# RECENT PROGRESS IN SAMPLING AND MEASUREMENT OF RADON AND THORON DECAY PRODUCTS

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***Abstract:*** **A new approach to sampling and measuring the unattached fraction of radon decay products is presented. Two methods of electrostatic sampling with polystyrene plates charged by friction or with copper plates connected to a high voltage source are compared to conventional sampling on wire mesh screens. A good correlation between Po-218 alpha activities for the three sampling methods for different radon concentrations and aerosol conditions was found. For total beta activities correlation between the methods is less obvious.**

## INTRODUCTION

The alpha-decay of Rn-222 and its recoil result in about 90% positively charged Po-218 ions. They have a high mobility, characterized by the diffusion coefficient, which controls the processes of aerosol attachment and deposition on surfaces. The diffusion coefficient of charged and uncharged Po-218 and Pb-212 has been studied with electric fields in diffusion chambers (1, 2). Despite the fast neutralization of the ions, collection is possible with a sufficiently high voltage. The efficient mean life time of free Po-218 ions due to decay, recombination and attachment is typically 12 s (3). Therefore it is possible to determine the Po-218 concentration by collecting the free ions. Likewise Pb-214 may be positively charged, but its amount is unknown, which makes the determination of the Pb-214 concentration by collecting the ions less certain. The recoil energy of the beta decay of Pb-214 to Bi-214 is not sufficient for ionization and thus there are no Bi-214 ions.

All radon decay products quickly attach to aerosol particles or deposit on surfaces. Airborne radionuclides are divided into an "attached" and an "unattached" fraction. Molecular clusters up to 1 nm in size are formed with water molecules and trace gases. The fraction with a thermodynamic diameter below 2 nm is defined as "unattached", the larger one as "attached".

Unattached radionuclides have a higher diffusivity and may be deposited on wire mesh screens. An alternative method is a collection on negatively charged plates, which attract the positively charged radon decay products. The unattached fraction is attracted much more effectively than the attached fraction, due to its much higher mobility. In the following the activity collected on charged plates is compared with the activity on wire mesh screens under different radon concentrations and aerosol conditions.

## MATERIALS AND METHODS

The alpha activity was determined by alpha spectroscopy with a 30 cm² surface barrier silicon detector in a vacuum chamber and a multichannel analyzer (EG&G, D-81677 München), and by a hand-held alpha spectrometer Alpha Analyzer with a 2" diameter silicon detector (SAIC, San Diego, CA 92121 USA). The beta activity was measured with the Living Level Monitor LLM 500 (Münchener Apparatebau, D-82024 Taufkirchen) with a 11 x 11 cm² proportional detector (4).

The attached fraction was sampled with glass fiber filters MN 85/90 (Macherey-Nagel, D-52348 Düren). The unattached fraction was deposited on wire mesh screens (Bucher, D-81669 München) with a mesh width of 0.04 mm and a wire diameter of 0.028 mm (147 mesh/cm = 374 mesh/in). Both have an effective diameter of 10 cm and are mounted in cardboard diskettes, which fit into the LLM. The wire mesh screen was fixed in front of the glass fiber filter on an air sampler (vacuum cleaner). The distance between the wire mesh screen and the glass fiber filter was 4 mm to avoid penetration by alpha-recoil-atoms. After 1 min of sampling at 250 L/min with the air sampler and 10 s transfer time, the filter and the screen were evaluated in the LLM within 1 min for beta activity and equilibrium equivalent radon concentration (EER). Additional screen samples were measured for alpha activity on the front side. 250 L/min through a 10 cm diameter screen yield a subcritical face velocity of 50 cm/s, for which the particle diameter for 50% collection efficiency is about 2 nm.

Collector plates for electrostatic collection are polystyrene plates (PS) of 130 x 200 x 2 mm³ charged by friction, "Philion Plates" (5), or Cu plates of 100 x 100 x 0.6 mm³, charged by a high voltage source. The PS plates were charged by rubbing with wood or wool. With charge saturation on the PS by appropriate rubbing, -20 kV are reproducibly achieved and confirmed by measurement with a "field mill" Static Monitor 140 C (John Chubb Instrumentation, Cheltenham, GL51 8PL, UK). The voltage on the plate decreases faster at higher radon concentrations. The Cu plate was connected to a constant voltage source of -16 kV. No significant differences between PS plates and Cu plates were found, so in the more results from the Cu plate are described, because they are assumed to be more convincing. After sampling for 1 min and transfer time of 10 s Cu plates were measured 1 min for beta and for alpha activity. Using large-area collectors and large-area detectors improves overall efficiency approx 100-1000 fold, relative to wires connected to a voltage source and an electrometer discharge first used by E. Rutherford in 1900 in the laboratory, and by J. Elster and H. Geitel in 1901 in the open air.

The samples were taken in a radon lab (46 m³), where uranium minerals were stored in air tight containers. The radon concentration was changed by opening and closing containers. The aerosol concentration was changed by burning candles and incense and filtering the air with an air cleaner Breatheasy (12 m³/min) (Biltwell, Mississauga, Ontario L5C 2Z2). The radon gas concentration was varied between 500 and 12000 Bq/m³.

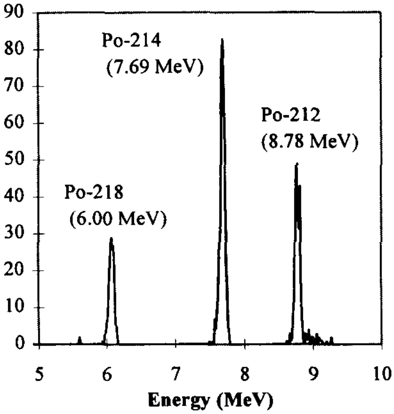


**Fig. 1.** Alpha spectrum of a Philion Plate, exposed to ambient air for 25 min; 3.7 h count with a 30 cm² silicon detector in a vacuum chamber

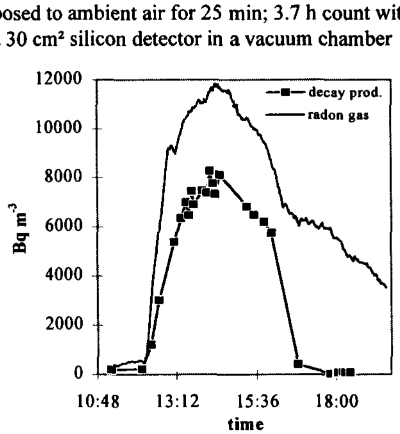


**Fig. 2.** Concentration of radon gas and EEC of radon decay products in the radon test room

RESULTS AND DISCUSSION

A problem with alpha measurements is the self-absorption in samples like glass fiber filters. Peaks in alpha spectra become very broad with crosstalk between Po-218 and Po-214 peaks. A problem with wire mesh screens is the dependence of the front to back ratio and screen losses on environmental parameters. Fig. 1 shows the alpha spectrum of a Philion Plate, which was exposed to ambient air for 25 min and measured with the 30 cm² silicon detector for 3.7 h in the vacuum chamber. The result is a neat spectrum of Po-218 and Po-214 from the radon decay series and of Po-212 from the thoron decay series. All decay products are on the surface of the plate, so there is no self-absorption like in glass fiber filters. Therefore it is possible to count the plates in open air with the SAIC Alpha Analyzer for Po-218 and Po-214 activities without crosstalk. The spectrum on a Cu plate is of similar appearance.

For fast measurements with good counting statistics, high radon levels were used. Fig. 2 shows the variation of radon gas, measured with an instrument Megarad, and of attached radon decay products EER, collected on glass fiber filters and measured with a LLM 500. The radon gas concentration was raised by opening a container with uranium minerals at 12:11. High aerosol particle concentrations of different size distributions were produced by burning candles from 12:45 to 14:40 and incense from 15:15 to 16:10. From 16:10 to 19:00 the air cleaner was put into operation. There is almost no thoron, so measurements were not disturbed by thoron decay products.

After sampling, the Cu plate and the screen were counted for alpha activity with the SAIC Alpha Analyzer and for beta activity with the LLM 500. In fig. 3 the alpha counts in 1 min of the Po-218 peak on the Cu plate and the screen are shown. Pairs of values, measured at nearly the same time are plotted in fig. 4. The numbers indicate corresponding values. There is a good correlation between the activities sampled on the screen and on the Cu plate. This confirms, that there is a fairly constant fraction of Po-218 ions within the unattached fraction for different aerosol and radon gas conditions. It also shows that the collected activity on the Cu plate or the friction charged polystyrene plate is a fair quantity for the unattached fraction under different aerosol and radon gas conditions.

The Po-214 activity was very low and showed no correlation for both methods of sampling. This is explained by the fact that there is almost no unattached and no charged fraction.

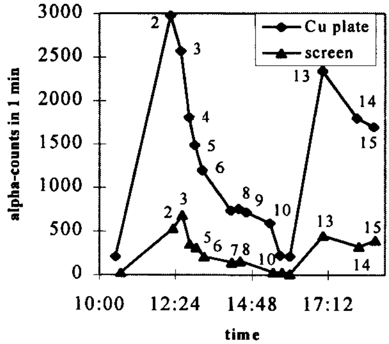


Fig. 3. Alpha counts of Po-218 on the Cu plate (1 min exposition at -16 kV) and on the screen (250 L of air sampled), measured with the SAIC alpha analyzer. Numbers refer to fig. 4

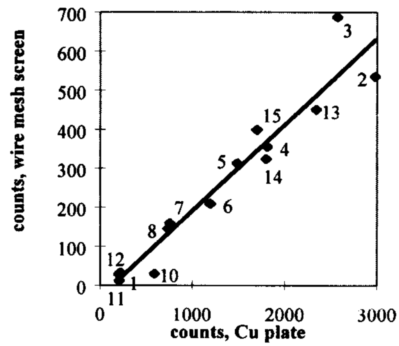


Fig. 4. Alpha counts of fig. 3, plotted as pairs, to show the correlation between the Po-218 activity on the screen and on the Cu-plate. Numbers indicate the points, taken from fig. 3

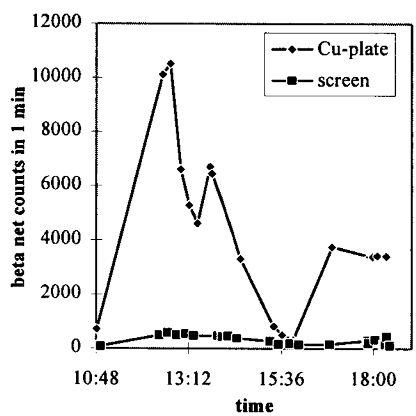


Fig. 5. Beta activity on the Cu-plate and the screen, measured with the LLM 500.

In Fig. 5 total beta counts are shown. There is no obvious correlation between the two series of measurements. The Cu plate shows a wider range of activities for different conditions. This leads to a vague exponential correlation between the two sampling methods. There are several reasons for not having a good correlation. First of all, decaying Po-218 adds to the beta activity. In addition, the correlation between the charged and the unattached fraction of Pb-214 is not known and depends on parameters like humidity and trace gases. Both charged plates sample only the charged unattached fraction, screens sample charged and uncharged, and partly the attached fraction.

CONCLUSIONS

The comparison of the wire mesh screen method with the two types of charged plates - Cu plates and the Philion Plates - gives the following results: A charged plate is a good sampling device for the unattached fraction of Po-218. Despite the very fast neutralization rate of the Po-218 ions, it is possible to collect them by this method, which is simpler and more efficient than wire mesh screens. Because of the sufficiently high voltage the ions are collected before recombination. The determination of the volume, from which Po-218 is sampled, and possible influences of water vapor are still under investigation.

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