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Radioactivity of combustion residues from coal fired power stations

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In Germany every year about 18 million tonnes of combustion by-products are produced in power stations. These by-products are boiler slag, bottom ash and fly ash. They are utilized to a great extent in the construction industry and in mining.

Like any other natural rock, coals and consequently coal combustion by-products contain radioactive nuclides from the natural decay series and potassium-40. This is why they emit ionizing radiation. An examination of the radioactivity of solid material passing through power stations has shown that more than 90 % of the radioactivity in coal is found in the by-products.

In this paper, it will be examined whether or not the handling of coal ash in power plants and its utilization as building material leads to an increase in radiation exposure which is injurious to health for the personnel or the public.

In conclusion it can be said that

- the exposure of workers handling coal ash inside the power station is only insignificantly higher than the natural human radiation exposure
- the use of by-products in building materials entails a negligible portion of radiation dose which results from living in dwellings only.

Introduction

Like any other natural rock, coal, and consequently coal ash, contains radioactive nuclides resulting from the natural decay series. This is why they emit ionizing radiation. In the following, it will be examined if operational handling and disposal of coal ash as well as its utilization in the production of building materials lead to an increase in radiation exposure that is health-endangering to the personnel or the public.

According to a publication of German authorities, the natural human radiation exposure in Germany is between 2 and 6 mSv/a (effective radiation dose) with a mean value of 2.4 mSv/a (table 1) [6]. Along with the civil radiation exposure of 1.5 mSv/a (primarily through the use of ionized rays and radioactive materials in medicine), the total exposure is some 3.9 mSv/a (mean value).

In 1996 17.8 mill. tonnes of ash were produced in coal-firing, in the form of boiler slag, bottom ash and fly ash in Germany. 7.5 mill. tonnes result from the burning of bituminous coal and 10.3 mill. tonnes from lignite (table 2). 98 % of the bituminous coal ash is utilized in construction and in mining industry. The majority of lignite ash is used for the filling and recultivation of open pit mines. It is utilized either unmixed or mixed with FGD gypsum and/or FGD water.

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Table 1. The mean effective radiation dose per year from natural radiation exposure to the public in the Federal Republic of Germany in 1994 according to [6]

Source	Mean effective dose in mSv/a
Cosmic radiation	0.3
Terrestrial radiation	
- Outdoors (5h/d)	0.1
- Indoors (19h/d)	0.3
Inhalation of radon decay products	
- Outdoors (5h/d)	0.2
- Indoors (19h/d)	1.2
Incorporated natural radioactive materials	0.3
Total natural radiation exposure	2.4

Table 2. Production and utilization of by-products from power stations in Germany in 1996

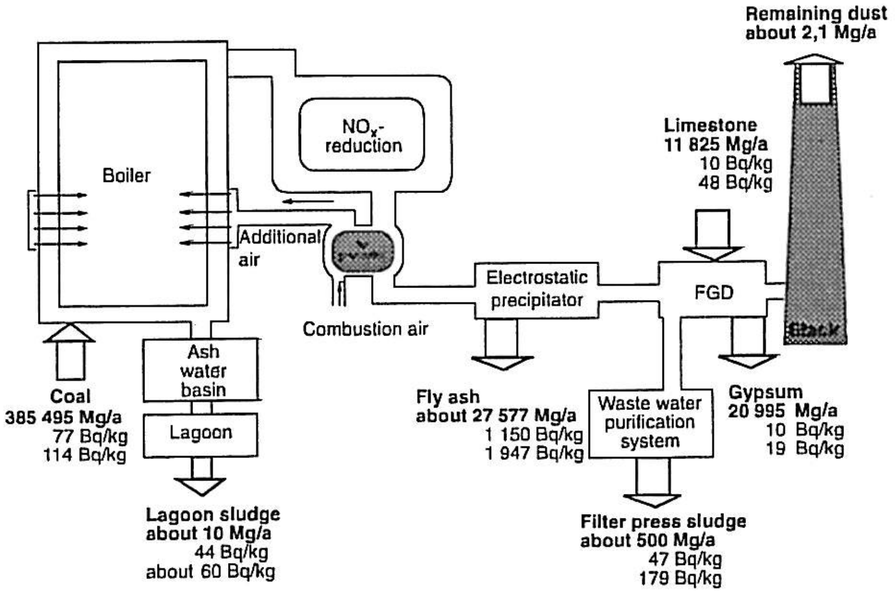
	By-product	Production mill. tonnes	Utilization mill. tonnes
Bituminous coal-fired power stations	Boiler slag	2.4	2.4
	Bottom ash	0.6	0.6
	Fly ash	4.1	4.1
	Fluidized bed combustion ash	0.4	0.4
	Subtotal	7.5	7.5
Lignite-fired power stations	Bottom ash	2.0	2.0
	Fly ash	8.0	8.0
	Fluidized bed combustion ash	0.3	0.3
	Subtotal	10.3	10.3
Total		17.8	17.8

Natural radioactive nuclides in power station coal ash

The natural radioactivity of coal, and thus of the ash produced through its firing, mainly results from radionuclides in the decay series of uranium-radium, thorium and uranium-actinium, as well as from potassium-40. The radon inert gas, which is also produced in the decay series, may partly escape through the solid substances into the air. Radon-220 is less important because of its half life period of 56 s. The uranium-actinium series is not dealt with because its content of nuclides is only small when compared with total radiation.

Because of the age of coal, the radionuclides are balanced, i.e. all nuclides in a given decay series have specific activities which, in a balanced position and depending on the ramification, are in a given relation to each other. For this reason, it is general practice to give an account of the activity concentrations of a representative radionuclide for each series, usually radium-226 and thorium-232 for the uranium and the thoron decay series, respectively.

In the firing of the coal, most of the radionuclides remain in the ash. The ash content of bituminous coal used in power stations in Germany is in the range of about 7 to 40 %, the mean being 15 %. Examination of the radioactivity of solid material passing through power stations has shown that more than 90 % of the radioactivity in coal is retained in the ash. Only a small percentage of the radioactivity can be found in flue gas desulphurization products like FGD gypsum (figure 1) [21]. Due to the ash content, the natural radioactive nuclide concentration in fly ash exceeds that in coal by a factor of 2 to 15.



upper value: Thorium series with the following nuclides: Th-232, Ra-228, Ac-228, Th-228, Ra-224, Rn-220, Po-216, Pb-212, Bi-212, Po-212 (64 %), Tl-208 (36 %)

lower value: Uranium series with the following nuclides: U-238, Th-234, Pa-234, U-234, Th-230, Ra-226, Rn-222, Po-218, Pb-214, Bi-214, Po-214

Figure 1. Mass balance and activity flow in a coal fired power plant according to [21]

The radionuclide concentration in an ash is determined by the radionuclide concentration in the coal, the ash content of the coal and the conditions at the power station. Owing to the low activity concentration in German lignite, the lignite coal ash has a very low value too. Its activity concentration is similar to that in natural soil and thus causes no increase in radiation. Therefore it is not further dealt with in this report.

A summary of international measuring results regarding activity concentration in bituminous coal fly ash can be found in table 3. The mean values for radium-226, thorium-232 and potassium-40 are 170, 114 and 652 Bq/kg, respectively.

Table 3. Activity concentration of bituminous coal fly ash (FA), in Bq/kg

Origin	Source	No. of samples	$C_{\text{Radium-226}}$			$C_{\text{Thorium-232}}$			$C_{\text{Potassium-40}}$		
			MV	min	max	MV	min	max	MV	min	max
D	[1]	28	210	80	390	130	60	260	700	330	1110
D	[11]	26	264	85	370	120	44	190	n.i.a.	n.i.a.	n.i.a.
PL	[22]	10	237	70	611	200	89	514	833	385	1776
D	[21]	3	126	93	137	121	96	155	700	459	877
GB	[9]	4	89	72	105	68	53	94	900	800	1250
AUS	[2]	9	90	7	160	150	7	290	220	20	570
diverse	[19]	50	127	75	235	96	42	131	437	205	765
D	[26]	5	172	148	204	118	100	140	955	881	1018
D	[3]	3	122	87	177	88	73	115	n.i.a.	n.i.a.	n.i.a.
D	[8]	20	90	51	165	83	59	120	1039	760	1480
D	[7]	6	224	192	281	106	93	122	827	740	1000
Overall		164	170[1]	7	611	114[1]	7	514	652[1]	20	1776

[1] Weighted mean value
MV : Mean value
n.i.a. : No information available

An evaluation of the values for bituminous coal ash from German power stations (table 4) shows that the mean values for radium-226 and thorium-232 are similar to those in table 3 (186 and 110 Bq/kg, respectively), whereas the mean value for potassium-40 is 785 Bq/kg which is slightly higher than the mean in table 3.

Table 4. Activity concentration of coal fly ash (FA) from German power stations, in Bq/kg

Source	No. of samples	$C_{\text{Radium-226}}$			$C_{\text{Thorium-232}}$			$C_{\text{Potassium-40}}$		
		MV	min	max	MV	min	max	MV	min	max
[1]	28	210	80	390	130	60	260	700	330	1110
[7]	6	224	192	281	106	93	122	827	740	1000
[11]	26	264	85	370	120	44	190	n.i.a.	n.i.a.	n.i.a.
G 1	60	235	80	390	123	44	260	722	330	1110
[21]	3	126	93	137	121	96	155	700	459	877
[26]	5	172	148	204	118	100	140	955	881	1018
[8]	20	90	51	165	83	59	120	1039	760	1480
[3]	3	122	87	177	88	73	115	n.i.a.	n.i.a.	n.i.a.
[23]	8	113	59	182	79	69	105	836	380	1240
G 2	39	111	51	204	90	59	155	881	380	1480
Overall	99	186	51	390	110	44	260	785	330	1480

MV : Mean value
n.i.a. : No information available
G 1: Total evaluation of measurings before 1980
G 2: Total evaluation of measurings after 1980

An interesting point to note is that the results in the later analyses [3,8,21,23,26] differ from those in the earlier ones [1,7,11]. The earlier investigations focused on the emission of radioactivity from coal-fired power stations through the chimney. To determine the emission, either so-called "clean gas ash" discharged with the flue gas, or fly ash in the last stage of the electrostatic precipitator were examined. In this stage, the ash is of great fineness and comparable to clean gas ash. It was already found out that the radioactivity of coarser ash, like that in the first stages of the electrostatic precipitator were about 98% of fly ash is sampled, is lower than that of the fine ash in the last stage. The later measurements, however, concentrated on the evaluation of the total ash collected in the electrostatic precipitator, just as it is utilized. These measurements then resulted in clearly lower values for radium-226 and thorium-232 whereas the value for potassium-40 was slightly higher.

The mean values in table 3, determined from all of the values measured, therefore display a safe-sided approximation. The overall mean values and their band-widths have been proven reliable through several independent studies.

The results shown in table 5 for <u>boiler slag</u> display slightly lower mean values.

Table 5. Activity concentration of boiler slag (BS), in Bq/kg

Source	No. of samples	$C_{Radium\text{-}226}$			$C_{Thorium\text{-}232}$			$C_{Potassium\text{-}40}$		
		MV	min	max	MV	min	max	MV	min	max
[3]	24	138	68	245	93	59	162	835	441	1240
[7]	3	195	181	211	93	85	104	765	666	851
[4]	4	166	144	207	136	107	170	512	337	677
[23]	2	130	121	140	119	76	162	910	660	1160
Overall	33	146	68	245	100	76	170	794	337	1240

MV: Mean value

In table 6, the analyses on <u>bottom ash</u> are displayed. In the tested cases, the activity concentration was lower than that of the respective fly ash.

Table 6. Activity concentration of bottom ash (BA), in Bq/kg

Source	No. of samples	$C_{Radium\text{-}226}$			$C_{Thorium\text{-}232}$			$C_{Potassium\text{-}40}$		
		MV	min	max	MV	min	max	MV	min	max
[23]	2	70	70	70	40	40	40	355	340	370
[28]	n.i.a.	108	46	166	79	25	120	514	196	742

n.i.a.: No information available
MV: Mean value

Besides the direct radiation of the radionuclides, the radioactive gases radon-222 and radon-220 (also called thoron), as well as their short-lived daughters, contribute to the radiation. In order to assess the radioactivity of a material, not only the activity concentration must be considered, but also the exhalation of the radon and thoron stemming from the material itself.

As a result of their production at high temperatures ranging between 1200 and 1700°C, the ashes from furnaces burning pulverized coal have a glassy structure (figure 3). This causes the ashes to have a relatively low emanation rate in comparison with other materials [13]. Emanation is understood to be the release of radon atoms from the solid matter into the pores of a material. Exhalation is the escape of the radionuclides out of the building material into the air by means of diffusion. The quantity of emanated radon atoms which actually are released into the air depends on the structure of the pore system in the material (figure 4). Therefore, in order to know the radiation contribution of a material, the content of radionuclides as well as the exhalation rate must be known. The exhalation rates of particular materials with and without bituminous coal ash will be dealt with later.

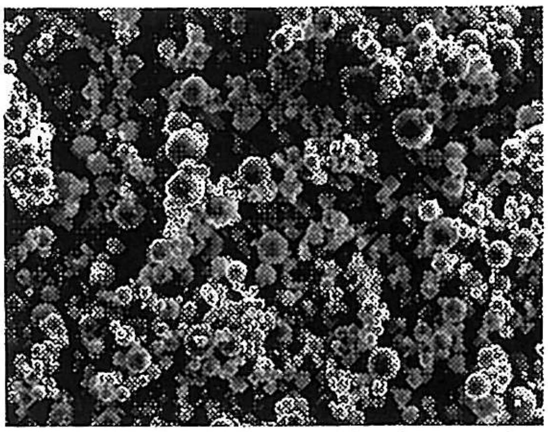


Figure 2. Glassy structure of bituminous coal fly ash (SEM, 2000x)

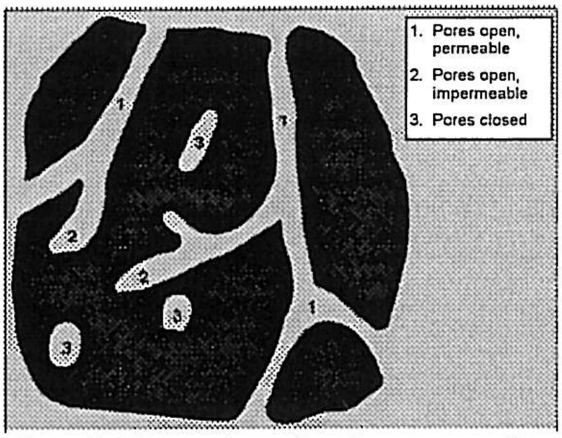


Figure 3. Different formation of pores according to [10]

The question arises, where do the above listed activity concentrations range compared to the limits of the Council Directive 96/26 EURATOM [32] setting the basic safety standards for the protections of the health of workers and the general public against the dangers arising from ionizing radiation, dated May 13, 1996.

First, Articles 40 and 41 dealing with considerably higher expositions from natural radiation sources are to be taken into consideration. Thus Article 40, Para. 2, stipulates that it must be ensured by member countries that those operations that are possibly concerned will be determined on the basis of investigations or other suitable measures. This includes operations resulting in residues that are normally not considered radioactive however contain natural radionuclides that increase the exposition of individual persons of the general public and possibly of the workers considerably. Article 41 describes the protection from exposition from terrestrial natural radiation sources and states in detail which parts of the Council Directive are in this connection to be taken into consideration. Under Section "Declaration and Approval of Activities" radioactive material that is exempt from the declaration is listed in Article 3.2 b, provided its activity concentration per mass unit does not exceed the exemption limits of column 3, Table A, Appendix 1 or - given extraordinary circumstances in a single member country - radioactive material that has deviating values which are approved by the authorities and which do not exceed the general basic criteria of Appendix 1.

If the stipulations of Appendix 1 are applied to the material under consideration, the figures as shown in the table 7 result. The sum of the ratios for each nuclide of the total amount present divided by the values listed in table A of [32] (10 Bq/g for radium-226, 1 Bq/g for thorium-232 and 100 Bq/g for K-40) should be less than 1 (Table 7, column 6). The ashes from coal-fired power plants do in no case exceed the exemption limits of the Council Directive.

Table 7. Radiological description of by-products from coal-fired power plants

Product[4]	Activity concentration			Exhalation rate meas.	Comparative value[5]	Radiation dose in mSv/a Calculated according to table 11[1]		
	^{226}Ra Bq/kg	^{232}Th Bq/kg	^{40}K Bq/kg	Φ Radon-222 Bq/m^{2}h	-	γ-Radiation mSv/a	Inhalation (2) mSv/a	Total (1)+(2) mSv/a
1	2	3	4	5	6	7	8	9
FA 3	92	69	990	0,002	0,088	0,230	0,00016	0,23
FA 4	182	87	520	0,110	0,110	0,375	0,009	0,38
FA 5	140	70	670	0,500	0,091	0,294	0,039	0,33
FA (3)	170	114	652	n.d.[3]	0,138	-	-	-
G1 (4)	235	123	722	n.d.	0,154	-	-	-
G2 (4)	111	90	881	n.d.	0,110	-	-	-
FA (4)	186	110	785	n.d.	0,136	-	-	-
FA (4)	390	260	1480	n.d.	0,314	-	-	-
BA (8)	108	79	514	n.d.	0,095	-	-	-
BA	70	40	370	< d.l.[2]	0,051	0,091	< d.l.	0,16
BS (8)	146	100	794	n.d.	0,123	-	-	-
BS	83	64	1000	1,440	0,082	0,220	0,112	0,33

[1] It gives no sense to evaluate the radiation dose if the exhalation-rate is not measured

[2] d.l.: detection limit

[3] n.d.: not determined

[4] The number in brackets refer to tables in this paper

[5] According to article 3 of the Council Directive a declaration or approval of an operation is not requested if a comparative value calculated according to appendix 1, clause 7 of the directive does not exceed a limit of 1.

According to [31], handling of such material meeting the exemption limits of the Council Directive will thus also in future not be liable to declaration or approval, because thorough investigations have shown that handling of such material leads, even in the most unfavourable case, to actual doses below 1 mSv per annum as specified in article 13 for individual persons of the public and in article 18 for workers. The radiation doses calculated according to table 11 are shown in table 7 (last three columns). The values are far below the limits layed down in the Council Directive.

Exposure of personnel

In the power station

Inhalation and ingestion of dust particles as well as direct radiation are the potential ways in which exposure may occur while handling ash. The handling of ash, e.g. transportation, storage and reloading, is determined by the physical structure of the ash. For example, boiler slag and bottom ash settle at the bottom of the furnace. These deposits are cooled in a water bath, then using a scraper, removed from the water bath and either collected in containers or stored loosely in specially designed storage areas in the open. Because of the low concentrations of fine particle dust, boiler slag is practically dust-free. Bottom ash consisting of sintered fly ash particles also produces very little dust due to its high water-binding capacity.

Dust-forming fly ash is most often forwarded pneumatically from the electrostatic precipitator through a closed system into a storage silo. Thus, in general, there is no direct contact between the personnel and the ashes in the transport-, storage- and loading facilities. Silos are emptied either

- through a mixing screw, with a 10 to 15 % addition of water, onto a truck or a conveyor belt, or
- through flushing pipes in suspension with water, or
- in dried form through a closed system into a silo-truck.

Exhaust- or filtering systems prevent the release of dust into the open. A considerable build-up of dust is only possible during system disruptances.
Radiation exposure is almost totally eliminated through the shielding effect of the walls of the forwarding mechanisms and silos. Direct contact of personnel with power station ash is restricted to repair works, i.e. to short intervals of time. For reasons of general health protection (exposure to dust), filtering masks and safety clothing must be worn when the occasionally necessary repair and inspection work is conducted in the precipitator or in the silos. Thus, ingestion and inhalation are almost eliminated.

Handling of ash from coal combustion in power stations does not lead to any significant increase in radiation exposure.

At the disposal site

Compared to some other industrial nations, very little of the ash from bituminous coal power stations is disposed in Germany. Only 75.000 tonnes, i.e. 1 %, of the bituminous coal ash produced in 1996 were disposed including 37.000 tonnes of fly ash. On account of its dusty, unfavourable form, the primary focus of the following examination will be on the fly ash mono-disposal site model. The fly ash is either transported to the disposal site by truck or by conveyor belt, after having been conditioned with approximately 10 to 15 % water in a mixing screw, or it is pumped by pipeline in suspension with water into a lagoon. In the former case the conditioned fly ash is unloaded and compacted by a bulldozer or other suitable appliance. If drying occurs at the surface, a dust build-up can be avoided through dampening or application of blanketing layers. Larger developments of dust can thus be excluded. The continuous presence of personnel at the unloading of dampened fly ash is not necessary. If the fly ash is being pumped as a suspension into a lagoon, the personnel is needed in the disposal area merely for conversion work. Therefore, the radiation exposure to those working at the disposal site is negligible.

The mean effective dose equivalent in Germany caused by terrestrial radiation has been measured to be on average 0.4 mSv/a [6]. Measurements at German disposal sites [3] displayed a local radiation of 0.14 µG/h, whereby a 2000 h/a stay outdoors would produce an effective dose of 0.28 mSv/a. This effective dose at a fly ash disposal site is within the scattering range of the natural terrestrial radiation too.

The measured radon concentrations at an open fly ash disposal site in UK and those in its direct vicinity, windward as well as leeward, have shown no significant difference [9]. Given a measured radon concentration of 4 Bq/m^3 and a stay of 2000 h, a disposal site worker would be exposed to an effective dose of 0.06 mSv/a.

Radiation exposure to the public through building materials containing power plant residues

General

In Germany, 98 % of the residues resulting from the firing of bituminous coal is utilized in civil engineering and in mining.

With respect to radiation exposure to the public, the cases of special consideration are those in which the ash is utilized in the manufacturing of building materials used for the building of dwellings. The most important cases in Germany are:

- the implementation of fly ash as a concrete addition or cement additive,
- the manufacturing of masonry blocks using fly ash, bottom ash and/or boiler slag as components.

Radiation exposure to the public through living in dwellings

The mean effective dose equivalent in Germany caused by living in dwellings has been measured to be 1.5 mSv/a [6]. This exposure level is a result of radon, thoron, their short-lived daughter nuclides and direct radiation of building materials. An average duration of stay of 19 h/d was used in the calculation of these radiation doses. With more than 80 % (1.2 mSv/a) of the radiation exposure in dwellings being caused by radon-222 (radon) and radon-220 (thoron) and their short-lived daughters, only 0.3 mSv/a comes from direct radiation.

It is known from systematic research that the mean value of the radon concentration in the air in Germany, in dwellings and in open air, is 50 Bq/m^3 and 14 Bq/m^3, respectively. In an extensive examination of more than 6000 dwellings, the radon concentration varied between 0 and more than 10000 Bq/m^3 [6]. According to a statement from the German Commission for Radiation Protection, no measures are necessary for radon concentrations less than 250 Bq/m^3 (normal range) [27]. If the concentrations are higher, the situation should be examined to determine if restoration measures with affordable expenditure are possible. The ICRP (International Commission for Radiation Protection) recommends that for concentrations higher than 200 to 400 Bq/m^3 the proper measures ought to be taken to reduce the radon concentration. Radiation sources are radon from the soil as well as the natural radiation of the building materials.

The entire building material in a house contributes an approximated 30 Bq/m^3 to the radon concentration [13], with higher concentrations originating in the soil [5,27]. Therefore, according to *Keller* [13], an examination of building materials is not necessary, since the building materials produce a relatively low radioactive exposition when compared to other sources. On the other hand, it is necessary to examine the radioactive properties of new building materials in order to prevent an increase in radioactive exposition.

In the past, various equations have been suggested for an approximation of the contribution of building material to the radioactive exposition in dwellings, whereby varying assumptions and maximum acceptable doses were given. Examples of these are: the *Krisiuk* equation [17] (often referred to as the Leningrad Equation) where only direct radiation is considered, the Austrian pre-standard S 5200 equation [25] and the recommendations from *Keller et al* [12,16] in which radon is considered in various ways. In all of these assessments, the actual concentration of radon in the air is approximated in a lump sum manner. This leads to false estimations when the actual physical characteristics differ greatly from the accepted parameters. Table 8 represents the differences between measured and calculated radon concentrations.

Besides radon-producing radionuclides in the building material, the emanation and the exhalation rate are of considerable importance with respect to the release of radon into the air. While the radionuclide concentration in composed building materials can be calculated from the radionuclide concentration of their components, such calculation cannot be used to determine the exhalation rate, since this is determined by the structure of the building material. As the radiation contribution of radon and its short-lived daughters from building materials is high compared to the quantity coming from direct radiation, the knowledge of the exhalation rate is of special importance for the assessment of radiation of building materials. Figure 4 shows a measuring device for the radon exhalation rate [33].

Table 8. Comparision of measured and calculated radon concentrations

Sample	C (Rn)^{mea} in Bq/m^3 [1]	C (Rn)^{cal} in Bq/m^3 [2]	C (Rn)^{cal} / C (Rn)^{mea}
Bottom ash	≤ 0,001	68,71	≥ 68710
Boiler slag	7,27	34,73	4,8
Fly ash 3	0,55	52,85	96,1
Fly ash 4	0,01	26,42	2642
Concrete 9	0,36	1,70	4,7
Concrete 10	1,27	4,53	3,6
Concrete 11	2,18	7,36	3,4
Concrete 12	1,45	18,12	12,5
Concrete 13	1,82	19,25	10,6
Concrete 14	2,55	19,25	7,5

[1] mea: measured value

[2] cal: calculated value based on the activity concentration

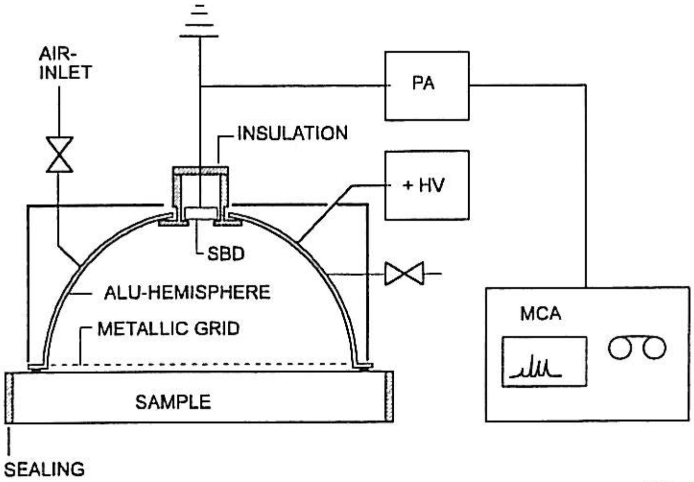


Figure 4. Measuring device for the alpha spectroscopic-determination of ^{222}Rn and ^{220}Rn-exhalation-rates. (SBD : surface barrier detector, PA : preamplifier, MCA : multi-channel analyzer) [33]

Radioactivity from Concrete

For reasons of concrete technology, the portion of fly ash used as a concrete addition normally ranges between 40 and 120 kg/m^3 of concrete. The measurements made on concrete in Germany and in other countries [15,20,24,30] have uniformly shown that the exhalation rates of concrete with fly ash addition, despite its higher activity concentration, increased negligibly or not at all. This is regarded to be due to the glassy structure of bituminous coal fly ash (figure 2) as well as to the alteration in the system of pores in the concrete (decrease of mean pore diameter). The compositions of some concretes are listed in table 9. Table 10 shows that, despite higher activity concentrations, sometimes the radon-222 exhalation rate was lower in concrete with fly ash addition when compared to concrete containing Ordinary Portland Cement only.

Table 9. Composition of concretes dealt with in table 10

	C 1 [30]	C 2 [30]	C 3 [30]	C 4 [30]	C 5 [30]	C 6 [30]	C 7 [30]
CEM I 32,5 R	360	320	360	270	200	320	-
CEM III / A 32,5 R	-	-	-	-	-	-	360
FA 1	-	80	90	67,5	100	-	-
FA 2	-	-	-	-	-	80	-
FA 3	-	-	-	-	-	-	-
FA 4	-	-	-	-	-	-	-
Boiler slag	-	-	-	-	-	-	-
Aggregate	1817	1777	1754	1814	1856	1771	1807
water	180	180	162	182	180	180	180
w/ (c+f)	0,5	0,5	0,4	0,6	0,72	0,5	0,5

	C 8 [30]	C 9	C 10	C 11	C 12	C 13	C 14
CEM I 32,5 R	-	240	240	240	240	240	240
CEM III / A 32,5 R	320	-	-	-	-	-	-
FA 1	-	-	-	-	-	-	-
FA 2	80	-	-	-	-	-	-
FA 3	-	-	120	-	-	120	-
FA 4	-	-	-	120	-	-	120
Boiler slag	-	-	-	-	625	625	625
Aggregate	1763	1914	1782	1782	1257	1126	1126
water	180	180	180	180	180	180	180
w/ (c+f)	0,5	0,75	0,5	0,5	0,75	0,5	0,5

The radiation dose of building material can be calculated if the radionuclide concentrations and exhalation rates are known. The equations used for calculation of the radiation dose in table 11 are taken from [15,29]. As the contribution of radon 220 to inhalation is very low, the exhalation of radon 220 was not measured and not included into the calculation of the radiation dose in column 8 of table 10. It can be seen that all concretes deliver small amounts of radiation (table 10). With values ranging between 0.01 and 0.27 mSv/a all concretes lay far under the mean radiation dose measured in dwellings. In conclusion, it can be said that concrete made from the common basic components delivers only a small contribution of radiation when compared with other building materials (see [5]). The addition of fly ash to concrete only slightly increases the radiation dose of concrete. The radiation doses are considerably below the limits of the Council Directive.

Table 10. Radiological description of concretes

	Activity concentration			Exhalation rate meas.	Comparative value[1]	Radiation dose in mSv/a Calculated according to table 11		
	^{226}Ra	^{232}Th	^{40}K	Φ Radon-222		γ-Radiation (1)	Inhalation (2)	Total (1)+(2)
	Bq/kg	Bq/kg	Bq/kg	Bq/m²h	-	mSv/a	mSv/a	mSv/a
1	2	3	4	5	6	7	8	9
C 1	15	17	238	2,87	0,021	0,046	0,224	0,27
C 2	16	19	270	0,78	0,023	0,051	0,061	0,11
C 3	17	19	275	0,65	0,023	0,052	0,051	0,10
C 4	16	18	265	1,50	0,022	0,051	0,117	0,17
C 5	17	19	278	0,72	0,023	0,054	0,056	0,11
C 6	19	19	263	0,60	0,024	0,055	0,047	0,10
C 7	19	19	230	1,19	0,023	0,056	0,093	0,15
C 8	23	21	256	0,96	0,026	0,064	0,075	0,14
C 9	6	3	125	0,08	0,005	0,013	0,006	0,01
C 10	7	8	130	0,25	0,010	0,022	0,020	0,04
C 11	10	13	130	0,44	0,015	0,034	0,035	0,07
C 12	26	32	370	0,29	0,038	0,087	0,023	0,11
C 13	21	34	330	0,36	0,039	0,081	0,028	0,11
C 14	29	34	405	0,50	0,041	0,092	0,039	0,13

[1] According to article 3 of the Council Directive a declaration or approval of an operation is not requested if a comparative value calculated according to appendix 1, clause 7 of the directive does not exceed a limit of 1.

Table 11a. The components of the radiation dose

Radiation dose

H_{eff} = γ - Radiation dose (1) + Inhalation dose (2)

1. **γ - Radiation dose**

$H\ (\text{Radium-226}) = 1.3 \cdot 10^{-3} \cdot C_{\text{Radium-226}}$
$H\ (\text{Thorium-232}) = 1.6 \cdot 10^{-3} \cdot C_{\text{Thorium-232}}$

2. **Inhalation dose**

$H\ (\text{Radon-222}) = De_1 \cdot F_1 \cdot S \cdot v^{-1} \cdot \Phi\ \text{Radon-222}$
$H\ (\text{Radon-220}) = De_2 \cdot F_2 \cdot S \cdot \lambda_2^{-1} \cdot \Phi\ \text{Radon-220}$

with

$De_{1,2}$	dose conversion factor (indoors) H/C_{eq}, in $mSv/(Bq \cdot m^3)$
$F_{1,2}$	equilibrium factor
S	surface volume ratio in m^{-1}
$\lambda_{1,2}$	decay constant in h^{-1}
v	ventilation rate in h^{-1}

Table 11b. Parameters used for calculation in table 11a

Parameter	Dimension	Radon-222	Radon-220
De	mSv/(Bq $\cdot$ m^3)	0,061	0,29
F	-	0,3	0,05
S	m^{-1}	1,7	1,7
λ	h^{-1}	0,00755	45,36
v	h^{-1}	0,4	0,4

In the aforementioned reflections it has not yet been taken into account that concrete in buildings is usually coated with floor pavement, plaster, wallpaper, paint or other coverings additionally reducing material exhalation [14].

Masonry Blocks

Only a few measuring results are available for radionuclide content and exhalation rate from masonry blocks containing ash from coal fired power stations. Calculations of *Green* [9] lead to the conclusion that the increase of activity concentration due to ashes is at least partly compensated by the small exhalation rate of the ashes. Therefore masonry blocks with coal ash do not significantly increase the radiation exposure compared to conventional masonry blocks. Table 12 gives some values for masonry blocks with by-products from coal fired power plants. They are below the limits.

Table 12. Radiological description of some masonry blocks

	Activity concentration			Exhalation rate meas.	Comparative value[1]	Radiation dose in mSv/a Calculated according to table 11		
	^{226}Ra Bq/kg	^{232}Th Bq/kg	^{40}K Bq/kg	Φ Radon-222 Bq/m^{2}h	-	γ-Radiation mSv/a	Inhalation (2) mSv/a	Total (1)+(2) mSv/a
1	2	3	4	5	6	7	8	9
Brick	29	31	487	< d.l.[2]	0,039	0,087	<d.l.[2]	0,09
Hollow block	78	63	591	0,216	0,077	0,202	0,017	0,22
Sand-Lime block	81	62	859	0,252	0,079	0,202	0,019	0,22

[1] According to article 3 of the Council Directive a declaration or approval of an operation is not requested if a comparative value calculated according to appendix 1, clause 7 of the directive does not exceed a limit of 1.

[2] d.l.: detection limit

Summary

In 1996 18 mill. tonnes of ash were produced by coal firing in power stations in Germany. The ashes are utilized to a large share in building and mining industry. At firing, the radionuclides mostly remain in the ash. In Germany, radionuclide concentration in lignite ashes is in the range of natural soils due to the very low radionuclide concentration of the coal. Ashes from bituminous coal contain radionuclides in the same amount as natural rocks.

The exposure of workers handling coal ash within the power station and at the disposal site is only insignificantly increased compared to natural radiation. There is no significant additional exposure of the public from ash disposal sites either.

The use of ash in building materials results in a negligible increase of radiation dose from living in dwellings.

All materials mentioned in this paper are considerably below the limits given by the Council Directive [32].

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