Radiation Protection Dosimetry (2006), 1 of 8
doi:10.1093/rpd/ncl161

INFLUENCE OF ENVIRONMENTAL CHANGES ON INTEGRATING RADON DETECTORS: RESULTS OF AN INTERCOMPARISON EXERCISE

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Received July 27 2006, amended September 18 2006, accepted September 23 2006

An intercomparison exercise for passive integrating radon detectors has been carried out with the participation of 12 detection systems from 10 laboratories. The detection systems comprise three commonly used in radon integrating measurements, tracks, activated charcoal canisters and electrets. The exposures were carried out in the radon and thoron chambers at the Institute of Energy Techniques (INTE) of the Technical University of Catalonia (UPC), which is considered to be the Spanish reference chamber. The detectors were exposed to three different temperatures (10, 20 and 30°C) and relative humidities (30, 45 and 80%). Furthermore, in three exposures radon concentration was drastically changed during the exposure period in order to study the efficiency of canister collection. The results indicated that only the charcoal canister response was found to be significantly influenced by external climatic conditions and radon fluctuations. Those track detectors, which are unable to measure thoron concentrations show thoron sensitivity and thus interfere with precise measurement of radon. Detectors for measuring thoron concentration show quite a different response, which could be related to their traceability.

INTRODUCTION

The 96/29/Euratom Directive aims to introduce radon regulation at work places in European countries. Within this framework, the Spanish nuclear regulatory body, the Nuclear Safety Council (CSN), is interested in obtaining a reliable quality level for radon measurements in Spain. The Radon Laboratory Group at the Institute of Energetic Techniques (INTE) of the Technical University of Catalonia (UPC) in Barcelona has been commissioned to set up its radon chamber for testing radon instruments and has carried out intercomparison campaigns.

The CSN, in collaboration with the INTE, has launched two series of intercomparison exercises among several Spanish laboratories which are involved in radon measurement studies. The first exercise was carried out during 2001–2002, with the participation of 13 passive integrating radon detectors from 11 groups. In this first campaign the environmental conditions were maintained constant under standard conditions and the results have roughly shown that the reliability of these systems was acceptable. The results and conclusions of this intercomparison were presented in Ortega *et al.*[1]

However, it is a well-known fact that these radon detectors could be sensitive to environment parameters such as temperature, humidity and other environmental parameters[2,3]. Thus, a second intercomparison exercise was carried out during 2004–2005. This intercomparison exercise was done in parallel with a continuous radon monitor intercomparison presented in a previous paper[4]. In these campaigns, the influence of changing exposure conditions on the measurement systems was analysed in order to study the response of the detectors used by the participating radon groups. Therefore, variation of radon concentration, temperature, humidity level and the presence of thoron were analysed by different exposures in the INTE radon and thoron chambers.

In this paper the results and conclusions of the second intercomparison for passive integrating radon detectors exercise carried out in the INTE chambers are presented.

MATERIALS AND METHODS

Exposure of radon detectors was carried out in the INTE radon and thoron chambers. A brief description of both is presented below.

The radon chamber

The radon chamber[5] at the INTE basically consists of a 20 m^3 volume with an air lock. The walls are made of electrically grounded stainless steel and it is thermally isolated to prevent any stray collection of radon progeny because of electrostatic effects and to ensure good temperature control within the chamber.

Radon measurements are continuously carried out at least every hour. The alpha-spectra obtained with the ATMOS reference instrument were analysed in order to estimate radon concentrations. A typical spectrum of the ATMOS can be seen in Figure 1.

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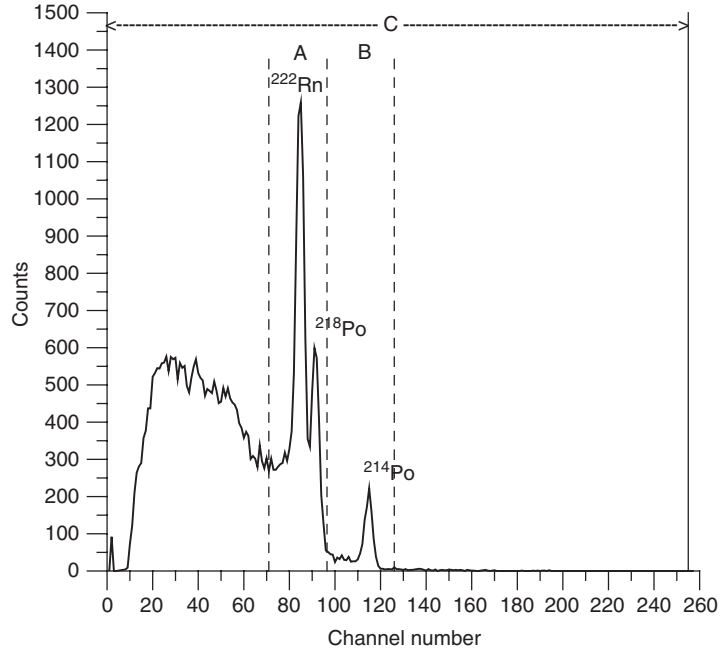

Figure 1. Alpha-spectrum obtained by the ATMOS radon monitor.

Radon concentration can be obtained by the following expression:

$$C_{222_{Rn}} = \frac{C_1(NC_{1n} - rNC_{2n})}{t - C_3 NC_3}, \quad (1)$$

where C_{Rn-222} is the radon concentration in Bq m^{-3}; $NC_{1n} = NC_1 - BKG_1$ are the net counts in region A (Figure 1); $NC_{2n} = NC_2 - BKG_2$ are the net counts in region B (Figure 1); NC_3 are the counts in region C (Figure 1); NC_1 are the counts in region A; NC_2 are the counts in region B; BKG_1 are the background counts in region A; BKG_2 are the background counts in region B; r is the interference parameter representing the ratio of the counts in region A to the count in region B due to decays of ^{214}Po; C_1 is the calibration parameter determined by means of a radon calibration in Bq m^{-3} s; C_3 is the dead time parameter which is determined by the manufacturer and its value is 0.0065; t is the measurement integration time in seconds.

The background parameters, BKG_1 and BKG_2, were estimated at the INTE by circulating synthetic-air within the detection volume of the monitor and analysing three 1 h spectra. The BKG_1 and BKG_2 were estimated at 3.8 and 0.25 h^{-1}, respectively.

The interference parameter, r, was determined by dividing NC_{1n}/NC_{2n} when the atmosphere within the ATMOS volume detection was free of ^{222}Rn and ^{218}Po and it only contained ^{214}Po. To generate this atmosphere, synthetic-air had been circulating for 30 min through the detection volume after an exposure at high radon levels. A 1 h spectrum was then analysed and the r interference parameter and its standard uncertainty was found to be 1.28 ± 0.10.

The calibration factor, C_1, was obtained according to the calibration procedure used in the INTE radon laboratory[5]. The C_1 value and its standard uncertainty was 2735 ± 112.

The combined uncertainty for radon concentration can be obtained from equation 1. The expanded uncertainty is ~10% with a coverage factor of $k = 2$, which corresponds to a coverage probability of ~95%.

The thoron chamber

The thoron chamber and measurement procedure has been described in a previous paper[4]. Basically, the alpha-spectra obtained with the ATMOS were analysed in order to estimate thoron concentrations. The alpha-particles from ^{220}Rn (6.29 MeV), ^{216}Po (6.78 MeV), ^{212}Bi (6.05–6.09 MeV) and ^{212}Po (8.78 MeV) can be counted by an analytical fitting procedure, using gaussian functions and a semi-empirical low-energy function, typical of the instrument. The monitor was calibrated at the Physikalisch-Technische Bundesanstalt (PTB) by comparing the reading with a calibrated ATMOS from the PTB. The measurement procedure and the calibration facility from PTB[6]. The uncertainty

Table 1. Participating laboratories in the intercomparison.

Department	Organisation	City
Radiation Protection Service	Health Institute Carlos III (ISCIII)	Majadahonda (Madrid)
Laboratory of Environmental Radioactivity	University of Oviedo (UO)	Oviedo
Laboratory of Environmental Radioactivity	University of Valencia (UV)	Valencia
Laboratory of Medical Physics and Environmental Radioactivity	University of La Laguna (ULL)	La Laguna (Tenerife)
Institute of Energy Technologies—Laboratory of Radon Studies	Technical University of Catalonia (UPC)	Barcelona
Department of Medical and Surgical Sciences—Laboratory of Medical Physics	University of Cantabria (UC)	Santander
Department of Psychiatry, Radiology and Public Health—Laboratory of Public Health	University of Santiago de Compostela (USC)	Santiago de Compostela
Department of Physics, Radiation Physics Unit	Autonomous University of Barcelona (UAB)	Bellaterra (Barcelona)
Research Center for Radiation Safety	National Institute of Radiological Sciences (NIRS)	Chiba (Japan)
Physics Department	University College Dublin (UCD)	Dublin (Ireland)

Table 2. Identification of each monitor and measurement system.

ID-detector	System	Measure	Calibration	Device
1	Tracks CR-39	Radon and thoron	NIRS	Rado-pot[7]
2	Tracks CR-39	Thoron	NIRS	NRPB/SSI
3	Tracks CR-39	Radon	Radosys calibration	Radosys RSFS
4	Tracks Makrofol	Radon and thoron	NRPB for radon. Theoretical for thoron	KfK FN Polyetheline and glass-fibre filter for radon KfK FN two glass-fibre for thoron
5	Activated charcoal	Radon	Ra-226 source and INTE-UPC chamber.	Canister EPA no diffusion barrier
6	Tracks CR-39 for radon and thoron	Radon and thoron	INTE-UPC chamber for radon. Theoretical for thoron	Radosys
7	Activated charcoal	Radon	Ra-226 source	Canister EPA no diffusion barrier
8	Tracks Makrofol	Radon	NRPB	KfK A
9	Activated charcoal	Radon	Ra-226 source	Canister EPA no diffusion barrier
10	Electret	Radon and thoron	NIST for radon. CANMET for thoron	E-perm SLT for radon E-perm SST for thoron
11	Tracks CR-39	Radon	INTE-UPC chamber	Landauer
12	Tracks CR-39	Radon and thoron	NIRS	Radouet

contribution from the reference instrument is ~5% (PTB certificate) and a 5% contribution is added due to the calibration of the instrument used for measuring thoron concentrations in the chamber. The combined uncertainty for the conversion factor from counts of thoron concentration was estimated from the square root of the quadratic sum of all the relative uncertainty contributions. The expanded uncertainty is ~15% with a coverage factor of $k = 2$.

Participants and passive integrating radon detectors

Twelve passive integrating radon detection systems were used in the intercomparison exercise involving ten laboratories, eight of which were from Spain. Table 1 shows the participants and Table 2 shows the detection system, the identification number, calibration and type of instrument. The ID-2 detector only participated in the thoron intercomparison.

The radon measurement systems for the different laboratories comprise the three most commonly used systems as follows: tracks, activated carbon canisters and electrets. All the measurement systems are passive and the air enters the detection volume by diffusion and, depending on design characteristics, a filter is used in order to prevent the radon progeny from getting into the volume. A description of the different measurement systems can be found elsewhere[8].

ORGANISATION

Spanish participants of the intercomparison were selected by the CSN by directly contacting the Heads of the different laboratories, and the intercomparison programme was sent to them. The two participants from outside Spain were contacted directly by the INTE. The programme contained the scheduled exposures, management of the detectors and data, confidentiality of the results from each laboratory and other conditions in order to explain the intercomparison process. Furthermore, the Heads of the laboratories were asked to fill in a questionnaire and to sign acceptance of the intercomparison conditions established in the programme.

For the different exposures in the radon and thoron chambers, three detectors of each type were exposed. For the laboratories which use charcoal canisters, six more detectors were needed in order to study their response due to temporal radon fluctuation. Finally, two detectors for each type were sent for transit. According to the intercomparison programme, 23 detectors were sent for tracks and electrets and 29 for carbon canisters.

Detectors were sent to the INTE radon laboratory with instructions for their correct use. Once the detectors were at the INTE, each one was identified and placed in a low-radon room. The detectors were then randomly selected for the different exposures and transit.

The detectors were placed homogenously in the radon chamber in a volume of ~ 2 m^3 in which the radon concentration homogeneity was determined to be better than 5% and the radon concentration stability did not exceed 10%.

Afterwards, the track detectors were exposed in the chambers. They were placed in a low-radon room for 1 h in order to allow the radon concentration to decrease in the detection volume. They were then placed inside air-tight bags and kept in a low-level radioactivity room until they were sent.

The carbon activated detectors and electrets were sent to the corresponding laboratory immediately after each exposure, track detectors were sent together at the end of the exercise.

The results from each laboratory were sent to the INTE in order to be analysed. A final report was given to the CSN and distributed to all the Spanish participants.

EXPOSURES

According to the intercomparison programme, detectors were exposed inside the radon and thoron chambers at different stages with the aim of determining the thoron response and the influence of changing temperature, humidity and radon concentration.

Exposures in the thoron chamber

Two series of exposures were carried out in the thoron chamber. In order to estimate the thoron uncertainty in the chamber, contributions from the thoron measurement instrument were considered and estimated as described above, and the thoron concentration heterogeneity in the chamber was estimated to be 5% of the thoron concentration level. Table 3 shows the exposure period of the two series, the mean thoron concentration, the expanded uncertainty with a coverage factor of $k = 2$ and the identification numbers of the exposed detector type. In these exposures radon concentration was lower than 30 Bq m^{-3}.

Exposures in the radon chamber

In order to estimate the radon uncertainty in the chamber, contributions from the radon measurement instrument were considered and estimated as described above, the radon concentration heterogeneity in the chamber was estimated to be 5% of the radon concentration level.

Table 3. Exposures carried out in the thoron chamber.

Exposure	Exposure period		Number of measurements by the reference instrument	$C_{220_{Rn}} \pm U$ ($k = 2$)	ID-detector
1st series	30/03/05	17:50	75	512 ± 89	5, 7, 8, 9, 10
	02/04/05	21:10			
2nd series	04/04/05	12:45	76	550 ± 94	1, 2, 3, 4, 6, 11, 12
	07/04/05	17:25			

Table 4. Exposures carried out in the radon chamber for climatic analysis.

Exposure	Exposure period		Number of measurements by the reference instrument	$C_{^{222}Rn} \pm U$ $(k = 2)_{reference\ (Bq\ m^{-3})}$	$T_{reference}$ (°C)	$Hr_{reference}$ (%)
A	15/04/05	08:50	76	8267 ± 1157	20	45
	18/04/05	09:20				
B	18/04/05	15:10	72	8545 ± 1196	20	30
	2104/05	15:30				
C	22/04/05	16:00	66	8333 ± 1167	10	45
	25/04/05	09:30				
D	25/04/05	15:55	71	9473 ± 1321	30	45
	28/04/05	15:30				
E	29/05/05	10:55	71	8932 ± 1250	20	80
	02/05/05	10:40				
F	02/05/05	13:20	69	9771 ± 1368	30	80
	05/05/05	10:45				

Table 5. Exposures carried out in the radon chamber for radon fluctuation analysis.

Exposure	Exposure period (d)	$C_{^{222}Rn} \pm U$ $(k = 2)$ reference (Bq m^{-3})	$C_{^{222}Rn} \pm U$ $(k = 2)$ reference average (Bq m^{-3})	$T_{reference}$ (°C)	$Hr_{reference}$ (%)
G	0–1.5	8249	8267 ± 1157	20	45
	1.5–3	8285			
H	0–1.5	10	4193 ± 587	20	45
	1.5–3	8285			
I	0–1.5	8249	4130 ± 578	20	45
	1.5–3	10			

Table 6. Average exposure obtained by transit detectors.

ID-detector	kBq m^{-3} h
1	3
2	0
3	29
4	0
5	0
6	32.5
7	0
8	27.5
9	0
10	0
11	0
12	7

In Table 4 the different exposure for analysing the climatic effects are presented. Exposures A, B and E were carried out in order to evaluate the influence of the relative humidity. The influence of temperature influence was analysed using exposures A, C and D. Exposure F was carried out in order to study the response of detectors in extreme conditions.

Finally, in order to study the response of activated charcoal detectors when radon concentration fluctuates, exposures G, H and I are divided into two periods, one at a low-radon concentration and the other at a high concentration. In Table 5 the different exposures are given.

RESULTS AND ANALYSIS

The results of the different laboratories are analysed in four sections, transit detectors, thoron response, climatic influence and effects of radon fluctuations in carbon canisters.

Transit detectors

The results of the transit detectors are presented in Table 6. These values are subtracted from the exposure results given by the different laboratories in the radon and thoron chambers.

The ID-8 has a high value in the transit detectors, probably due to the laboratory bags not being properly air-tight. The ID-3 and ID-6, both Radosys systems, have also high values in the transit detectors

Table 7. Results of radon concentrations obtained in the presence of thoron.

Exposure	$C_{220_{Rn}} \pm U$ ($k = 2$)	ID-detector (RT%)											
		1	2	3	4	5	6	7	8	9	10	11	12
1st series	512 ± 89					0		0	48	0	9		
2nd series	550 ± 94	16	[a]	118	21		0					161	10

[a]No data

Table 8. Response to thoron concentrations.

Exposure	$C_{220_{Rn}} \pm U$ (Bq m^{-3}) reference ($k = 2$)	ID-detector $C_{220_{Rn}}$ (Bq m^{-3})					
		1	2	4	6	10	12
1st series	512 ± 89					1226	
2nd series	550 ± 94	1486	1353	489	391		1004

and these laboratories should analyse the reason for these high levels.

Influence of the presence of thoron

In Table 7, the response of the radon detectors to the presence of thoron is shown. In order to carry out the analysis of the influence of thoron, the RT(%) parameter was introduced, which is defined as a ratio expressed in percentage, between radon concentration measured by the detection system and thoron concentration in air. It can be seen that the RT parameter is higher for ID-3, ID-8 and ID-11, which are the track detectors that can not measure thoron concentration.

The results are presented in Table 8 for detectors which are able to measure thoron concentration. Detectors ID-1, ID-2 and ID-12 have the same traceability to the National Institute of Radiological Sciences (NIRS, Chiba, Japan), ID-10 was calibrated by Rad Elec Inc. Company and is traceable to the thoron test facility of CANMET, Elliot Lake Laboratory (Canada), and thoron concentration in ID-4 and ID-6 detectors are theoretical calibrations.

Influence of changing climatic conditions

The influence of changing climatic conditions was analysed using the calibration factor, FC.

$$\mathrm{FC} = \frac{C_{\mathrm{ref}}}{C_{\mathrm{eq}}} \tag{2}$$

where C_{ref} is the mean radon concentration measured by the reference instrument; and C_{eq} is the mean radon concentration measured by an ID-detector minus its transit concentration.

The calibration factor uncertainty, U(FC), is estimated by combined uncertainties according to the EA-4/02 guide[9]. In Table 9, calibration factor and expanded uncertainty U(FC) with a coverage factor k, which for a t-distribution with v_{eff} estimated from the Welch–Satterthwaite formula corresponds to a coverage probability of ~95%, are shown for the exposures and detectors.

Influence of changing radon concentration

Exposures G, H and I show the effect of varying radon concentration on the calibration factor for activated charcoal canisters. The calibration factor and expanded uncertainty are presented in Table 10. From these data it can be seen that the final days weigh much more than the first few days for radon concentration estimation. This fact is due to the adsorption and desorption process in the activated charcoal, as has been described in a previous study[10].

Since radon concentration fluctuation is not known during the measurement period, it is important to consider that an error can be occur if the possible fluctuation is not taken into account in the uncertainty evaluation. It is therefore appropriate for radon concentration measurements carried out with canisters that the radon concentration is maintained as constant as possible by closing windows, doors and other variable ventilation systems.

CONCLUSIONS

The 12 detectors from 8 Spanish laboratories and two from outside Spain, all of which belonged to public institutions, comprise the three most common techniques used for passive integrating

Table 9. Calibration factor for different environment conditions and detectors.

ID-detector	Exposure					
	A (20°C 40%)	B (20°C 30%)	C (10°C 45%)	D (30°C 45%)	E (20°C 80%)	F (30°C 80%)
1	0.89 ± 0.15	0.79 ± 0.15	0.98 ± 0.14	0.90 ± 0.18	1.02 ± 0.18	1.00 ± 0.16
3	0.93 ± 0.18	0.98 ± 0.17	0.94 ± 0.14	0.97 ± 0.15	0.95 ± 0.19	1.13 ± 0.18
4	1.12 ± 0.17	1.02 ± 0.15	1.04 ± 0.16	1.19 ± 0.16	1.09 ± 0.15	1.23 ± 0.18
5	0.98 ± 0.13	0.83 ± 0.13	0.77 ± 0.11	1.55 ± 0.21	0.97 ± 0.20	[a]
6	1.00 ± 0.14	1.07 ± 0.16	1.10 ± 0.17	1.15 ± 0.16	1.23 ± 0.17	1.20 ± 0.16
7	0.97 ± 0.18	0.94 ± 0.13	0.75 ± 0.11	1.32 ± 0.22	[a]	[a]
8	1.72 ± 0.32	2.03[b]	1.64 ± 0.24	1.68 ± 0.29	1.67 ± 0.35	1.68 ± 0.26
9	0.92 ± 0.12	0.87 ± 0.12	0.74 ± 0.10	1.22 ± 0.16	[a]	[a]
10	1.01 ± 0.14	0.98 ± 0.14	0.98 ± 0.14	1.03 ± 0.14	1.02 ± 0.14	1.02 ± 0.14
11	1.41 ± 0.29	1.47 ± 0.23	1.50 ± 0.22	1.66 ± 0.31	1.95 ± 0.31	2.37 ± 0.47
12	0.86 ± 0.13	0.88 ± 0.12	0.83 ± 0.11	0.92 ± 0.12	0.95 ± 0.13	1.01 ± 0.17

[a]Out of range calibration
[b]Only one detector was exposed

Table 10. Calibration factor for charcoal canister detectors for different radon concentration temporal evolution.

Exposure	Exposure period (d)	Concentration in the two periods (Bq m^{-3})	C_{ref} average (Bq m^{-3})	FC ID-detectors		
				5	7	9
G	0–1.5	8249	8267	0.98 ± 0.13	0.97 ± 0.18	0.92 ± 0.12
	1.5–3	8285				
H	0–1.5	10	4193	0.57 ± 0.10	0.57 ± 0.10	0.57 ± 0.10
	1.5–3	8285				
I	0–1.5	8249	4130	4.93 ± 0.74	5.86 ± 1.05	2.83 ± 0.42
	1.5–3	10				

radon concentration measurement: tracks, electrets and activated charcoal canisters.

Different exposures were carried out in order to analyse the reliability of the detectors to changing climatic conditions and radon concentrations, and the influence of the presence of thoron concentration.

Analysis of the results has led to the following conclusions:

In exposure A (20°C, 45%), which is considered as standard climatic conditions, the calibration factor for detectors ID-8 and ID-11 shows a significant deviation from the true value.

Electret detectors show a correct response to temperature and humidity variation.

Track detectors show, in general, a correct response to temperature and humidity variation. Only ID-11 shows a significant deviation for high humidity exposures E (20°C, 80%) and F (30°C, 80%).

Activated charcoal canisters are sensitive to temperature. From exposure A (20°C, 45%), C (10°C, 45%) and D (30°C, 45%) it can be concluded that the lower temperature, the higher the radon absorption.

The laboratories which use activated charcoal canister employ a correction factor by measuring the weight increment due to water adsorption. If the detector is not saturated, then the response is acceptable.

Activated charcoal canisters show that they do not measure a real integration of the exposure due to the absorption desorption process. The weight of the final period of the exposure is much more significant than the first period.

Activated charcoal canisters do not measure thoron and there is no influence by the presence of this radionuclide in the air.

Those track detectors with no capacity to measure thoron concentration (ID-3, ID-8 and ID-11) are sensitive to thoron and interfere with precise measurement of radon, if thoron is present in air.

Detectors with the capacity to measure thoron concentration show quite different results. It has been observed that these differences could be due to different traceabilities.

The intercomparison study carried out for passive integrating detectors in Spain provides the CSN with a tool for obtaining better knowledge of the level of

quality of different laboratories involved in radon measurements. Furthermore, the intercomparison allows the laboratories to improve the quality of their radon measurements.

Finally, due to the importance of maintaining a good quality level of radon measurements, it is expected that these intercomparison studies will be carried out biennially.

ACKNOWLEDGEMENTS

This work has been partially supported by the Spanish CSN. The authors would like to thank José Luís Martín Matarranz (CSN) for his work in organising the intercomparison, and Vicente Blasco (INTE) and Jaume Bonet (INTE) for their collaboration in the preparation and development of exposures in the radon chamber.

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