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NATIONAL INTER-CALIBRATION EXERCISE ON POLYCARBONATE TRACK-ETCH DETECTORS

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Abstract

Since the results from previous national intercalibration exercises on polycarbonate track-etch detectors showed a spreading of more than 20 %, a new intercalibration exercise was organized and performed at SCK•CEN. The results are reported and compared with those previously obtained. From the data it turned out, that the conversion factor between the track density and the radon exposure is not constant. Indeed, the sensitivity increases with increasing values of the average track-diameter.

1. Introduction

In 1994 three intercalibration exercises on polycarbonate track-etch detectors have been organized by the Instituut voor Hygiëne en Epidemiologie (IHE), the Institut Supérieur Industriel de Bruxelles (ISIB) and the University of Gent (RUG). The results of the intercomparison organized by the RUG are given in Tab. 1 as an example. The intercomparison shows a scatter around the obtained mean radon exposure of more than 10 %. At SCK•CEN, the exposure (E) is calculated from the number of tracks (y) according to the equation from ref. [1] :

$$E = -\ln(1 - y/a)/b \quad , \tag{1}$$

with $a = 7000$ tracks/cm^2 and $b = 7.8 \times 10^{-5}$ m^3/(kBq h), if $y < 2700$ tracks/cm^2. The initial slope (ab) is 0.546 tracks/cm^2/(kBq/m^3 h) and is compatible to the earlier value of 0.55 tracks/cm^2/(kBq/m^3 h), which was confirmed within 5 % by previous international intercalibration exercises [2,3]. Therefore, the deviation of the results obtained by the SCK•CEN group from the mean value, although not the largest as it is shown in Tab. 1, is unexpectedly large. As a result, it was decided to invite the participants to attend a new intercalibration exercise organised at SCK•CEN. During this exercise great interest was attached in particular on additional radon measurements as a reference to cross-check the whole procedure.

Table 1: Results from the intercalibration exercise on polycarbonate track-etch detectors organized by the University of Gent in summer 1994. The scatter of the data reaches 27 %.

	SCK•CEN	RUG	ISIB	IHE
exposure (kBq/m^3 h)	746	640	710	565
$\Delta E/\overline{E}$ (%)	12	-4	7	-15

In the next chapter the experimental conditions are described. Then, the results obtained by the SCK•CEN group and reported from the other participants are summarized. In the fourth chapter all results are compared and discussed and in the last chapter the consequences for the SCK•CEN calibration factor are introduced.

2. Experimental setup

The track-etch detectors were exposed to radon in a chamber with a volume of about 3.6 m^3. The air was circulated continuously through a radon source. The radon activity concentration was monitored with a continuous radon monitor [4,5] integrated over periods of 2 h. Figure 1 shows, that the intercalibration exercise was performed under well defined as well as stable conditions. The slightly decreasing activity concentration between about 120 h and 200 h correspond to the loading and removing procedure of the detectors. Since the variation of the radon activity concentration was gradually and very slow compared to the monitoring interval, it was possible to determine the radon exposure to the detectors with an accuracy within 1.8 %.

In order to have additional and independent information on the radon exposure air samples were taken before and after the exposure of the detectors as reference points. At later stages additional samples were taken as controls. These air samples were then analysed at the SCK•CEN low-level radioactivity measurement group [6] by means of adsorption on activated charcoal and transfer to a Lucas-cell. From totally five sampling series the second one had to be rejected, since the obviously too low value obtained for the radon activity concentration indicated bad vacuum conditions inside the sampling volume. In Fig. 2 the reported average radon activity concentration of each sampling series is shown (upper part) together with its deviation with respect to the values obtained with the continuous radon monitor (lower part). The grab-sample measurements are less than or equal to the values obtained with the continuous monitor. Since the maximum deviation is less then 9 %, this may be seen as a support for the validity of the furtheron reported radon exposures.

3. Experimental results

In total four different exposures have been applied to detectors of the SCK•CEN with

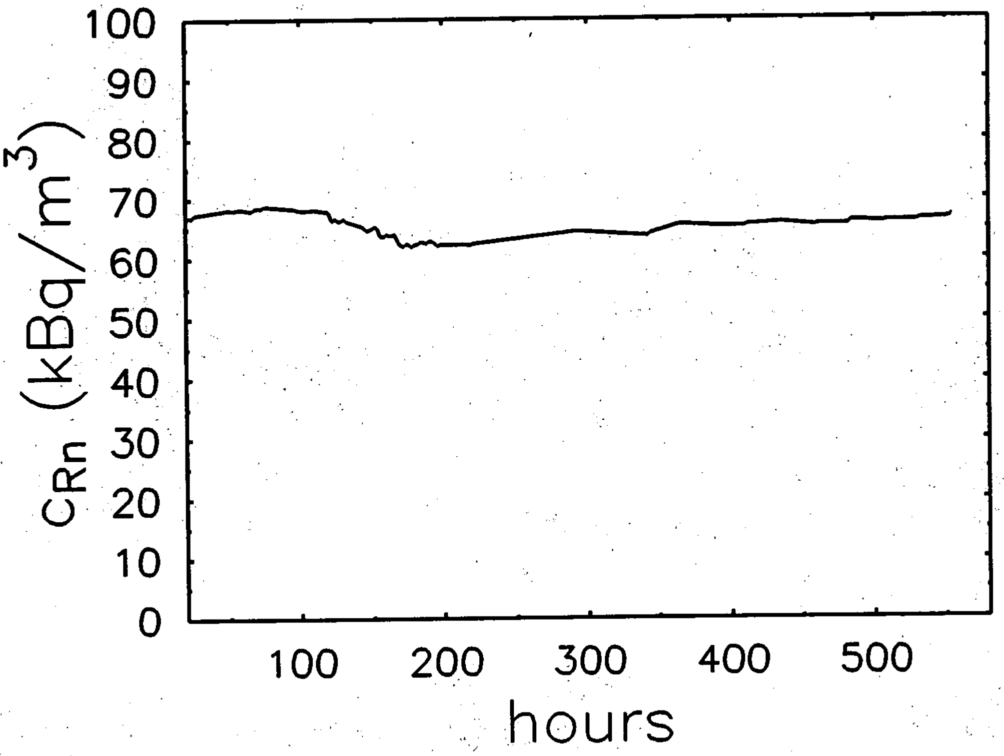


Fig. 1: Radon activity concentration c_{Rn} as a function of time. The slightly decreasing of c_{Rn} between 120 h and 200 h correspond to the loading and removing procedure of the detectors.

exposures ranging from 450 kBq/m^3 h to 1.9 MBq/m^3 h. At two intermediate exposures the detectors from the other participants were added.

3.1 Results of SCK•CEN

After exposure the polycarbonate foils were pre-etched for 30 min in a mixture of ethyl alcohol and a 6N KOH solution with a volume ratio 1:4 in order to reduce background tracks. Then, the foils were electrochemically etched in the same solution for 3 h at 25°C applying an effective voltage of 800 V at 2 kHz [1].
From the central region of the detector an area of 1.0 cm^2 was magnified by a factor of 23 with a microfiche reader and printed on paper. The tracks were then counted manually.

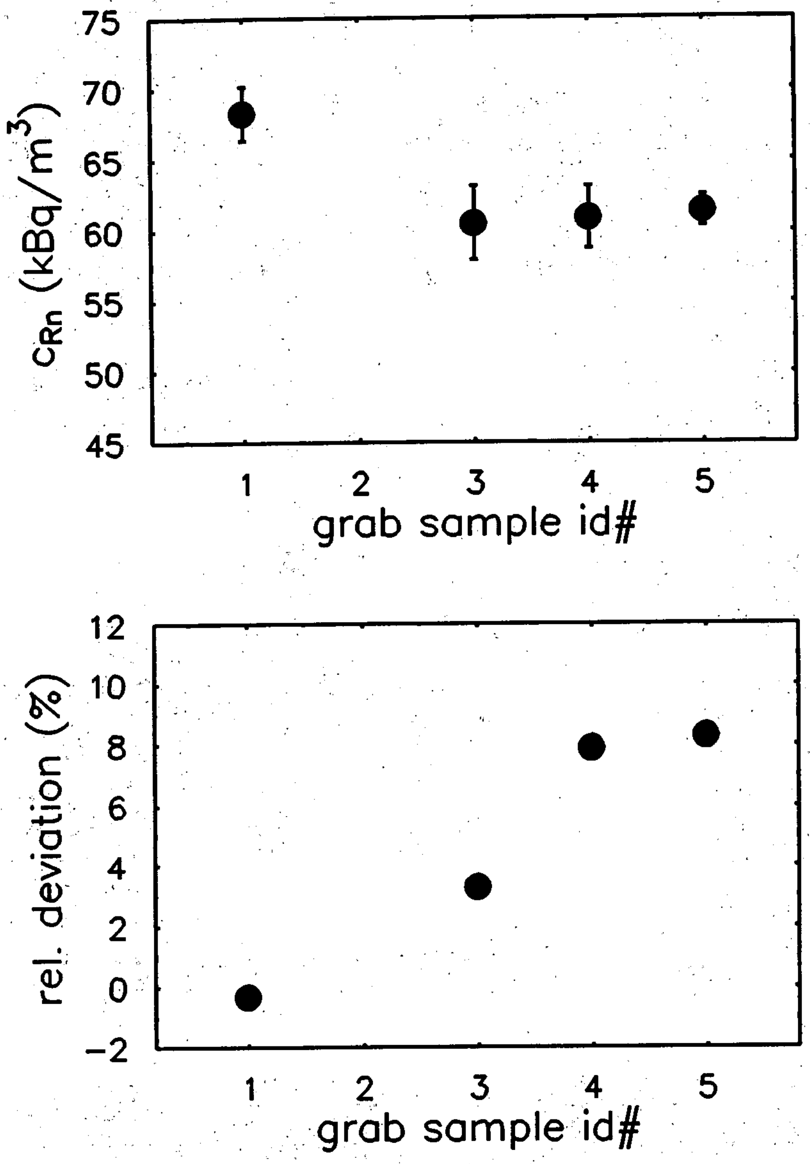


Fig. 2: Radon activity concentration c_{Rn} measured by means of grab-sampling (upper part). In the lower part the deviation is shown with respect to the values obtained with the continuous radon monitor.

The average background track density was determined from 12 unexposed detectors to be 4.4 ± 2.7 tracks/cm^2. In Fig. 3 the obtained net track densities of each detector and each exposure are shown. The upper part shows the track densities after medium exposure, i. e. run (1) and (2), and the lower part shows the track densities after low (•) and high (■) exposure, i. e. run (3) and (4), respectively. The uncertainties due to counting statistics are not indicated in the figures. The average track densities and the corresponding exposures using the conversion factor from ref. [1] are the following :

number of run	1	2	3	4
mean track density (tracks/cm^2)	565	588	293	1186
exposure (kBq/m^3 h) [1]	1079	1125	548	2380
exposure (kBq/m^3 h) (contin. radon mon.)	853.1	882.8	456.0	1898.9
rel. deviation (%)	26.5	27.4	20.2	25.3

However, if we compare these results with the data we got from the continuous radon monitor as mentioned in the previous chapter, the exposures obtained with the polycarbonate detectors are higher by more than 20 %. In Fig. 4 the track density is shown as a function of the radon exposure E from the continuous radon monitor. The ratio between both sets of exposures α is determined by means of a least square analysis

$$\frac{\partial}{\partial\alpha}\sum_{i=1}^{4}((\alpha E_i^1 - E_i^c)/E_i^c)^2 = 0 \quad , \tag{2}$$

with E^1, E^c denoting the exposure calculated from eq. (1) and obtained with the continuous radon monitor, respectively. From this minimization α turned out to be 0.801, which leads to an initial slope of 0.682 tracks/cm^2/(kBq/m^3 h).
Before the existing discrepancy will further be discussed, the results from the other participants are presented next.

3.1 Results reported from RUG, ISIB, IHE

The detectors from the other participants were exposed simultaneously with those from

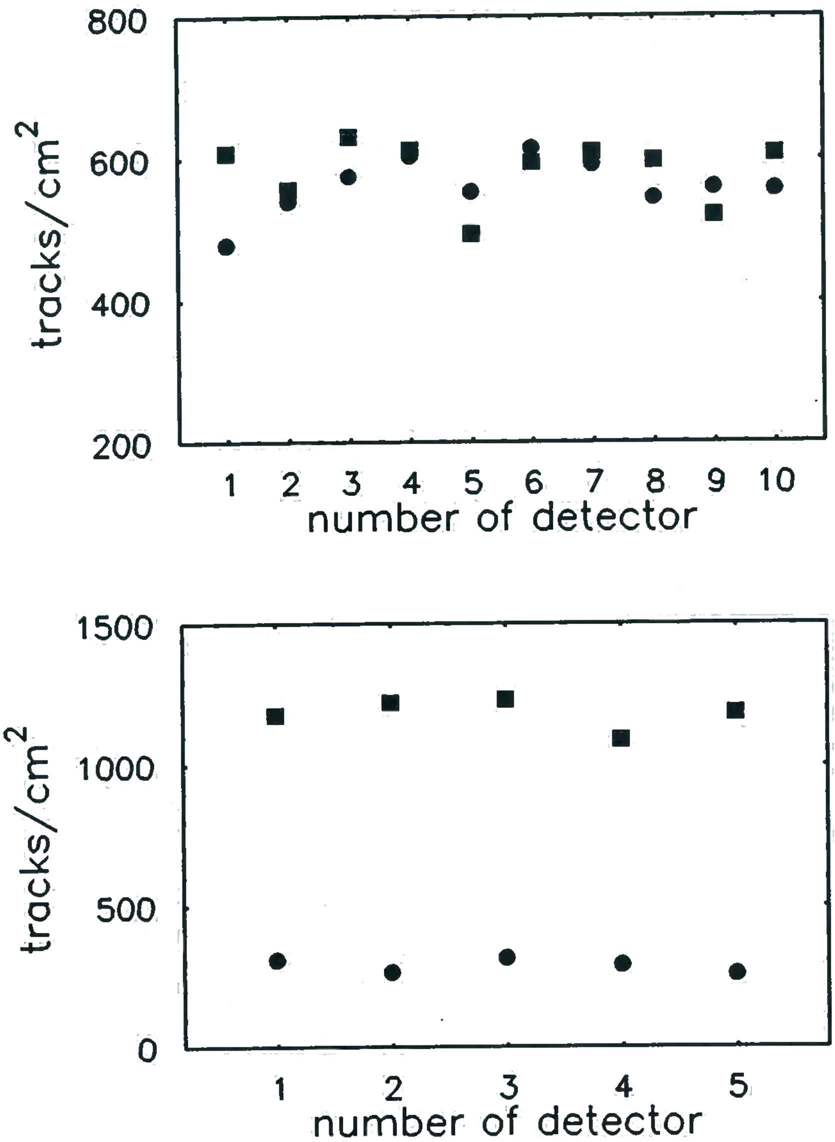


Fig. 3: Net track density obtained by manually counting after a magnification of 23. In the upper part the track densities correspond to medium radon exposures, run (1) and run (2). In the lower part data indicated by a • or a ■ correspond to a low or high radon exposure, i. e. run (3) and run (4), respectively. Uncertainties due to counting statistics are not indicated.

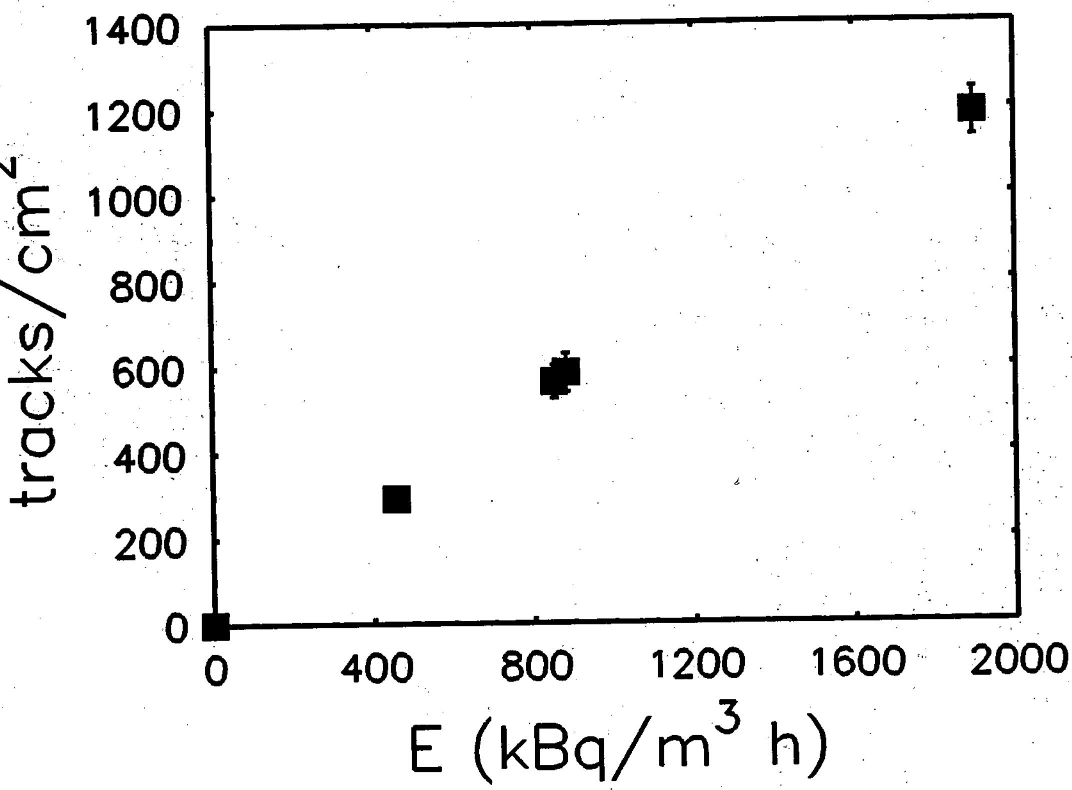


Fig. 4: Average net track density obtained at SCK•CEN as a function of the radon exposure E measured with the continuous radon monitor.

SCK•CEN at medium exposures corresponding to run (1) and (2). The University of Gent (RUG) and the Instituut voor Hygiëne en Epidemiologie (IHE) sent us 10 detectors each, which were equally distributed on both runs. From the Institut Supérieur Industriel de Bruxelles (ISIB) 20 detectors were obtained, 10 of them underwent a thermal treatment to decrease the background. They were exposed during run (2). In Fig. 5 the reported track densities are shown. In the upper part the data from ISIB are shown and in the lower part those from IHE. From the RUG only average track densities were reported. The average track densities and the corresponding exposures for both runs are given in Tab. 2.

4. Discussion

The intercomparison of the reported radon exposures from all participants again show a spreading of more than 20 %. This is although the track densities of the same group

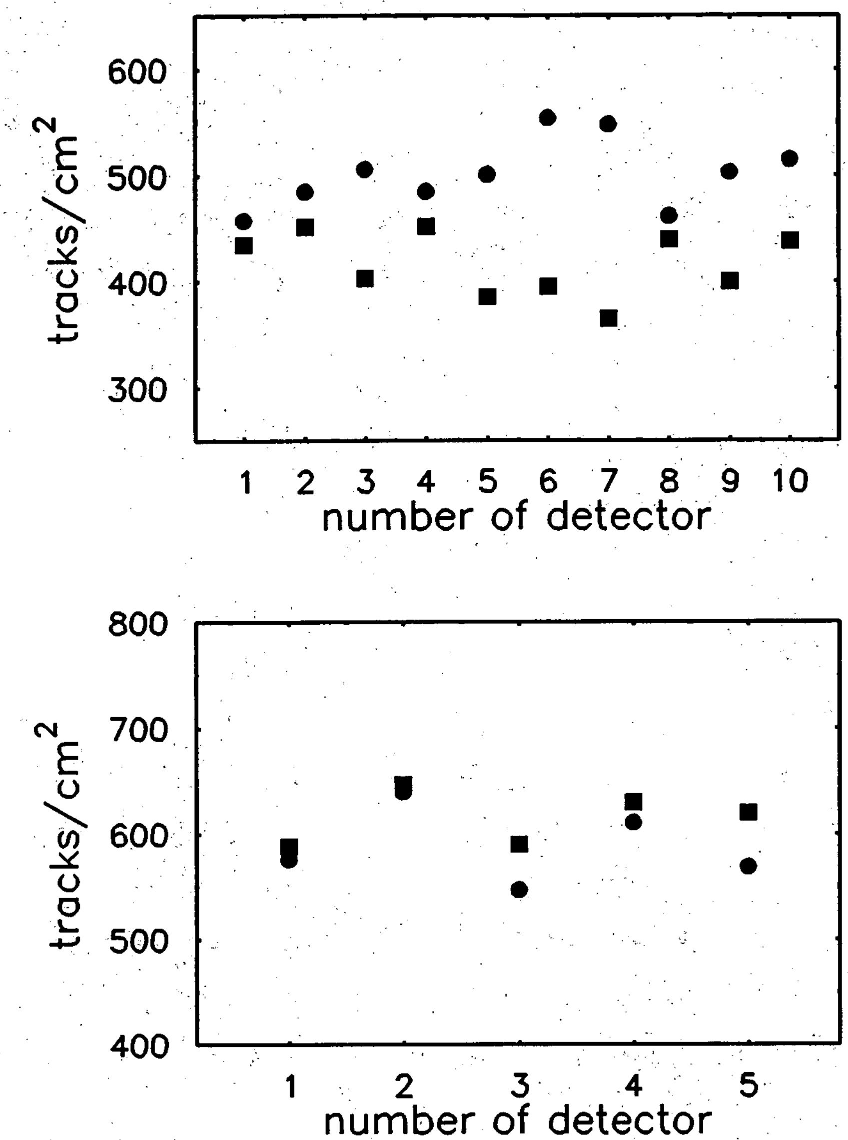


Fig. 5: Net track density reported from the other participants. In the upper part the data from ISIB are shown and in the lower part those from IHE. Different symbols (■ and •) indicate different exposures. In the upper part the higher track density corresponds to a lower exposure, because a heating procedure was applied to these detectors prior to exposure. Uncertainties due to counting statistics are not indicated. The RUG reported average track densities only.

Table 2: Average track densities and corresponding exposures reported by RUG, ISIB and IHE. The (*) indicates the batch which underwent thermal treatment prior to exposure.

		RUG	ISIB	IHE
run (1)	mean track density (tracks/cm^2)	516	455	589
	exposure (kBq/m^3 h)	870	880	1079
run (2)	mean track density (tracks/cm^2)	558	394*	615
	exposure (kBq/m^3 h)	950	940	1129

are equal within counting statistics as shown in Figs. 3 – 5. Furthermore, the data from SCK•CEN enforce a readjustment of their conversion factor for polycarbonate detectors by about 20 %. Therefore, in the following we first have to derive an approximation to the true exposures based on the results of all measurement techniques involved. As already explained in Chapts. (1) and (2) the results from the grab-sampling support the validity of the calibration factor of the continuous radon monitor, which were lower within 9 %. Additionally, our investigations on volume-traps as retrospective radon monitors [7] lead to a confirmation of the continuous radon monitor within 6 %. On the other hand the previous calibration factor of 0.55 tracks/cm^2/(kBq/m^3 h) was confirmed within 5 % at two international intercalibration exercises [2,3].
In Tab. 2 the results of all participants are summarized. The indicated error on the reported exposure, $\Delta E/E$, is with respect to the reference value of SCK•CEN.

Thus, in the following we have to look for a possible explanation for the variability of the calibration factor. In ref. [8] it was shown that the sensitivity increases with increasing values of the average track diameter. Parameters, which were identified to influence the track diameter, are the temperature and concentration of the etching solution, the length of the etching period, the applied voltage and frequency. A temperature variation of 1°C e. g. results in a difference of 7 %. Thus, a cumulating effect could be responsible for the observed deviation of up to 20 %. In Fig. 6 etched detectors from ref. [1] and from the current exercise after approximately the same exposure are compared. The average track

Table 3: Reported exposures E from all participants of the intercalibration exercise at SCK•CEN from December 1994. The deviation $\Delta E/E$, is with respect to the reference value of SCK•CEN; SCK*) without readjustment of the calibration factor and SCK**) calculated from the present experimental data.

	SCK•CEN	RUG	ISIB	IHE	SCK*	SCK**
exposure (1) (kBq/m^3 h)	853	870	880	1079	1034.8	864
$\Delta E/\bar{E}$ (%)	-	2.7	3.2	26	21.3	1.3
exposure (2) (kBq/m^3 h)	883	950	940	1129	1076.9	901
$\Delta E/\bar{E}$ (%)	-	7.6	6.5	28	22.0	2.0

size of the current detector is significantly larger leading to a higher conversion factor. Unfortunately, it is not possible to determine retrospectively, which parameters caused the difference in track sizes. Furthermore, there may be an additional effect on the sensitivity from ageing of the polycarbonate detector material. From the results of the ISIB detectors it is clear, that detectors undergoing a thermal treatment to decrease the background have a different calibration factor.

5. New SCK•CEN calibration factor

From the present intercalibration exercise two main conclusions may be drawn. First, there is a dependence of the calibration factor, i. e. read-out sensitivity, with respect to the mean track-diameter of a polycarbonate detector. The introduction of a correction factor as a function of the mean track-diameter is only possible, if the read-out procedure is performed involving computer-based image-analysis techniques. Since such a system has already been used at SCK•CEN for the calibration of CR-39 track-etch detectors [9], detailed investigations will start after some minor modifications will have been applied to the system.
Secondly, as long as the observed variation of the read-out sensitivity is not under control, the parameters (a, b) from eq. (1) for polycarbonate detectors at SCK•CEN will be adapted

to

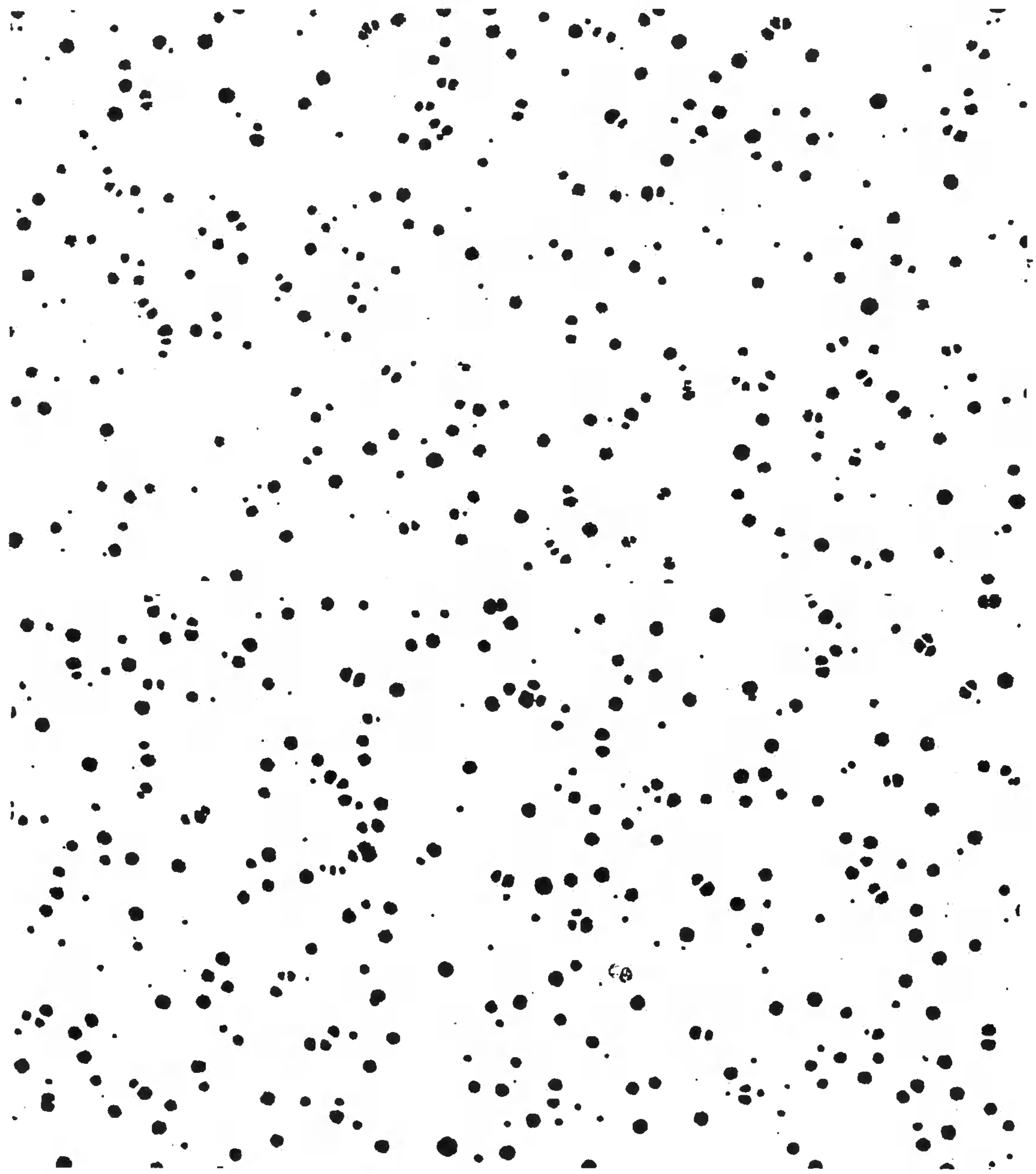
$$y < 2700 \text{ tracks/cm}^2$$
$$a = 7000 \text{ tracks/cm}^2$$
$$b = 9.0 \times 10^{-5} \text{ m}^3/(\text{kBq h})$$

$$y > 2700 \text{ tracks/cm}^2$$
$$a = 2.3 \times 10^4 \text{ tracks/cm}^2$$
$$b = 2.3 \times 10^{-5} \text{ m}^3/(\text{kBq h})$$

The initial slope (ab) is 0.63 tracks/cm^2/(kBq/m^3 h), which is lower than the value derived from the current exercise but higher than the one determined in ref. [1]. On that basis our present results agree within 10 % with the reference values from the continuous radon monitor.

A recalculation of the data from the RUG intercalibration (E = 746 kBq/m^3 h), where the same polycarbonate batch as for the present exercise was used brings our result back (E = 647 kBq/m^3 h) to reasonable consistency with the average exposure of 641 kBq/m^3 h.

a)



b)

Fig. 6: Differece in average track-diameter in etched polycarbonate detectors. Here, detectors are shown after approximately the same exposure. The upper part show tracks from ref. [1] having a smaller average diameter than from the current exercise (lower part), which in the present case leads to a higher conversion factor.

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