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RECOIL-DEPOSITED Po-210 IN RADON DWELLINGS

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Prologue

George Formby (1904-1961) was a popular filmstar, singer and ukulele musician. One of his greatist hit, "When I'm cleaning windows", was first heard in the movie "Keep your seats, please" from 1936. The song was such a great success that the tune was re-recorded in 1937 under the title "Window cleaner No. 2", starting off with these lyrics:

> You've heard about my capers when windows I've to clean
> Now I'd like to tell you of a few more things I've seen
> I've seen miss Thompson in her flat
> take off her shoes, her coat and hat
> I've seen her take off more than that
> When I'm cleaning windows

Partly inspired by Mr Formby, we have also joined the window cleaning business. Unluckily, and despite our use of 96% ethanol, we haven't been able to spot miss Thompson. Instead we have observed the young granddaughter of miss (!) Lea D. Longlived, miss Polona, dressed up in a very thin (50 nm) and tranparent glassy fabric. Our assistant, mr Pulse Ironchamber, is especially sensitive to miss Polona, and he claims to see her radiant beauty in most dwellings.

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ABSTRACT

Short-lived decay products of Rn-222 plate out on all surfaces in a house containing radon gas. Following the subsequent alpha decays of the mother nuclei, the daughter products Pb-214 and Pb-210 are superficially and permanently absorbed. Due to its long half-life (22 y) the activity of absorbed Pb-210 accumulates in the surface. The activity of Pb-210, or its decay products, can thus reflect the past radon daughter and plate-out history of a house over several decades.

Our results and experience from measurements of Po-210 and Rn-222 in 22 dwellings will be presented. In these studies the Po-210 surface activity of one plane glass sheet per dwelling (window panes were not used) has been determined and compared with the period of exposure times the mean radon concentration measured over a two-month period. Considering the large uncertainty in the integrated radon exposure estimate the surface ^{210}Po correlates well (r=0.73) with the accumulated radon exposure. The ^{210}Po activity of the glass samples has been measured non-destructively using an open-flow pulse ionization chamber and this detector has also been successfully applied in field exercises.

Vitreous glass is close to ideal as a substrate. It can be found in all dwellings and migration of the surface activity caused by diffusive transport is thought to be negligible. The results obtained indicate that practical exploitation of the ^{210}Po activity in glass surfaces as an estimator of lung cancer risk is feasible, but the method must be utilized great care in order to minimize the influence from non-dose related factors. The accuracy of the glass-polonium method and conversion factors between surface ^{210}Po and lung cancer risk are still to be assessed.

INTRODUCTION

In the search for the answer to the question which serves as the topic of this conference, we are hampered by the fact that we cannot accurately assess the radon exposure history of a person. When observing the incidence of lung cancer today it is evidently the radon daughter levels several years back that are relevant, not the levels here and now. Even if we do not know the mechanisms or time parameters behind the carcinogenicity of radon it is of interest to investigate if retrospective assessment of radon exposure (RARE) is feasible. ('Radon' in the acronym is used in a broad sense to identify the source term, not the active lung cancer agent. Our primary interest is, of course, to assess the accumulated lung cancer risk, and in doing this air-borne short-lived progenies and their size distribution is much more important than the radon gas itself.)

The basic principles behind RARE, as proposed by Samuelsson, 1988, are presented schematically in Figure 1. All radon atoms present today will, within a few weeks, turn up as ^{210}Pb atoms. Luckily, Mother nature "stows" some of these long-lived atoms away for later measurement. All indoor surfaces accumulate long-lived radon daughter atoms and a more or less permanent record of past radon daughter concentration levels is created in hard materials. Unfortunately, time resolution is not possible, and due to the decay of ^{210}Pb the 'radon daughter memory' is limited in time.

The embedding of long-lived nuclides in the surfaces of a room is a two-step process:

1) surface plate-out of the short-lived progenies
2) alpha decay causing daughter nuclei to recoil into the surface layer.

The range distribution of the recoiling nuclei in hard materials is such that most of them will be safe from normal surface cleaning procedures.

Typically, the embedded surface activity of ^{210}Pb will be very low. A surface exposed to 100 Bq/m^3 (2.7 pCi/ℓ) of radon for ten years exhibits roughly only one decay of ^{210}Pb per m^2 per second (1 Bq m^{-2}). In view of this low surface activity a discussion concerning detection methods is appropriate.

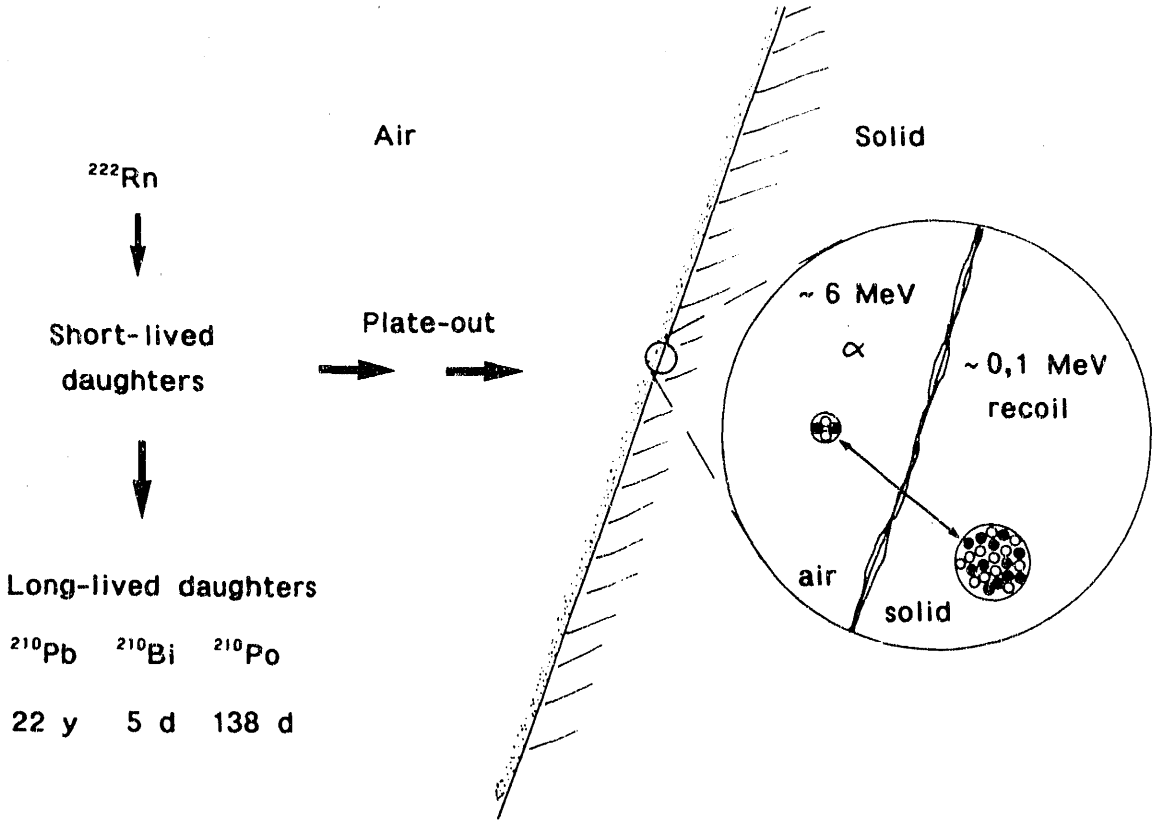

FIGURE 1. Surface plate-out followed by alpha recoils embeds long-lived daughter activity in a surface. The range of the recoiling nuclei in glass is about 50 nm (C. Landsheere, 1989).

DETECTION METHODS

The superficially embedded-long lived radon daughters provide a very thin α/β source and it would be foolish not to try to take advantage of this fact when looking for suitable methods of analysis. Bearing in mind the expected surface activity of about one decay per hour per cm^2, we were discouraged from a direct determination of the β-emitting ^{210}Pb itself, or its β-decaying daughter ^{210}Bi, and turned towards the use of large-area samples. The absence of intensive gamma lines suitable for spectrometry left only alpha detection methods to be explored. Alpha spectrometry is feasile as the grand-daughter of ^{210}Pb, ^{210}Po, decays while emitting a 5.3 MeV alpha particle. Sensitivity is, however, still a problem as the normal solid-state devices of today, with excellent background and energy resolution characteristics, are intended for small-area samples only. This speaks in favor of dusting off the alpha spectrometer of the fifties, the pulse ionization chamber, a detector which can easily accomodate samples with surface areas of several square decimetres, (1 dm^2= 0.1 sq.ft).

Due to their complexity and cost 'electronic' detectors are less suitable for large-scale *in-situ* measurements; extensive epidemiological studies, for instance. For large-scale studies, an autoradiographic technique using track-etch alpha detectors may be feasible, but this would require some sort of energy resolution or discrimination against the alpha background of the substrate material. Depending on origin and manufacturing technique sheets of glass, for instance, may exhibit quite different alpha background count rates, making it difficult to apply any type of total-alpha detector.

In Figure 2 the energy resolution of our 30-litre pulse ionization chamber (PIC) and a 300 mm^2 surface barrier detector (SBD) are compared. The sample is a sheet of glass taken from a window exposed to roughly 6 kBq y m^{-3} during a period of 25 years. The ^{210}Po peak is easily detectable, even with this small SBD, but in order to achieve satisfactory counting statistics the time of mesurement must be exessively long, five days in the illustrated case. In our closed type of PIC spectrometer the sample size is about 200 cm^2 and the net count rate in the ^{210}Po peak is about a hundred times that from the 300 mm^2 SBD.

Our closed PIC accommodates sheets of glass with diameters of less than 180 mm. Larger samples have to be cut down in size in order to be analysed. In field measurements in private dwellings such a destructive procedure is a definite drawback. A special PIC with the ability to analyse sheets of glass non-destructively was therefore constructed. This new PIC is of an open type requiring a continuous supply of counting gas. Due to the low weight of this chamber it is apt to field use.

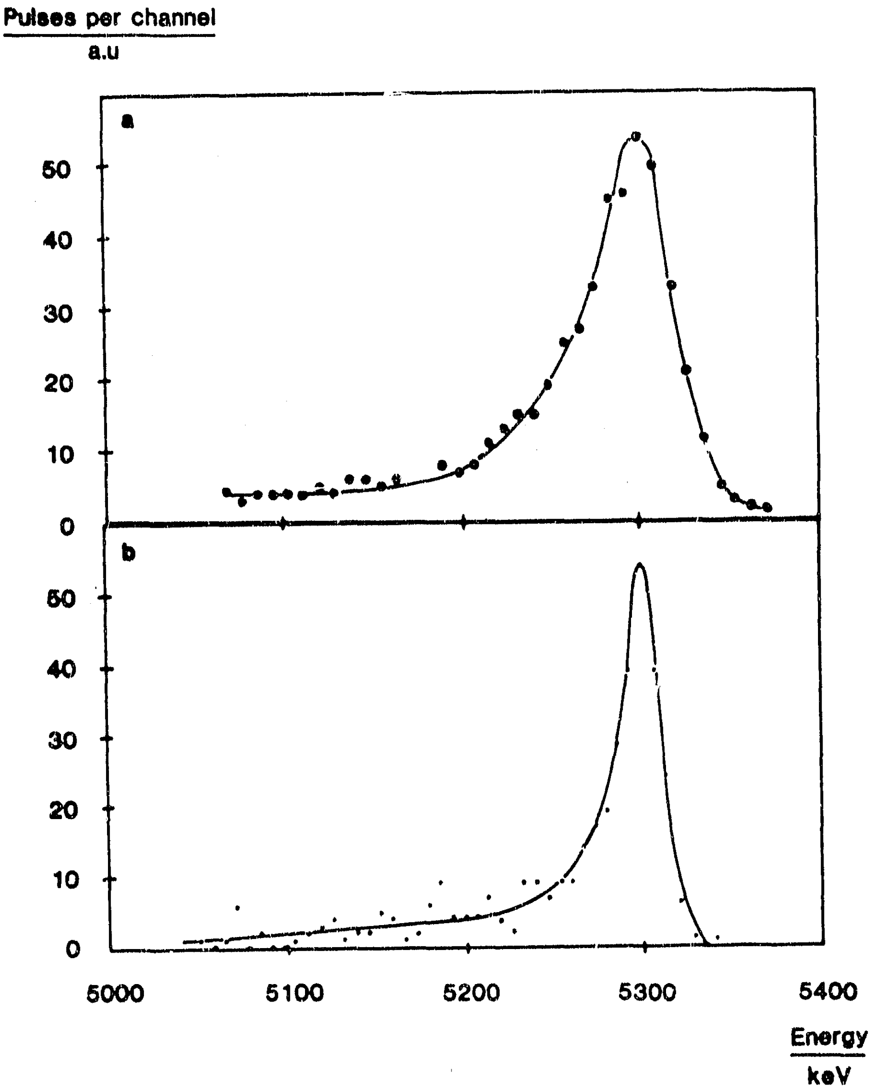

FIGURE 2.
The 5.3 MeV ^{210}Po peak measured in a) a closed pulse ionization chamber and b) a 300 mm^2 surface barrier detector. The sample is a sheet of glass exposed from 1963 to 1988 to roughly 6 kBq y m^{-3}. The net pulse count rate in a) is about 100 times that in b). The FWHM and sample size are 56 keV and 212 cm^2 in a) and 26 keV and collimated to 2.5 cm^2 in b)

MATERIALS AND METHODS

The Akarp Study

With the objective of investigating the relationship between radon concentration levels and long-lived daughter activity of surfaces, a group of detached houses in the village of Akarp near Lund was investigated. All dwellings in the area suffer from an increased indoor radon levels due to the construction material, alum shale lightweight concrete ('blue concrete'). About 60 households were asked by letter if they were willing to help us in a RARE research project and 25 of them answered favourably. The participating home owners were asked to place a piece of glass from a photograph frame or the like at to our disposal for a few days and in return they were offered a radon measurement free of charge. The National Institute of Radiation Protection (SSI) in Stockholm supplied and evaluated the track-etch films (pers. comm. Hans Mellander). The radon cup holding the CR-39 film strip is of the same type as that used by NRPB (Barlett and Bird, 1987).

All dwellings in the study are about 23 years old and only a few of them have undergone major reconstructions since erection. The houses have no basement. The majority of the dwellings are self-draught ventilated and have their own oil-fired heating systems distributing hot water to radiators under the windows.

In each dwelling, one radon cup was placed in the room from which the piece of glass was taken and another cup was exposed in the bedroom. The exposure period covered two months, from March to April 1990. The weather conditions were warmer than normal for southern Sweden, with very few nights below freezing point. The heating systems were in use throughout the major part of the exposure period.

Upon returning the radon cups the home owners answered a questionaire about the age and location of the glass and more general questions about the construction, heating and ventilation of the house. Most of the pieces of glass supplied were analysed non-destructively in the open PIC. Usually, two or three glass samples were analysed in our laboratory per day.

In-house Variations of ^{210}Po

It is obviously of interest to investigate the homogeneity of superficially trapped long-lived daughters in houses. In an effort to check the variability of ^{210}Po activity in window surfaces and at the same time test the open PIC under field conditions, the necessary equipment was installed in a van and parked outside the dwelling to be investigated. From this house (designated No. 10 below) mirrors, some cupboard glass and most of the windows were brought to the van one at a time and analysed for ^{210}Po. House No. 10 is a 'blue concrete' house with a basement and the moderately enhanced radon levels indicates that radon from the ground is of little importance. The house is not situated in Akarp, but a town 300 kilometres north of Lund.

RESULTS AND DISCUSSION

The Akarp Study

Of the participating 25 dwellings, 22 have supplied useful sheets of glass so far. For each of these 22 dwellings the accumulated radon exposure was calculated by multiplying the age (i.e. exposure time in the house) of the glass by radon concentration value given by the single track-etch film exposed in the same room as the glass sheet. The measured ^{210}Po activity of the glass sheets was corrected for decay (old glass samples) or non-equilibrium with ^{210}Pb (young samples) and the corrected values were plotted against the estimated radon exposure (Figure 3).

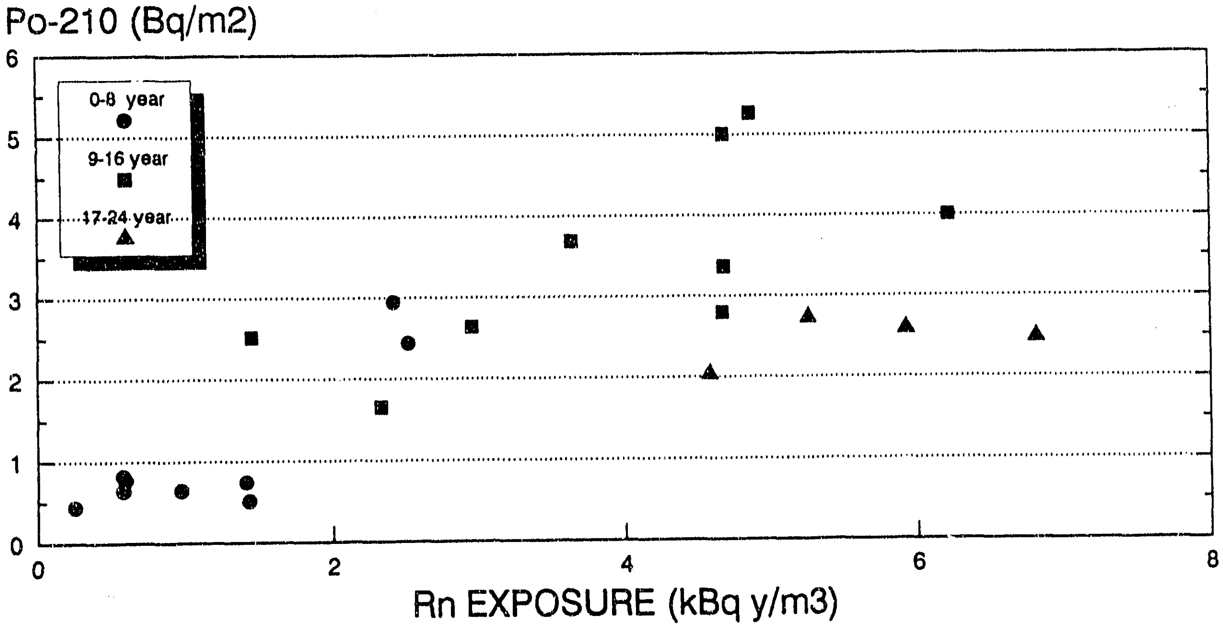

FIGURE 3. The surface activities of all the glass items, divided into three age groups, in the Akarp project, corrected for decay and lack of equilibrium with ^{210}Pb. The radon concentration was measured with the aid of a CR-39 radon film exposed for two months, March-April, 1990, in the room from which the glass sample was taken. The radon exposure displayed is obtained by multiplying the measured radon concentration by the age of the glass.

The uncertainty in the individual CR-39 film values depends on the number of tracks counted. This statistical error is insignificant in Figure 3, corresponding to a relative standard deviation of less than 1% in most cases. The relative standard deviation from counting statistics in analysing ^{210}Po is, in most cases, about 5 %. The assumption that the measured radon level has persisted for the whole exposure period, up to 24 years in Figure 3, can involve a substantial error in some cases. Also, the exposure history of a sheet of glass can be more or less accurate depending on how well the home owner remembers, or can trace by notations, the 'hanging history' of that very piece of glass. Realistically, large errors must be associate with the accumulated radon exposure, and due to the involvement of human authenticity outliers cannot be excluded. Considering the uncertainty in the exposure estimate, based on a single track film value, and the variability in the plate-out phenomenon, the spread in Figure 3 is not surprising.

There is a tendency for old glass samples to show a lower ^{210}Po value than expected from the linear trend. This is an expected feature since the radon levels several years ago were generally lower than today, due to less ambitious energy-saving measures. The coefficient of correlation (r^2) is equal to 0.54 in Figure 3.

In-house Variations of ^{210}Po

The predictions of different air-mixing models indicate a substantial variation in indoor plate-out velocities. Several factors, e.g. air movement patterns, temperature differences, particle size distribution, can have a significant influence on the plate-out rate of the short-lived radon daughters. A relationship between absorbed surface activity and accumulated radon exposure can easily be swamped in local in-house variations of short-lived daughter plate-out rate. In the authentic indoor environment, several of the parameters governing the plate-out rate are difficult, if not impossible, to quantify. One might expect that the inward facing side of window panes would be subject to substantial variations in both plate-out rates and in radon concentration levels. The air-tightness of a window frame, ventilation habits, proximity to ventilators etc. effect the radon level. The air movement patterns and turbulence caused by curtains, radiators, ventilators and window recesses are of importance concerning both the supply of airborne daughters to the window surface and the actual deposition velocity of the decay products. In addition, any temperature difference from room temperature will cause thermophoresis, i.e. a cold surface attracts and a warm surface repels the airborne decay products.

Bearing all these influencing factors in mind, the small spread, less than a factor of 2, in window values from house No. 10 (Figure 4) is somewhat surprising, and it is obvious that several of the short-term, diurnal and seasonal variations of the plate-out rate are levelled out during the long exposure times involved. It should be pointed out that radiators are situated beneath all windows except the terrace door. The two low values ('vent') both belong to windows with special window-catches for ventilation purposes. According to the owners the left window ('vent') in the large bedroom for instance, is kept slightly open throughout most of the year during sleeping hours. The absence of a radiator may explain the low polonium/radon ratio of the terrace door in Figure 4. Excluding the door and the windows used for ventilation the spread in the window values Figure 4 is only ± 10%. This small figure is presumably due to a lucky coincidence. The corresponding variation for another house now under evaluation is preliminarily ± 30%.

Nearly all glass samples analysed have been exposed in house No. 10 since it was built. Exceptions are a mirror exposed for 5 years prior to measurement, a photograph frame (13 years) and the cupboard door in the dining room (vitrine) which is a few years older than the house. At the moment, we have no explanation of the high Po-210/Rn-222 value for the 'young' mirror in the hall. One possible cause has already been mentioned in connection with the results from the Akarp study (Figure 3), the overestimated accumulated radon exposure for old glass samples. When calculating the Po-210/Rn-222 ratio for the cupboard door glass in Figure 4 we have ignored the unknown radon exposure prior to 1966.

The general trend in Figure 4 is that windows, at least those not used for airing, show a higher ^{210}Po value than true indoor glass samples This is probably due to the enhanced mixing of room air close to a window caused by the warm radiator beneath. Since the surface of the window panes is cooler

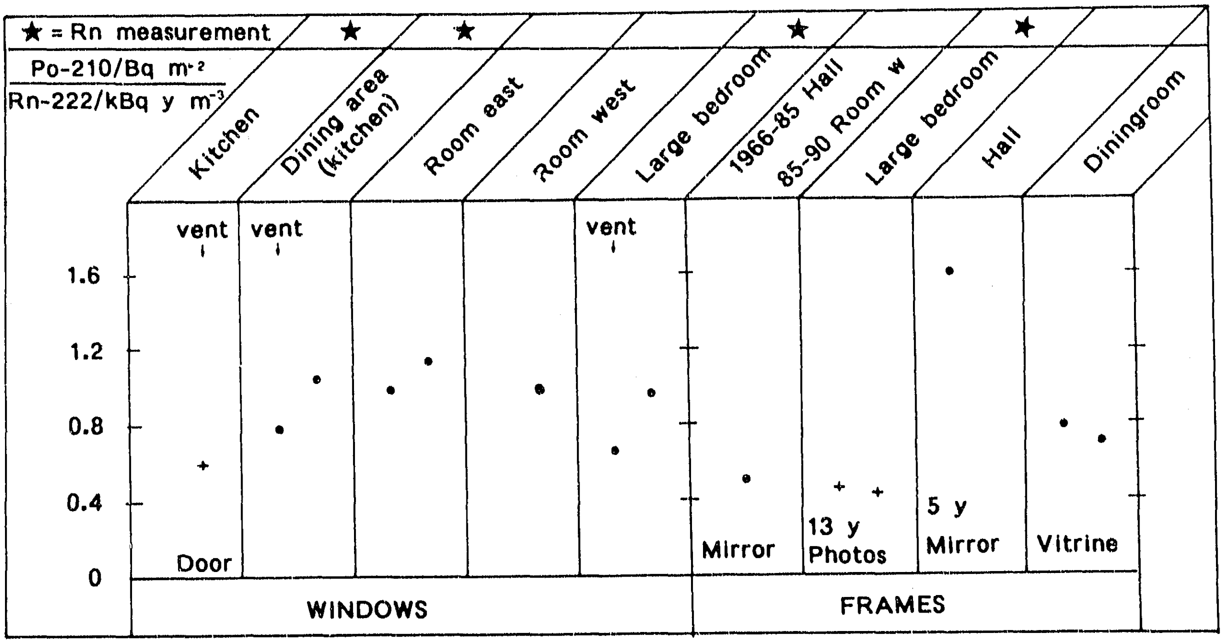

FIGURE 4. The normalized ^{210}Po surface activities of 14 different glass samples taken from dwelling No. 10. Radon measurements were performed in rooms marked with an asterisk. All sheets of glass had been exposed for 22 or 23 years prior to measurement, except when stated otherwise. Samples marked '+' were sent to Lund and analysed in January 1989, all other ^{210}Po measurements were made during a visit to the house in July 1990.

than room temperature during most of the year, thermophoretic effects also contribute to the enhanced surface activity of windows.

If dust and grease are allowed to build up on a glass surface, the alpha recoils will increasingly be trapped in the contaminant and not in the glass. Consequently, the non-removable part of the ^{210}Po activity will be reduced compared with a clean piece of glass. The cleaning practices of the household in house No. 10 are such that mass load and/or cleaning effects are negligible and can not explain the differences in Figure 4. This has been verified by investigating some of the glass items before cleaning. The normal procedure is to clean all samples before measurement with 96% ethanol.

The data presented in Figure 4 are from a self-draught ventilated house situated in a cold temperate region in which the building material constitutes the main radon source. The trends and values in Figure 4 are not necessarily applicable or typical for differently constructed houses in other climates.

CONCLUSIONS

This investigation has proven that field studies of ^{210}Po in glass surfaces can be performed non-destructively using an open pulse ionization chamber. The measured ^{210}Po activity in 22 dwellings is positively correlated (r=0.73) to the estimated radon exposure. As long as we do not know how well the ^{210}Po activity correlates with the true integrated radon concentration, or better, the tumorigenic potential of the indoor atmosphere, it is difficult to judge the feasibility and accuracy of the polonium-glass method as a retrospective radon/lung cancer risk monitor. It is clear that windows situated above radiators normally show enhanced ^{210}Po activity compared with other openly exposed glass surfaces away from radiators. The improved air mixing during the winter season is thought to be responsible for the enhancement, but thermophoretic forces may also play a part.

In our opinion the practical exploitation of ^{210}Po in glass as a RARE monitor must proceed with great care, and the influence of modifying and non-dose related factors must be taken into account. At the present stage of knowledge, it is advisable to analyse more than one glass sample in a dwelling, in order to increase the precision.

Acknowledgements

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