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ANALYSIS OF HAIR FOR POLONIUM-210 α-PARTICLE EMISSIONS*

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ABSTRACT

A pilot study was performed in which hair samples were collected from 14 persons including 7 cigarette smokers (CS), 4 male, 3 female and 7 nonsmokers (NS), 4 male, 3 female. These subjects had been environmentally exposed to different concentrations of bedroom radon-222 (CS, 0.61 ± 0.8 kBq/m^3; NS, 1.1 ± 1.6 kBq/m^3). The hair samples were washed, labeled with a trace dose of Po-208/Po-209 (6 mBq), digested, and the polonium isotopes (208, 209 and 210) were auto-plated onto polished silver disks. Polonium-210 activity was detected in all hair samples (CS, 3.9 ± 3.7 mBq/g; NS, 4.1 ± 4.7 mBq/g). Experiments indicated that Polonium-210 entered the hair through an internal pathway.

INTRODUCTION

Scalp hair levels of a number of trace metals are indicative of body tissue levels in humans (Smith 1987). These elements include: cadmium (Petering et al. 1973), chromium (Hambidge 1972), selenium (Keshan Disease Research Group 1979), strontium (Weyne et al. 1981) and magnesium, copper, strontium and barium (Smith 1987). The primary objectives of this investigation were to develop a reproducible method of measuring polonium-210 (Po-210) [a radioactive decay product of radon-222 (Rn-222)] activity in hair and to test the hypothesis that scalp hair Po-210 activity is dependent upon (a) bedroom radon exposure and/or (b) Po-210 inhalation from smoking. A suitable method of digesting hair, adding a radioactive trace dose of Po-208/209 ("spiking") and auto-plating the three isotopes of polonium (208, 209, and 210) onto polished silver disks was perfected. A secondary objective was to determine whether the Po-210 hair activity was a result of prior environmental exposure of the subject and not a result of external contamination acquired during sample digestion and subsequent auto-plating.

EXPERIMENTAL DESIGN

Hair Samples and Population

Twenty three hair samples of approximately one gram mass each were obtained from a local barber shop and 13 similar hair samples were obtained from persons who had Rn-222 (mean concentration ± standard deviation, m ± S.D., 281 ± 106 Bq/L) in their well water. These hair samples were used to develop a working procedure for washing, digesting and subsequently auto-plating 6 mBq trace amounts of Po-208/209 ("spikes") and the Po-210 in the hair samples onto polished silver disks†.

Measurements of bedroom radon concentrations with charcoal canisters‡ were performed in exchange for one gram hair samples. Both cigarette smokers (CS) and nonsmokers (NS) participated. A one-year residence was required whenever bedroom radon-222 concentrations exceeded 3 kBq/m^3. Each subject was requested to expose a charcoal canister in his/her bedroom prior to obtaining a hair cut. The subject was required to return a one gram hair sample with an exposed canister. Two subjects were asked to return up to 5 g hair samples approximately 12 cm in length.

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† 1.000 OD × 0.005 TK, 99.93/99.95 CC FINE AG, Lucas-Milhaupat, Inc., Cudahy, WI 53110.

‡ Radon canister RC-41, Ink Filtering Systems, Cumming, GA 30130.

Washing

Hair samples were washed in a clarifying gel[§] to remove build-ups of chlorine, medication, waxes, metals and other impurities and rinsed in distilled water. A beaker containing the sample and solution were agitated in the water bath of an ultrasonic cleaner[‖] in each of the above steps. The samples were dried for approximately 4 h, rinsed in acetone[¶], dried for 2 h and weighed on an electronic balance[#].

Digesting

The clean hair samples were digested in 10 ml concentrated nitric acid[**] after approximately 6 mBq Po-208/Po-209[††] had been added as a reference "spike". This solution was heated at 85° C until dry (approximately 0.5 h), and 3 ml concentrated nitric acid and 10 ml of 30% hydrogen peroxide[‡‡] were added. The hydrogen peroxide was added to aid in oxidation of fat molecules. The solution was then heated until it was dry (approximately 0.5 h). This procedure of addition of acid and hydrogen peroxide was repeated up to three times until there was no visible evidence of fat. Five ml of 12 molar hydrochloric acid[§§] and distilled water (5 ml) were added to the dry sample and the resulting solution was heated at 85° C until dry (approximately 0.5 h). This latter process removed nitrates which interfere with the Po plating[‖ ‖].

Disk Polishing and Plating

Distilled water (120 ml) and approximately 10 mg ascorbic acid powder were added to the dry mixture described above. A solution with pH<2 was required for plating the polonium[¶¶]. A silver disk was polished by applying approximately 100 mg silver polish[##] with a cotton swab[***] and rubbing the cotton swab on the disk surface in a circular motion for approximately 120 s. Excess polish was removed by washing the silver disk with distilled water. The silver disk was painted[†††] on one surface and was placed in a teflon holder containing a magnetic stirring bar[‡‡‡]. The sample holder was placed in the "spiked" plating solution and the polonium isotopes (208, 209 and 210) were plated. The beaker containing the plating solution and the teflon holder with a polished silver disk were placed on a hot plate with a built-in stirrer (stirring hot plate[§§§]). Polonium plating took place over a 6-8 hour period at a solution temperature of approximately 85° C and a stirring frequency of approximately 2.5 Hz[‖ ‖ ‖].

Control Samples

Blank samples were prepared simultaneously in conjunction with hair samples in three trials so as to provide a measure of background contamination.

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§ Aveda Corporation, Minneapolis, MN 55413.

‖ Model 450, E/MC RAI Research Co. Hauppauge, NY 11787.

¶ Klean-Strip, W.M. Barr, Inc., Memphis, TN 38101-1879.

Model GT210, OHAUS Corporation, Florham Park, NJ 07932.

** Reagent A.C.S., Fischer Scientific, Fair Lawn, NJ 07410.

†† Isotope Products Laboratories, Burbank, CA 91504.

‡‡ J.T. Baker Chemical Co., Phillipsburg, NJ 08865.

§§ Reagent A.C.S., Fischer Scientific, Fair Lawn, NJ 07410.

‖ ‖ Personal communication (1992), K. Oralandin, Argonne National Laboratory, Argonne, IL.

¶¶ Willner Chemist, Inc. New York, NY 10016).

Wright's Silver Cream, J. A. Wright & Co., Keene, NH 03431.

*** Q-tips, Chesebrough-Pond's USA Co., Greenwich, CT 06830.

††† Krylon semi-flat black, The Sherman-Williams Company, Solon, OH 44139.

‡‡‡ Personal communication (1991), P. O. Jackson, Battelle Pacific Northwest Laboratory, Richland, WA 99352.

§§§ Equatherm model 267-914, Curtin Matheson Scientific, Inc., Houston, TX 77251.

‖ ‖ ‖ PNL ADMINISTRATIVE PROCEDURES FOR GRD (1985), RSD-51-DIS 8, Battelle Pacific Northwest Laboratories, Richland, WA 99352.

A control hair sample of 8 g was obtained from one individual whose bedroom radon concentration was approximately 0.130 kBq/m^3. The 8 g hair sample was used to estimate the contribution of external Rn-222 exposure to measured hair Po-210 activity and to assess the effectiveness of the washing procedure. Four grams of this 8 g hair sample were exposed to a Rn-222 atmosphere of approximately 0.95 ± 0.51 MBq/m^3 for 7-10 d in a "Radium Ore Revigator" (Landa et al. 1988). The exposed hair was divided into four 1 g samples. One of the exposed hair samples was digested, "spiked" and the polonium isotopes (208, 209, & 210) were plated onto a silver disk. The remaining three samples were treated as follows: one was washed in a clarifying gel[§], the second was washed in an acetone solution[¶], and the third was washed in a 2N HCl solution. Finally, all three samples were "spiked", digested and the polonium isotopes were plated onto silver disks. Polonium-210 activities of these hair samples were compared to activities obtained from the remaining four 1 gram hair samples which had not been exposed to a controlled Rn-222-laden atmosphere but had been "spiked", digested and plated in an identical manner. This experiment was repeated twice for a total of three trials.

Sample Analysis

The plated silver disk was cooled and transferred to the counting chamber of an ion-implanted-silicon detector alpha particle spectrometer[¶¶] where it was counted for 24/48 h. Counting chamber backgrounds (24/48 h) were acquired weekly.

RESULTS

Control Samples

No net Po-210 activity above background was found in any of the three blank control samples which had been analyzed throughout the study. For this reason, it was deemed unnecessary to have analyzed a blank control sample with every hair sample digestion.

The results from the controlled Rn-222 atmosphere exposure experiment are shown in Table 1. A one way analysis of variance (ANOVA) (Kleinbaun and Kupper 1978) showed that there were significant differences among the measured Po-210 hair activities of the Rn-222 exposed and washed hair samples (E-W), the Rn-222 exposed and not washed (E-NW) hair samples and the hair samples not intentionally exposed to Rn-222 but washed (NE-W). The mean ± standard deviations for these Po-210 activities were (E-W, 1.4 ± 0.5 mBq/g; E-NW, 2.9 ± 0.5 mBq/g; NE-W, 1.5 ± 0.4 mBq/g). The Tukey contrast (Kleinbaun and Kupper 1978) indicated that these differences were significant at the $p<0.05$ level. The results of one specific control hair sample experiment are shown in Fig. 1.

Hair sample Po-210 activities in a 16 year old male cigarette smoker (CS) who had smoked cigarettes for 4 years (4 y @ 20 cigarette/d) as well as those in hair from a former 27 year old male cigarette smoker (FS) (6 y @ 20 cigarette/d) were dependent upon distance from the scalp as measured along the hair shaft. The hair Po-210 activities were greater in 5 cm hair segments adjacent to the scalp than in 5 cm segments including the far end of the hair segment. Respective Po-210 activities expressed in mBq/g were 12 and 2.3 (CS) and 3.5 and 2.3 (FS).

Hair Samples

A total of 108 hair samples were digested and plated. Only 71 of these hair samples were analyzed for α-particle emissions. However, complete data were available for only 55 of these hair samples of which 26 were used in duplicate analyses. Thirty-six hair samples were used to develop the procedures used. A baseline value of (1.4 mBq/g) was found in hair from persons with low bedroom radon concentrations (0.060 kBq/m^3), greater activities (14 mBq/g) were found in hair from persons with higher bedroom radon concentrations (3.4 kBq/m^3). Non-smoking persons with higher bedroom radon concentrations had higher hair Po-210 activities than those with relatively low bedroom radon concentrations. A contrast between hair Po-210 activity from a 76 year old male (NS) with a bedroom Rn-222 concentration of 3.4 kBq/m^3 and another 28 year old male (NS) with a bedroom radon concentration of 0.6 kBq/m^3 is shown in Fig. 2. In general hair samples from CS or FS contained higher Po-210 activities than hair from NS persons. A representative comparison of hair Po-210 activity between hair from a 16

¶¶ ULTRA CAM detector, ORTEC 576A Alpha Spectrometer, Ortec, EG&G Oak Ridge, TN 37831-0895.

year old male (CS) and hair from a 62 year old male (NS) with bedroom Rn-222 concentrations of 0.133 and 0.06 kBq/m^3, respectively, is shown in Fig. 3.

Linear regression analyses of hair Po-210 activity as a function bedroom Rn-222 concentration yielded R squared values of approximately 0.78 for the NS group and 0.43 for the CS group. Measured hair Po-210 activities and bedroom Rn-222 concentrations for the 14 subjects studied are shown in Table 2. A two way ANOVA to test if hair Po-210 activity correlated with ambient bedroom Rn-222 concentration was significant. Spearman correlation coefficients (Kleinbaum and Kupper 1978) of r=0.605 and p=0.0218 were found when the variables were ranked.

DISCUSSION

Control experiments demonstrated that Po-210 hair activity was not a result of external contamination from either exposure of hair to Rn-222 in the bedroom atmosphere or from the "spiking", digesting, or plating procedures. Thus, hair Po-210 activity may reflect body tissue content as it is true of some other trace metals (Smith 1987). The lack of net Po-210 activity in the three blank control samples indicated that no net detectable Po-210 activity was introduced into the sample during the "spiking" and the plating processes.

A weak correlation between bedroom Rn-222 concentration and hair Po-210 activity was found ($R^2 \approx 0.44$, for the 7 CS and the 7 NS subjects considered). This was in agreement with a previous finding by (Lykken and Alkhatib 1993) and it is in support of a finding that Pb-210 hair content correlated with bedroom Rn-222 concentration ($R^2 \approx 0.98$) (Lykken et al. 1991). However, complete histories of the subjects were not considered and subject lifestyle, including time spent in the bedroom, smoking history, and occupational as well as environmental exposure were not considered. The weak correlation between bedroom Rn-222 concentration and hair Po-210 activity and the relatively higher hair Po-210 activities in the CS group may indicate that cigarettes were a source of Po-210 (Martell 1974; Martell 1975).

It remains to be shown that hair Po-210 activity is indicative of tissue Po-210 activity in humans.

REFERENCES

Hambridge, K. M. M. Hair chromium concentration: effect of sample washing and external environment. Am. J. Clin. Nutr. 25:384-389; 1972.

Keshan Disease Research Group of the Chinese Academy of Medical Sciences, Beijing. Relation of selenium deficiency to the occurrence of Keshan disease. Chinese Med. J. 92(7):477-482; 1979.

Kleinbaun, D. G.; Kupper L. L. Applied regression analysis and other multi-variable methods. Scituate, MA: Duxbury Press; 268-271; 1978.

Landa, E. R.; Miller, C. L.; Brich, R. F. Radioactive and nonradioactive solutes in drinking water from rn-charging devices. Health Phys. 54(1):99-106; 1988.

Lykken, G. I.; Bushaw B. A.; Frey W. H. Analysis of biological samples for lead-210 by isotopically selective RIMS using double-resonance excitation with single-frequency cw laser. (Post-deadline paper). 1991 Meeting of the Southeastern Section of the American Physical Society, Duke U, Durham, N.C.; 1991.

Lykken G.I.; Alkhatib H. A. Analysis of hair for polonium-210 alpha-particle emissions. FASEB J. 7(4):A676; 1993.

Martell, E. A. Radioactivity of tobacco trichomes and insoluble cigarette smoke particles. Nature 249:215-217; 1974.Martell, E. A. Tobacco radioactivity and cancer in smokers. Amer. Scientist 63:404-412; 1975.

Martell, E. A. Polonium-210 alpha particle irradiation of the sperm of cigarette smokers. Unpublished manuscript: 1-11; 1987.

Petering, H. G.; Yeager, D. W.; Witherup. S. O. Trace metal content in hair. Arch. Env. Health 27(11):177-180; 1973.

Smith, B. L. Cardiovascular risk as related to an element pattern in hair. Trace Elem. Med. 4(8):131-133; 1987.

Weyne, J.; DeHemptinne; Leusen, I. Effects of strontium ions on contractibility of isolated heart preparations from Handbook of Stable Strontium. Skoryna (ed) Plenum Press, N.Y.; 1981.

Table 1. Effect of Rn-222 exposure on measured Po-210 hair activity.

Sample ID	NE-W[a]	E-W[b]	E-NW[c]
Trial 1a Exposure Rn-222 Concentration (kBq/m^3)	0.13	1200	1200
Trial 1a Po-210 Activity (mBq/g)	1.2	1.0	3.4
Trial 1b Exposure Rn-222 Concentration (kBq/m^3)	0.13	1300	1300
Trail 1b Po-210 Activity (mBq/g)	1.9	2.0	2.6
Trail 1c Exposure Rn-222 Concentration (kBq/m^3)	0.13	360	360
Trial 1c Po-210 Activity (mBq/g)	1.3	1.2	2.6

[a]Hair sample had not been exposed but washed.

[b]Hair sample had been exposed and washed.

[c]Hair sample had been exposed but not washed.

Table 2. Bedroom radon-222 concentration and hair polonium-210 activity in cigarette smokers and non- smokers.

Subject (sex, ID)	Age (y)	Rn-222 (kBq/m3)	Po-210 (mBq/g)	Rn-222/Po-210
Women				
CS-A	53	0.13	1.6	0.08
CS-B	36	1.80	4.0	0.45
CS-C	40	0.13	1.2	0.11
Men				
CS-D	41	1.80	3.0	0.60
CS-E	33	0.13	2.0	0.06
CS-F	28	0.13	3.5	0.04
CS-G	16	0.13	12.0	0.01
Women				
NS-I	73	3.40	4.6	0.74
NS-J	52	0.06	0.8	0.08
NS-K	55	0.06	1.2	0.05
Men				
NS-L	76	3.40	14.0	0.24
NS-M	28	0.56	1.4	0.40
NS-N	63	0.06	1.8	0.03
NS-O	54	0.13	5.2	0.02

Fig. 1 Respective 24 h Po-210 α-particle counts from control hair samples either intentionally exposed to a Rn-222 concentration of 0.4 MBq/m^3 or not exposed (see text). Exposed and not washed, E-NW, 2.6 mBq/g; exposed and washed, E-W, 1.2 mBq/g; not exposed but washed, NE-W, 1.3 mBq/g. Numbers above peaks correspond to polonium isotopes.

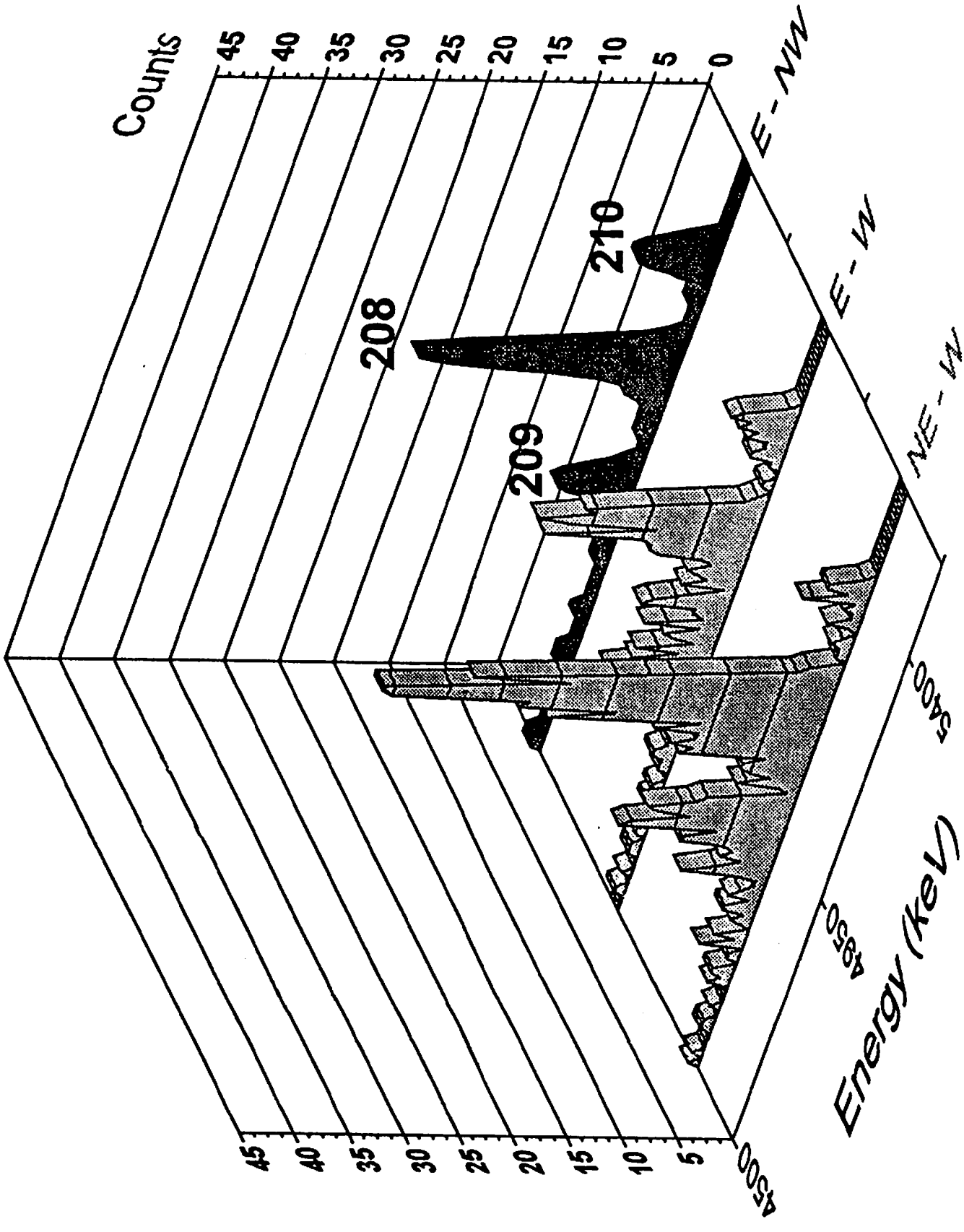

Fig. 2 Contrast between hair sample 24 h Po-210 α-particle counts from a NS male (hfk) with a bedroom Rn-222 concentration of 0.6 kBq/m^3 and similar counts from a NS male (hkj) with a bedroom Rn-222 concentration of 3.4 kBq/m^3 (see text). Numbers above peaks correspond to polonium isotopes.

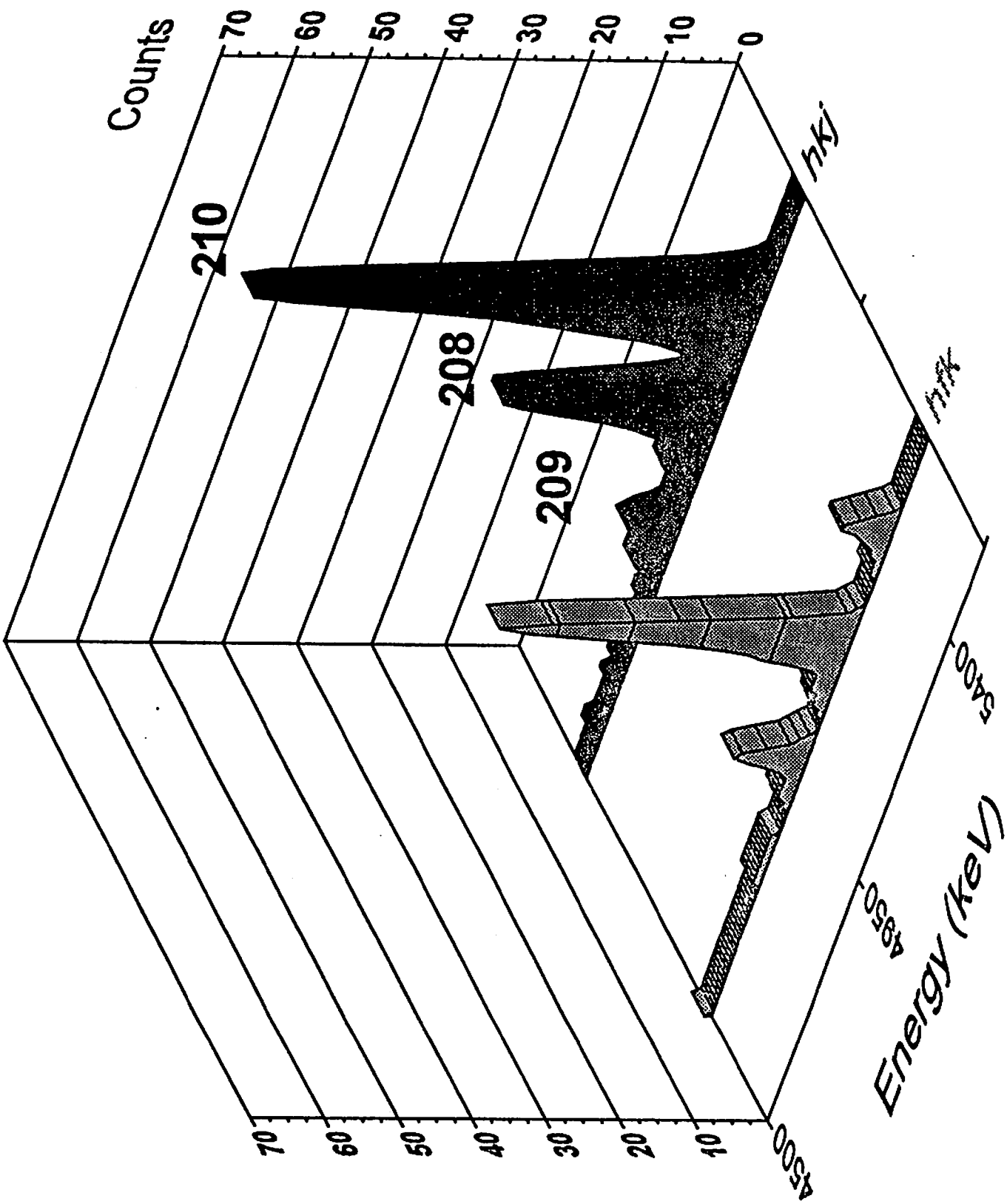

Fig. 3 Comparison between hair sample Po-210 α-particle counts from a CS male subject (hct) with a bedroom concentration of 0.13 kBq/m^3 and similar counts from a NS male subject (hmm) with a bedroom Rn-222 concentration of 0.06 kBq/m^3 (see text). Numbers above peaks correspond to polonium isotopes.