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Natural Radioactivity in Tanzania Cements and their Raw Materials

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Abstract: This paper presents the study of natural radioactivity in Tanzania Portland cements and their raw materials. Samples collected as raw materials were pozzolan, sandstone, limestone, clay, gypsum and cement as finished products. The natural radioactivity due to the presence of radium ^{226}Ra, thorium ^{232}Th and potassium ^{40}K were measured by means of gamma spectrometer coupled with HPGe detector. The mean measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the raw materials range from 2.6 to 93.2, 1.3 to 172.8 and 6.3 to 997 Bq/kg, respectively with higher activity concentrations in pozzolan and lower in gypsum. Activity concentrations of natural radionuclides in raw materials (excluding some materials from Songwe deposits in Mbeya region) are comparative with the worldwide average concentrations of these radionuclides in soil. The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the cements are 46, 28 and 228 Bq/kg, respectively. The calculated values of radiological indices are below 60% of the upper recommended values for building materials. The average annual effective dose to an occupant from use of these materials equals to 0.45 mSv. Average activity concentrations of the mentioned radionuclides in Tanzania cements are in the middle of the variability interval of the national averages.

Keywords: Annual effective dose, building materials, gamma-ray spectrometry, radium equivalent, radiation hazard

INTRODUCTION

The natural radionuclides from the uranium-radium series, thorium series and potassium K-40 are present in all raw and manufactured building materials. The Portland cement which is the finished good produced from the rock and soil materials is usually incorporated with natural radionuclides therefore, their use in the construction of residential and commercial buildings results into external and internal radiation exposure to the population. From the radiological point of view, the most important exposure with regards to health is from radon (^{222}Rn, ^{220}Rn) and its short-lived decay products which become the major source of internal exposure when inhaled. The external dose is due to radiation exposure by gamma rays emitted from the radionuclides in the building materials. Determination of radionuclides present in cement is crucial and vital because several investigated building materials including cements in some regions around the world were identified to possess elevated levels of natural radionuclides contributing to an annual effective dose of more than 1 mSv which is the maximum permissible level (Stoulos *et al*., 2003; Krsti *et al*., 2007). Based on the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in the building materials, one can assess the radiological hazard connected with the use of these materials.

Cement is one of the most important materials used by the building industry in Tanzania. Most often the buildings are constructed from cement blocks and concrete; furthermore cement is used for plastering buildings. The five brands of cement available in the country trademarked Tembo, Simba, Twiga, Rhino and Kilwa, are produced from different mixture of raw materials include limestone, clay, sandstone, pozzolan and gypsum quarried from different places of different geological, geographical and geochemical conditions. The Tembo cement in Mbeya Southern Tanzania for instance, is made from limestone, clay, sandstone and pozzolan quarried from Songwe in Mbeya region and gypsum obtained from Makanya and Itigi in Kilimanjaro and Singida regions, respectively. The Rhino, Twiga and Kilwa cements produced in Dar es Salaam and Lindi regions are made from gypsum quarried from Kilwa and the rest of their raw materials obtained from Wazo Hill deposits in Dar es Salaam. On the other hand the Simba cement produced in Tanga cement factory use gypsum from Kilwa and Makanya and the rest of its raw materials obtained from its quarry located at Pongwe. Occasionally, Wazo Hill cement

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factory imports gypsum from Oman used for Twiga cement manufacture. It is worth adding that pozzolan is seldom directly used in Tanzania for plastering (with few centimetres thin wall) in local communities as well as gypsum as thin (1-3 cm) gypsum boards for walls and ceilings.

The levels of natural radioactivity in Tanzania gypsum from Makanya and Itigi used for cement and chalk manufacture has been assessed (Msaki and Banzi, 2000). However, the detailed information of the activity concentration of natural radionuclides in cements and other raw materials in Tanzania is not available.

The objective of this study therefore was to determine the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in Tanzania cements and raw materials using gamma-ray spectrometry. The obtained results were used for assessment of the adequate radiological indices, for classification of the investigated building materials and for estimation of the radiation hazard due to use of those materials.

MATERIALS AND METHODS

Sampling and sample preparation: In this study, four regions of Tanzania were selected for collecting raw samples used in the production of five brands of cements. The regions include: Lindi (Southern east of Tanzania) where the gypsum deposits are located in Kilwa at Makaanga village, Mbeya region, southern Tanzania, where the limestone, sandstone, pozzolan and clay deposits are located at Songwe. The other regions include Dar es Salaam and Tanga, where the limestone, red clay and sandstone are found at Wazo Hill and Pongwe deposits respectively located close to the their respective cement factories.

A total of 25 raw material samples in amounts, weighing from 0.5 to 1 kg were collected. Part of these samples was directly collected from Songwe and Kilwa deposits and other samples were collected from Tanga and Wazo Hill cement factories. The 4 samples of different brands of cements were collected from the local market. The localizations of sampling sites are summarized in Table 1 and in Fig. 1; elevation and coordinates of the sampling sites were determined using a global positioning system (GPS map 60CSx).

The raw material samples were collected according to standard sampling procedures (IAEA, 1989), for environmental sampling. The collected samples were then placed in labelled polyethylene bags and transported at AGH University of Science and Technology in Krakow, Poland, where the measurements of the natural radioactivity were conducted.

Gamma spectrometry and activity measurement: The preparation of samples for radiometric measurements was performed in accordance with IAEA recommendations (IAEA, 1989) for environmental samples. The rock fragments, exceeding in a diameter of 3 mm, were discarded, the remaining material was dried to a constant weight at 105°C and milled to a grain size below 1 mm. The milled material was placed in aluminium cylindrical vessels of a volume of 121 cm^3 or Marinelli beakers of a volume 710 cm^3. The samples were sealed to prevent radon escape from the sample and were left for 4 weeks to reach secular equilibrium between the ^{226}Ra, ^{224}Ra and short-lived daughters of ^{222}Rn and ^{220}Rn, respectively.

The activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K were measured by gamma-ray spectrometry using a semiconductor HPGe detector (Canberra GX4020) with 42% relative efficiency and resolution of 1.9 keV at the line of 1333 keV, placed in lead housing with walls 10 cm thick. The IAEA reference materials RGU, RGTh and RGK (IAEA, 1987) were used as a standard source in the determination of the ^{226}Ra, ^{232}Th and ^{40}K, respectively. The uncertainty of the activity of these nuclides in standards should not exceed 2% (IAEA, 1987).

The ^{226}Ra activity concentration was determined from gamma lines of 609, 1120 and 1764 keV from ^{214}Bi. The ^{232}Th activity concentration was determined

Table 1: Mean activity concentrations of the investigated materials and the sampling coordinates

		Coordinates			Mean activity concentration Bq/kg		
Material	Location	Latitude	Longitude	Altitude (m asl)	^{226}Ra	^{232}Th	^{40}K
Pozzolans	Songwe	8˚56'10'' S	33°14'28" E	1291	93.2±2.8	172.8±5.2	997±29
	Pongwe	5˚06'36'' S	38°59'40" E	1279	35.2±1.1	50.8±1.5	225.7±6.8
Sandstone	Songwe	8˚55'03'' S	33°13'28" E	1253	18.3±0.5	30.3±0.9	501±15
	Wazo	6˚39'53" S	39°10'07'' E	1277	7.7±0.2	12.8±0.4	180.4±5.4
Clay	Songwe	8˚55'00'' S	33°13'27'' E	1254	90.7±2.7	123.3±3.7	137.7±4.1
	Pongwe	5˚06'36'' S	38°59'40'' E	1279	23.4±0.7	43.2±1.3	21.2±0.8
	Wazo	6˚39'53'' S	39°10'07" E	1277	14.6±0.5	38.0±1.1	164.6±4.9
Limestone	Songwe	8˚55'00" S	33°13'20" E	1268	6.4±0.4	15.2±2.5	80±11
	Pongwe	5˚06'36'' S	38°59'40'' E	1279	25.2±0.8	1.3±0.3	13.2±1.8
	Wazo	6˚39'53" S	39°10'07" E	1277	14.9±0.5	3.3±0.4	22.6±1.9
Gypsum	Kilwa	9˚29'44" S	39°18'36" E	1282	9.8±2.5	4.4±1.8	81 ±16
	Wazo	6˚39'53'' S	39°10'07'' E	1277	2.6±0.4	3.0±0.3	6.3±1.9

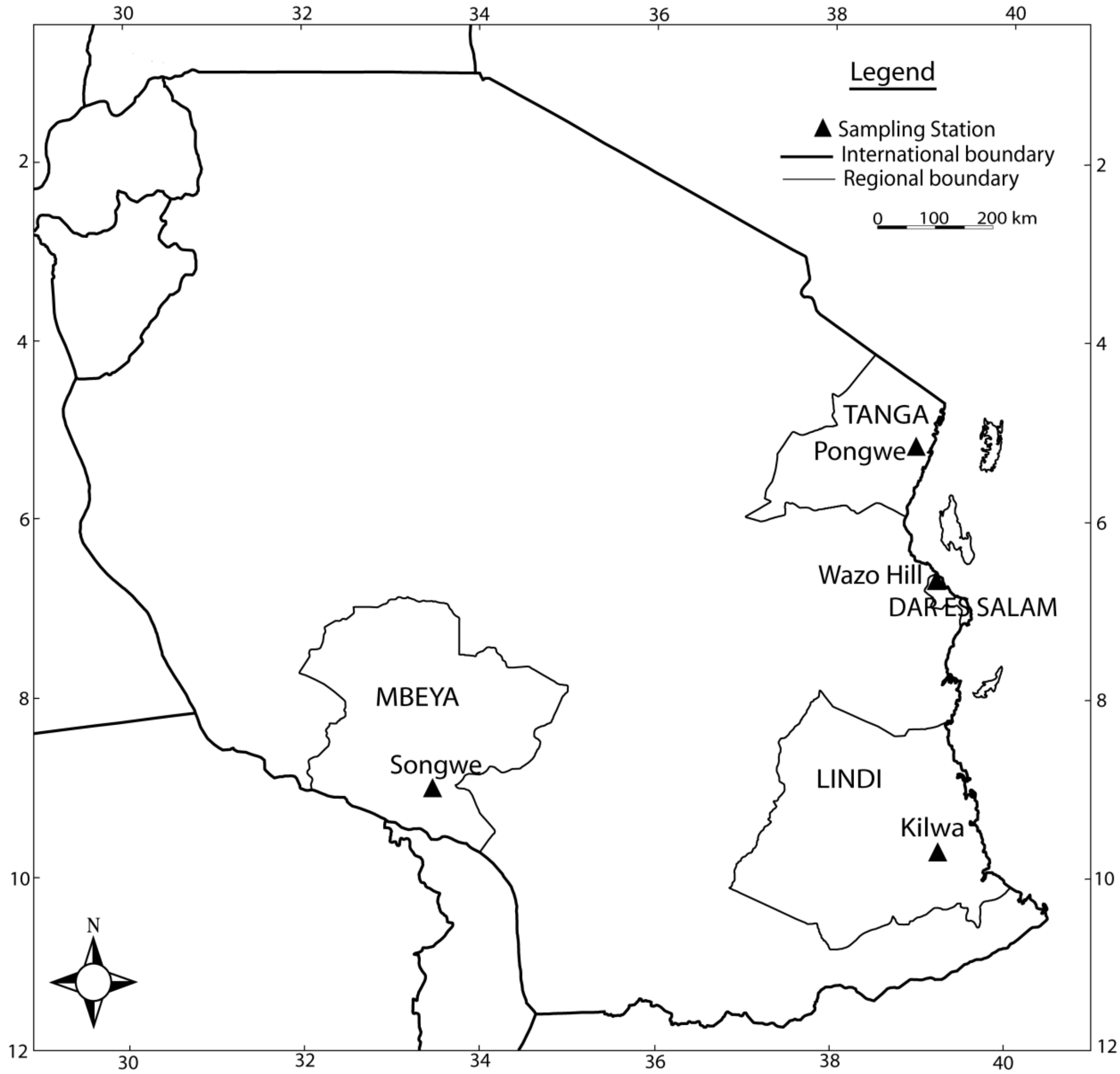

Fig. 1: Sketch of Tanzania showing sampling sites (names of the regions in capital letters)

from gamma lines of 911 and 969 keV from ^{228}Ac and 583 and 2614 keV from ^{208}Tl; in a case of disequilibrium between Ac-228 and Tl-208, gamma lines of Ac-228 were taken into account only. ^{40}K activity was determined from its 1461 keV gamma line.

The measurement time was generally chosen so that the relative uncertainty of the net counts of the peaks of 1120 keV, 969 keV and 1461 keV was less than 3%; in the case of very low active samples measurement time 40 h was chosen. In further calculation the differences in the densities of sample and standard were considered. A detailed description of the methodology is set out elsewhere (Jodłowski, 2006; Jodłowski and Kalita, 2010).

Radiation indices: The radiological hazard involved to the building materials is often assessed based on the radiological indices and dose which are calculated using the measured activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K

The most frequent used index is radium equivalent index Ra_{eq} (Bq/kg), defined as follows (Beretka and Mathew, 1985):

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (1)$$

where, $C_{Ra,}$ C_{Th}, C_K are the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

Recommended upper limit value of R_{eq} for dwellings is 370 Bq/kg.

The other parameters are external (H_{ex}) and internal hazard index (H_{in}), estimated by the following formulas (Beretka and Mathew, 1985):

$$H_{ex} = \frac{C_{Ra}}{370} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (2)$$

$$H_{in} = \frac{C_{Ra}}{185} + \frac{C_{Th}}{259} + \frac{C_K}{4810} \quad (3)$$

The external index corresponds to the gamma dose and the internal corresponds to the inhalation of the radon and its short-lived progenies. Both indices are dimensionless and their recommended upper limit value is 1.

In some recent publications (Trevis *et al*., 2012; Szabó *et al*., 2013) a dimensionless activity concentration index I, has been introduced and calculated as follows (EC, 1999):

$$I = \frac{C_{Ra}}{300} + \frac{C_{Th}}{200} + \frac{C_K}{3000} \quad (4)$$

The upper limit of this parameter amounts to 1.

The indoor absorbed dose rate in air D ($nGyh^{-1}$) and annual effective dose E involved from external exposition (mSv), can be calculated by formulas (EC, 1999; UNSCEAR, 2008):

$$D = 0.92\, C_{Ra} + 1.1 C_{Th} + 0.08 C_K \quad (5)$$

$$E = D \times F \times T \quad (6)$$

where, F is dose conversion factor from absorbed dose in air to effective dose Sv/Gy and T in annual indoor occupancy time 7000 h (0.8×365 days×24 h), assuming indoor occupancy factor 0.8 (UNSCEAR, 2008).

The I, D and E were estimated for a room with dimension 4×5×2.8 m with walls 20 cm of thickness and 2350 kg/m^3 of density (EC, 1999).

RESULTS AND DISCUSSION

The measured average activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K for the raw materials and their cement products and calculated associated radiological indices are presented in Table 1 to 3.

The radioactivity of the investigated raw materials, excluding pozzolan and clay from Songwe, vary in the intervals: 3-35 Bq/kg for ^{226}Ra, 1-51 Bq/kg for ^{232}Th and 6-500 Bq/kg for ^{40}K. These values are comparable with the worldwide average concentrations of these nuclides in soil equals to 32, 45 and 412 Bq/kg, respectively (UNSCEAR, 2008). All raw materials from Songwe deposit have higher activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K than the rest of materials in question except limestone where ^{226}Ra concentration is lower than those from Pongwe and Wazo. The fact can be connected with the local geology in Songwe deposits. The ^{40}K activity concentration in gypsum from Wazo is several times higher than those in the gypsum from Kilwa; on the hand results for ^{226}Ra and ^{232}Th are in agreement with those reported by Msaki and Banzi (2000).

The indices in all raw materials were also found to be within the reference levels except pozzolan from Songwe that was found to be up to few dozen per cent above the reference levels.

Generally the radioactivity in the analyzed limestone and gypsum from Tanzania is comparable with the radioactivity of these materials in other countries (cf. Turhan, 2010).

The activity concentration of ^{226}Ra in the cements was found in the range of 30 to 61 Bq/kg with an average of 46 Bq/kg, ^{232}Th from 18 to 48 Bq/kg with an average of 28 Bq/kg and ^{40}K varied from 78 to 500 Bq/kg with an average of 228 Bq/kg. The activity

Table 2: Calculated radiological indices of the investigated raw materials

Material	Location	Ra_{eq} (Bq/kg)	H_{ex} -	H_{in} -	I -	D (nGy/h)	*E (mSv)*
Pozzolans	Songwe	417	1.13	1.39	1.50	356	1.74
	Pongwe	125	0.34	0.44	0.45	106	0.52
Sandstone	Songwe	100	0.27	0.32	0.38	90.2	0.44
	Wazo	39.9	0.11	0.13	0.15	35.6	0.17
Clay	Songwe	278	0.76	1.01	0.96	230	1.13
	Pongwe	86.8	0.23	0.30	0.30	70.7	0.35
	Wazo	81.6	0.22	0.27	0.29	68.4	0.34
Limestone	Songwe	34.3	0.09	0.11	0.12	29.0	0.14
	Pongwe	28.1	0.80	0.15	0.09	25.7	0.13
	Wazo	21.4	0.06	0.10	0.07	19.2	0.09
Gypsum	Kilwa	22.3	0.06	0.09	0.08	20.3	0.10
	Wazo	3.5	0.01	0.02	0.01	3.2	0.02

Table 3: Activity concentrations of the investigated cements and radiological indices

	Activity concentrations			Radiological indices					
Brand	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Ra_{eq} (Bq/kg)	H_{ex} -	H_{in} -	*I* -	*D* (nGy/h)	E(mSv)
Rhino	61.0±1.8	25.7±0.8	168.2±5.1	110	0.34	0.41	0.39	97.6	0.48
Tembo	56.0±1.7	47.6±1.4	500±10	163	0.44	0.60	0.59	143.7	0.70
Kilwa	29.7±0.9	17.9±0.5	164.8±4.9	68	0.18	0.27	0.24	60.2	0.29
Simba	38.3±1.2	22.7±0.7	78.0±2.9	77	0.21	0.32	0.27	66.4	0.33
Average	46	28	228	104	0.28	0.40	0.37	91.4	0.45

Table 4: The average activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K in cements from several countries in the world (Khan and Khan, 2001) and references therein) including those of Tanzania cements and radiological indices calculated by the authors of this paper

	Activity concentration			Calculated radiological indices					
Country	^{226}Ra (Bq/kg)	^{232}Th (Bq/kg)	^{40}K (Bq/kg)	Ra_{eq} (Bq/kg)	H_{ex}	H_{in}	I	D (nGy/h)	E (mSv)
Australia	51.5	48.1	114	129	0.35	0.49	0.45	109.5	0.54
Austria	26.7	14.2	210	63.2	0.17	0.24	0.23	57.0	0.28
Algeria	41	27	422	112	0.30	0.41	0.41	101	0.50
Bangladesh	62.3	59.4	329	172	0.47	0.63	0.61	149	0.73
Brazil	61.7	58.5	564	189	0.51	0.68	0.69	166	0.81
China	56.5	36.5	173	122	0.33	0.48	0.43	106	0.52
Egypt	78	33.3	37	129	0.35	0.56	0.44	111	0.55
Europe[3]	45	31	216	106	0.29	0.41	0.38	92.8	0.46
Finland	40	20	251	87.9	0.24	0.35	0.32	78.9	0.39
Ghana	35.9	25.4	233	90.2	0.24	0.34	0.32	79.6	0.39
Greece[3]	85	19	257	132	0.36	0.59	0.46	120	0.59
India	37	24.1	432	105	0.28	0.38	0.39	95.1	0.47
Italy[3]	41	63	357	159	0.43	0.54	0.31	76.6	0.38
Japan	35.8	20.7	139	76.1	0.21	0.30	0.57	66.9	0.33
Netherlands[3]	62	64	271	174	0.47	0.64	0.62	149	0.73
Pakistan	26.7	28.6	273	88.6	0.24	0.31	0.32	77.9	0.38
Tunisia	21.5	10.1	176	49.5	0.13	0.19	0.18	44.9	0.22
Turkey	40	28	248	99.2	0.27	0.38	0.36	87.5	0.43
Tanzania present work	46	28	228	104	0.28	0.41	0.37	92.0	0.45

[3]quoted from Trevisi *et al*. (2012)

concentrations of ^{40}K and ^{232}Th were observed to be highest in Tembo cement; and in the case of ^{226}Ra the concentration was highest in Rhino and Tembo cements. The radioactivity of the Tembo cement is attributed to its raw materials and especially pozzolan from Songwe deposit that was observed to be elevated. The anomalous concentration of ^{226}Ra (61 Bq/kg) in Rhino cement may be connected with the use of other raw materials and it should be checked in future.

The calculated radiological indices of the Tanzania cements were found in the following ranges:

Ra_{eq} values-from 68 to 163 Bq/kg with an average of 104 Bq/kg
H_{ex} values-from 0.2 to 0.4 with an average of 0.3
H_{in} values-from 0.3 to 0.6 with an average of 0.4
I values-from 0.2 to 0.6 with an average of 0.4

All the values of the mentioned indices are below 60% of the upper recommended limits.

Indoor absorbed dose rate D originating from the Tanzania cements varied from 60 to 144 nGy/h with an average of 92 nGy/h, while the worldwide average indoor and outdoor absorbed dose rate amount to 84 nGy/h and 58 nGy/h, respectively (UNSCEAR, 2008). The annual effective dose to an occupant ranges from 0.30 to 0.70 mSv with an average of 0.45 mSv, while the worldwide average indoor exposure is equal to 0.41 mSv (UNSCEAR, 2008). Annual excess effective dose from the investigated cements (the excess in comparison with the dose outdoors (EC, 1999)) to an occupant is not higher than 0.2 mSv.

Table 4 summarizes average activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and corresponding radiological indices of cements from several countries in the world (Khan and Khan, 2001). Average activity concentrations of the Tanzania cements are in the middle of the variability range of mentioned national averages.

CONCLUSION

The activity concentrations of natural radionuclides of ^{226}Ra, ^{232}Th and ^{40}K and the associated radiological indices in Tanzania cements and their raw materials have been determined using gamma-ray spectrometry and adequate formulas.

Activity concentrations of natural radionuclides in raw materials (excluding some materials from Songwe deposits in Mbeya region) are comparable with the worldwide average concentrations of the radionuclides in soil.

The average activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in the cements were 46, 28 and 228 Bq/kg, respectively. The natural radioactivity was observed to be highest in Tembo cement; this fact is connected with the use raw material with elevated radioactivity from Songwe for its production. The variations of the radioactivity of the cements are due to the varying amounts of uranium, thorium and potassium contents in the different geological materials, where the deposits were exploited for producing the Portland brand cement. Average activity concentrations of the Tanzania cements are in the middle of the variability ranges in comparison from those of the dozen countries in the world.

All the values of the mentioned indices are below 60% of the recommended limits. The average annual effective dose to an occupant equals to 0.45 mSv; implies that for the investigated cements the annual excess effective dose does not exceed 0.2 mSv.

Based on the values of the radiological parameters obtained in this study, it can be concluded that cements used in Tanzania in the construction of dwellings is considered to be safe for occupants.

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