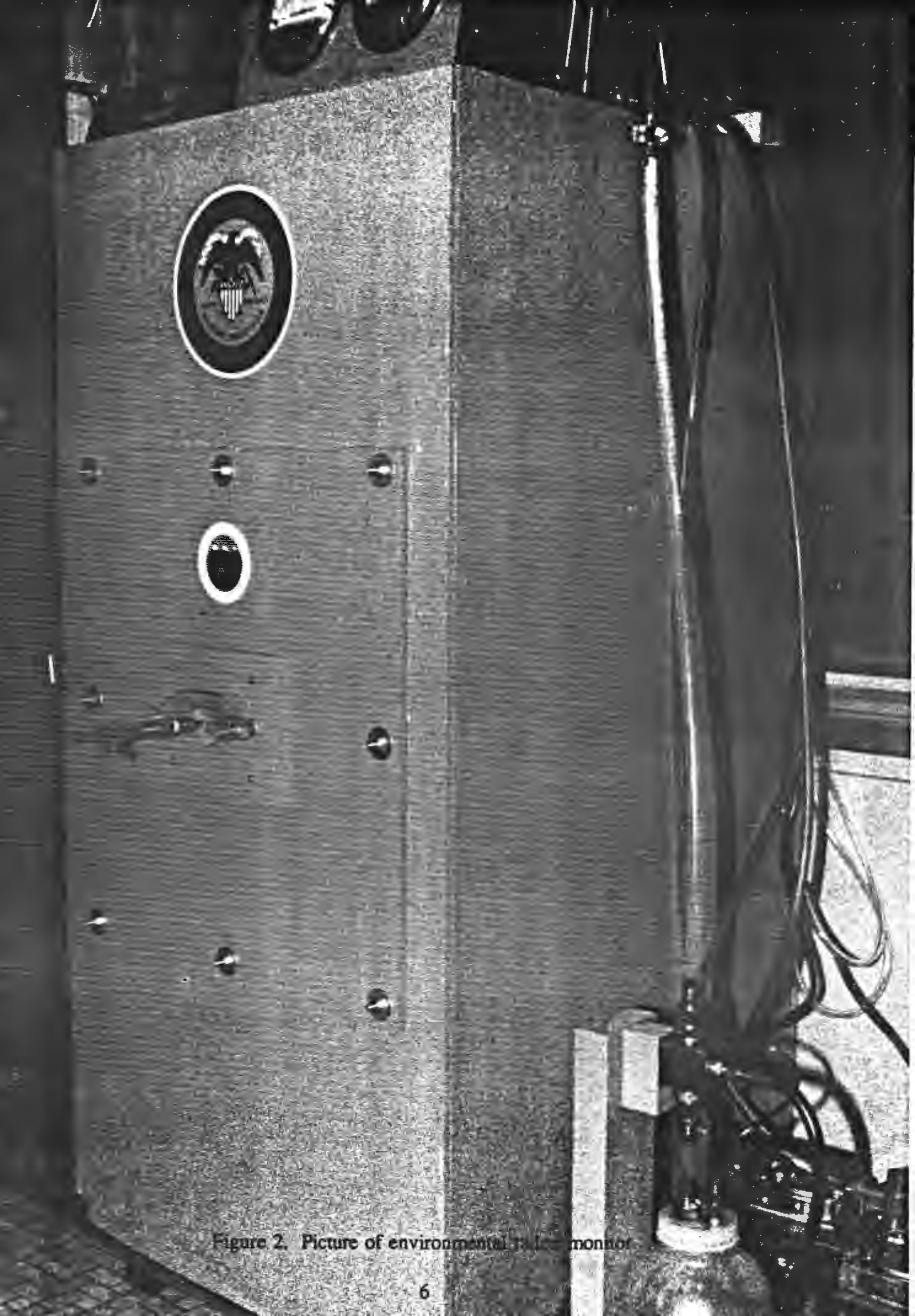


Figure 2. Picture of environmental test chamber

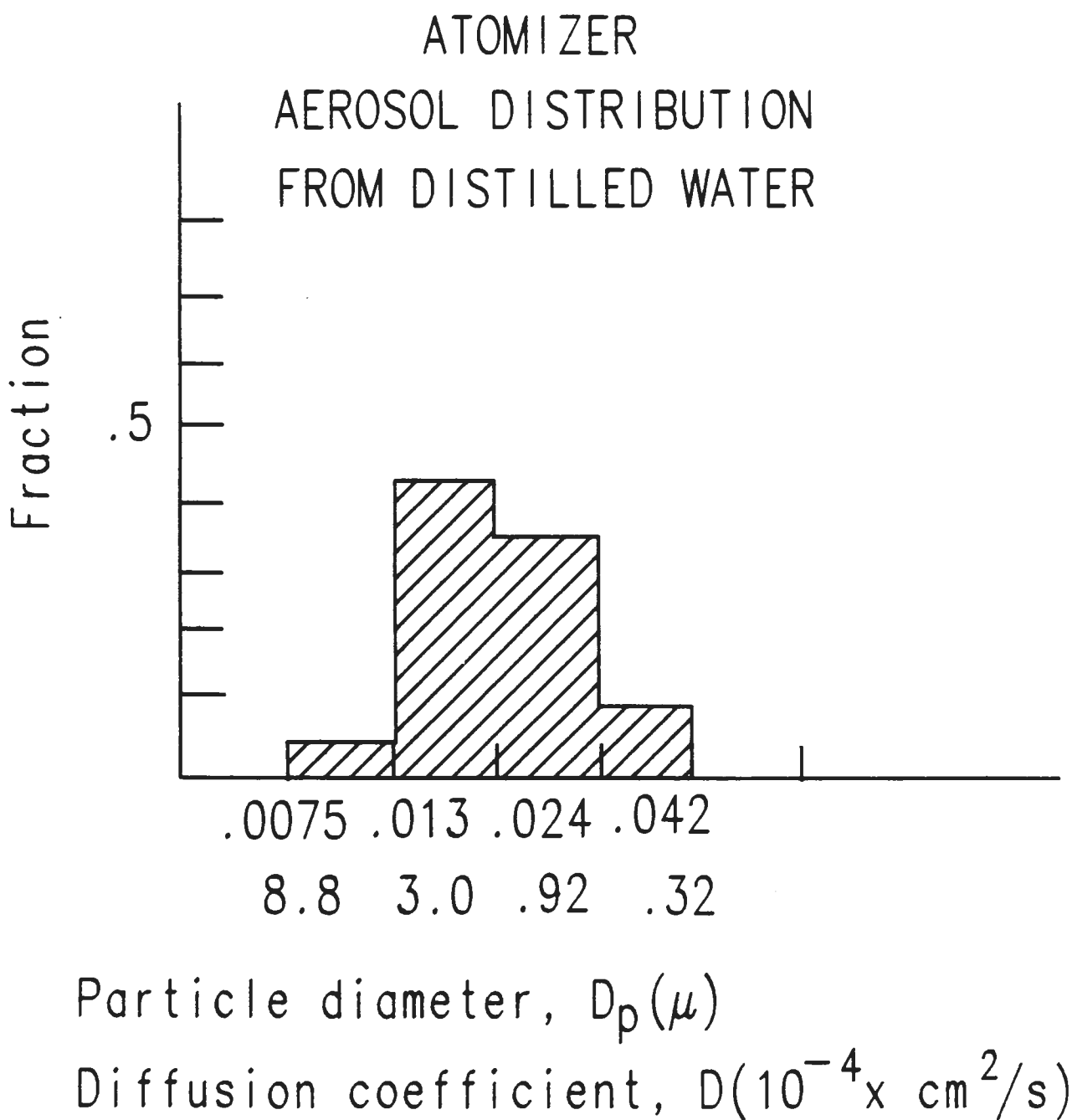


Figure 3. Typical diffusion battery condensation nuclei counter size distribution measurement of aerosols from pure "distilled water.

WALL DEPOSITION, β , FOR THREE TURBULENT SYSTEMS

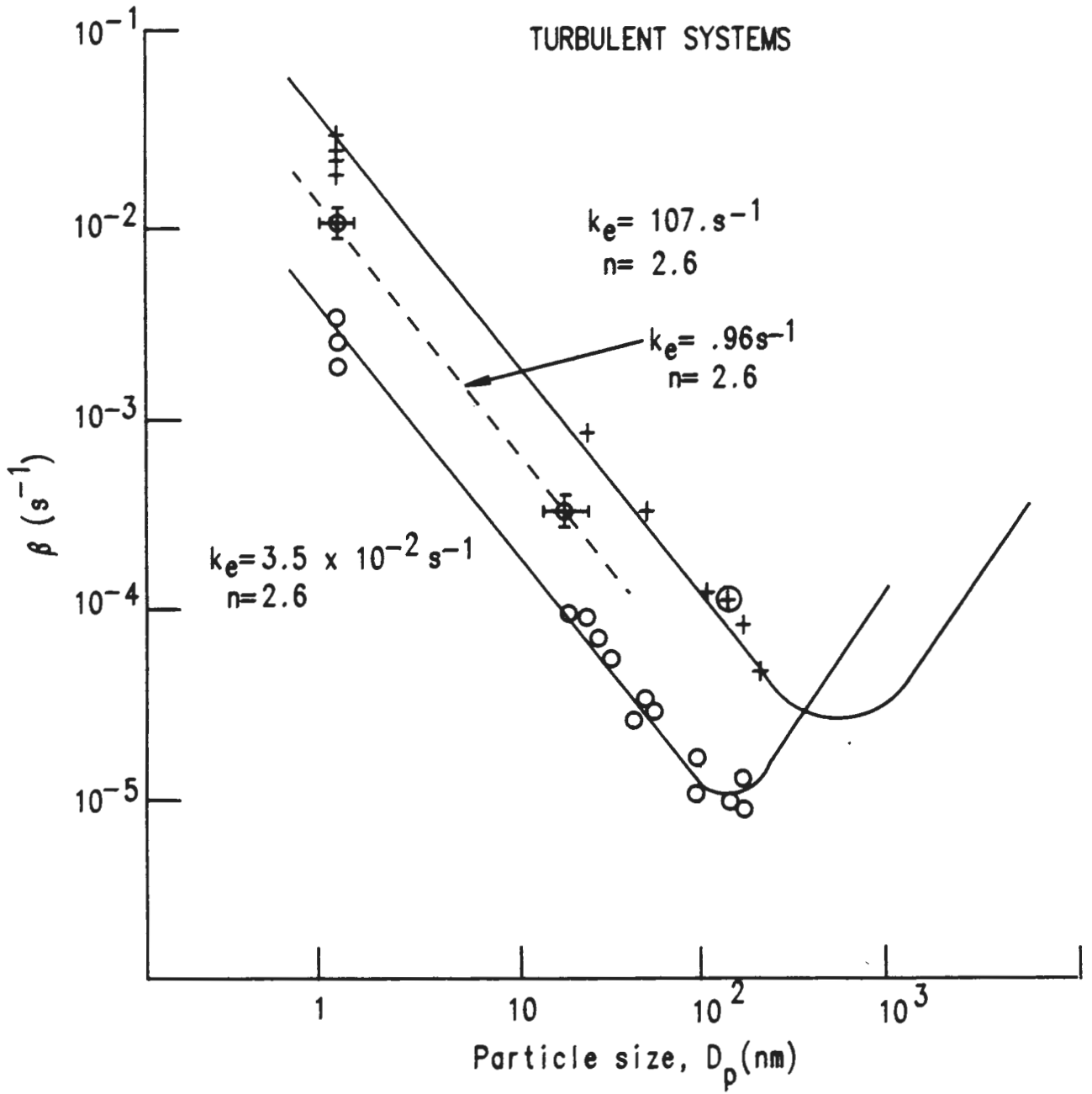


Figure 4. Wall deposition coefficient as a function of particle size for three different turbulent systems.

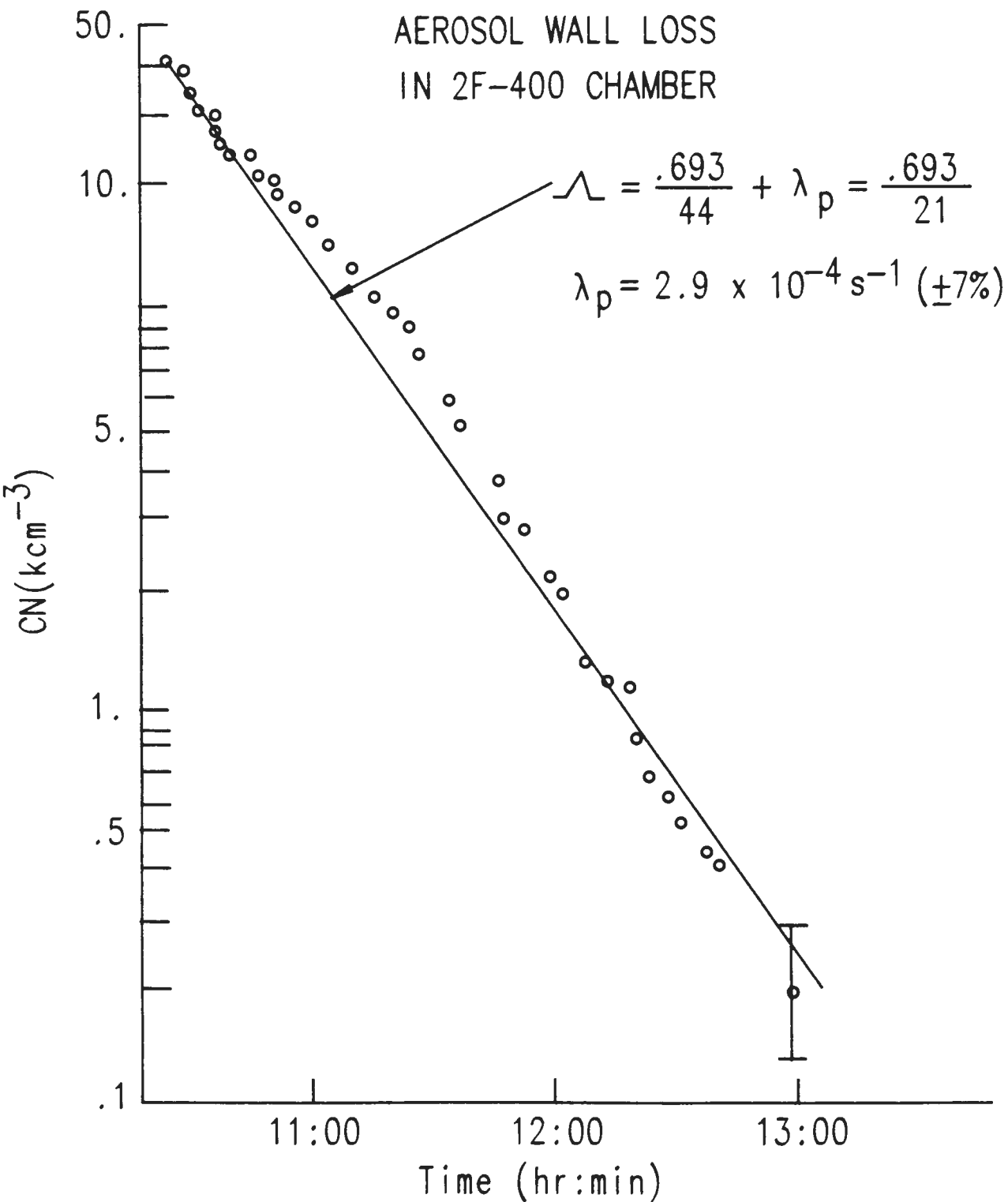


Figure 5. Condensation nuclei concentration as a function of time in the Bureau's environmental radon monitor chamber.

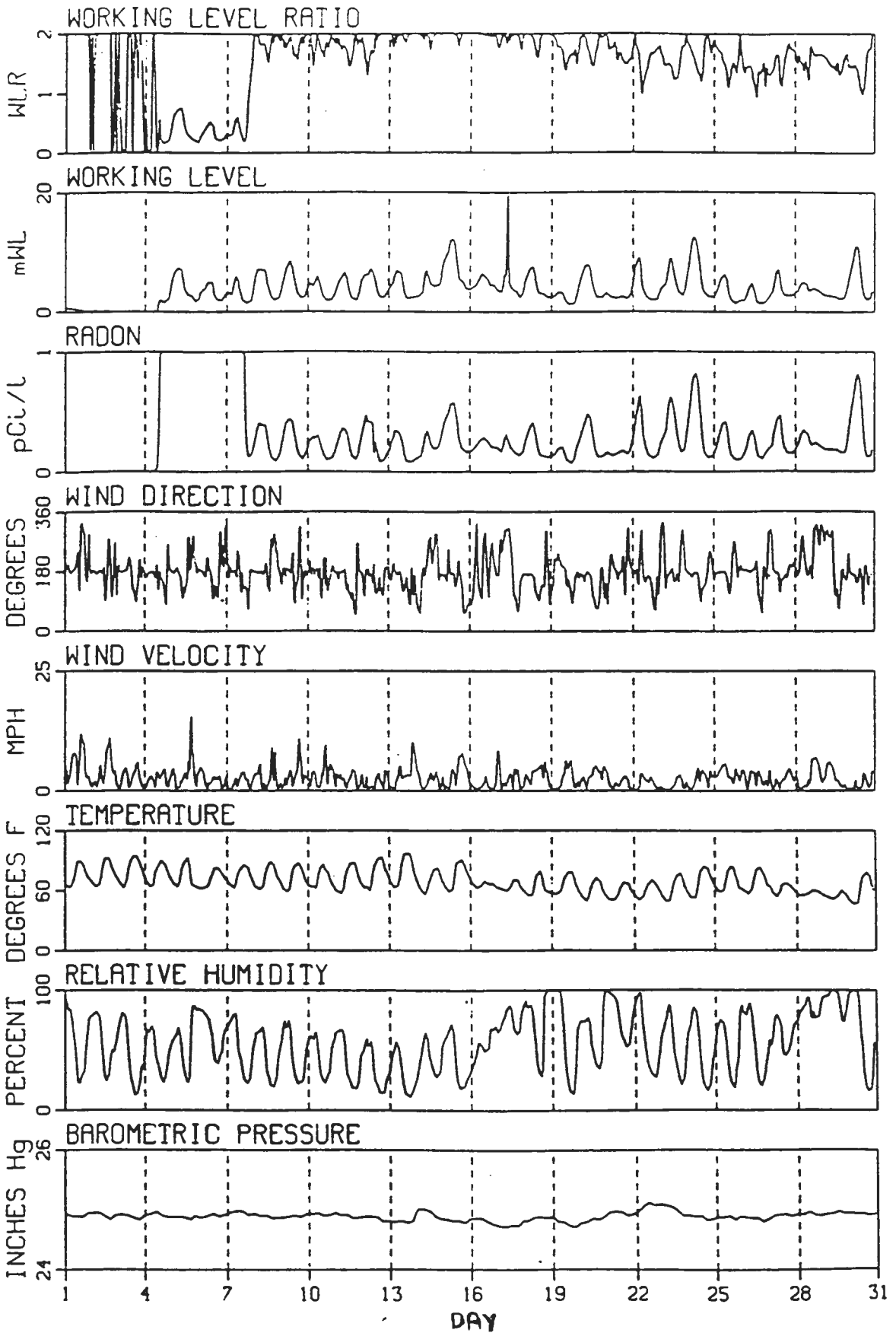


Figure 6. A typical set of measurements from the downtown Denver site.