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IOP Publishing J. Radiol. Prot. 43 (2023) 021502 https://doi.org/10.1088/1361-6498/acc596

Journal of Radiological Protection

OPEN ACCESS

RECEIVED
6 February 2023

REVISED
16 March 2023

ACCEPTED FOR PUBLICATION
20 March 2023

PUBLISHED
6 April 2023

PAPER

Radioactivity of residues from waste incineration facilities in Finland

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Keywords: radioactivity, waste incineration, municipal solid waste, fly ash, bottom ash, bottom slag

Abstract

Waste incineration in Europe has been increasing in the past few decades as there is a need to reduce the burden on landfills and their associated environmental concerns. While incineration reduces the volume of the waste, the volume of slag and ash is still substantial. To find out potential radiation risks that incineration residues could set to workers or the public, the levels of radioactive elements in these residues were investigated from nine waste incineration plants in Finland. Natural and artificial radionuclides were detected in the residues, but in general the activity concentrations were low. This study shows that the level of Cs-137 in the fly ash from municipal waste incineration follows the pattern of 1986 fallout zones in Finland, although the levels are significantly lower than in ash from bioenergy production from the same areas. Am-241 was also detected in many samples, although the activity concentrations were very low. Based on the findings in this study, the typical ash and slag residues from municipal waste incineration do not need radiation protection measures for workers or the public even in regions that received up to 80 kBq m^{-2} of Cs-137 fallout in 1986. The further use of these residues need not be restricted due to radioactivity. Hazardous waste incineration residues and other special cases need to be considered separately, depending on the original waste composition.

1. Introduction and background

The purpose of this study was to characterise the radioactivity of typical waste incineration residues in Finland in order to examine if there is a need for protective measures or restrictions due to radioactivity in the recycling or utilisation of the residues. Due to the global need to advance sustainable development and the circular economy, and mitigate the effects of climate change, measures are needed to reduce waste, increase reuse, recycling and utilisation of residues, and reduce the burning of fossil fuels (e.g. United Nations 2030 Agenda for Sustainable Development [1], the European Green Deal [2] and the European Circular Economy Action Plan [3]). One result of these efforts is that the incineration of municipal waste in Europe has doubled from 1995 to 2019 [4], although the long-term goal in Europe is the prevention and reduction of waste [3] rather than increasing waste incineration further. Previously there have been only a few studies of the radioactivity of waste incineration residues internationally [5–8], and no studies in Finland. Due to the levels of heavy metals in the residues [9, 10] the beneficial use of waste incineration residues can be limited, but as advanced forms of reuse and recycling are developed, such as more efficient metals recovery [11], it is also important to investigate and document the levels of radionuclides in the residues. For this purpose, in this study the residues from nine waste incineration plants in Finland were characterised for natural and artificial radionuclides.

1.1. Waste incineration

The composition of incinerated waste and the technology of the incineration plant are the main factors affecting the solid residue types and their composition [12, 13]. Municipal solid waste incineration (MSWI) facilities typically produce 160–300 kg of bottom ash and slag per 1000 kg of incinerated waste [13, 14]. Bottom ash and slag consist of a variety of melting products and also non-combustible glass, metal, ceramic, rock material and organic matter [15]. After screening for metals (iron, copper, aluminium and steel), bottom slag may be used as a building material intended for field structures or other earthworks [13, 16] or manufactured construction materials [17], if the heavy metal and soluble salt concentrations are suitable based on environmental standards. The ash residues separated from the flue gases, such as fly ash, air pollution control (APC) residue and boiler ash, are most often classified as hazardous waste [13, 14, 16].

In Finland in 2021 from the total amount of municipal waste (3.3 million tonnes), 2 million tonnes (62%) was utilised in energy recovery and 1.3 million tonnes (38%) was utilised in material recovery. Approximately 1.7 million tonnes of the municipal waste utilised in energy production was mixed waste [18]. In six of the nine studied incineration facilities the fuel was reported to be sorted municipal solid waste (MSW). Municipal waste is defined as waste generated in households, including separately sorted wastes (paper, cardboard, glass, metal, textile, biowaste), and wastes comparable to household waste generated in administrative, service and business activities (e.g. schools, hospitals, offices, stores, accommodation and catering services, industry and primary production), excluding septic tank sludges [19]. It has been estimated that, in 2015, 65% of the municipal waste in Finland originated from households and 35% from comparable waste from administrative, service and business activities [20]. The composition of incinerated MSW is very heterogeneous [19] and the sampling and compilation of reliable data on the composition of municipal waste is challenging. Without reliable data on the composition of the waste it is difficult to investigate the correlation of the quality of incinerated waste with ash or slag quality [13, 19].

MSW can contain radionuclides of natural origin such as U-238, Th-232, Ra-226, Ra-228 and K-40 as well as artificial radionuclides such as Cs-137, I-131 and Am-241. The natural radionuclides and Cs-137 are present in small activity concentrations in, for example, food waste, wood waste, garden waste and brushwood, and waste from wastewater treatment that may end up in incineration plants [21–24]. Also, hazardous waste and some wastes from industry, agriculture, forestry and construction can contain natural radionuclides or Cs-137 and may be incinerated with municipal waste. Artificial radionuclides such as Lu-177 and I-131 can end up in the ash or slag from the incineration of medical wastes originating from hospitals or patients' households, and residues from wastewater treatment [24]. Other artificial radionuclides such as Am-241 may end up in incineration residues from industrial waste which is below exemption level, or municipal waste which is not disposed of appropriately. To our knowledge, only one of the incineration facilities in this study had radiation monitors installed for the detection of radioactivity from incoming waste fluxes.

1.2. Legal background

In Finland, the directive 2013/59/Euratom (EU-BSSD) has been implemented with the Radiation Act 859/2018, and notification and exposure assessment are required if the activity concentrations of natural radionuclides exceed exemption and clearance levels (naturally occurring radioactive material, NORM). Therefore, the general exemption and clearance levels of 1 kBq kg^{-1} for U-238, Th-232 and their progeny and 10 kBq kg^{-1} for K-40 apply also to residues from waste incinerators, even though the Radiation Act does not explicitly mention the operations of waste incineration plants. There are also regulations in place concerning the radioactivity of building materials including natural radionuclides and fallout Cs-137 [25]. Due to the presence of fallout Cs-137 in the Finnish environment [26] originating from the Chernobyl nuclear accident in 1986 and atmospheric nuclear weapons testing in the 1950s and 1960s, the general exemption and clearance level of 0.1 kBq kg^{-1} for Cs-137 from the EU-BSSD is not practical in Finland. For example, almost all ash from bioenergy production in Finland exceeds this level for Cs-137, often by a large margin [27, 28]. In Finland, the exposure from fallout Cs-137 is considered as part of natural radiation and thus fallout Cs-137 is included in the regulations for natural radiation [25]. The reference levels used for natural radiation in Finland are 1 mSv a^{-1} for workers and 0.1 mSv a^{-1} for the public (excluding radon, building materials and background). For the exposure of the public to external gamma radiation from building materials, the reference levels are 1 mSv a^{-1} for indoor exposure in buildings (additionally 0.1 mSv a^{-1} from Cs-137 alone) and 0.1 mSv a^{-1} for outdoor earthworks such as roads, streets and field structures.

Based on evaluations made by the Radiation and Nuclear Safety Authority (STUK), specific exemption from exposure assessment is given to materials (excluding building materials) containing fallout Cs-137 in activity concentrations less than 5 kBq kg^{-1} and 1 kBq kg^{-1} for workers and public, respectively [25]. In EU-BSSD Annex VIII a screening index is used for the indoor gamma radiation emitted by buildings, and this is implemented in the Finnish regulations [25]. This index can also be applied to screening the

constituents of mixed building materials (e.g. mixing ash into concrete), taking into account that for the final mixed material the proportions of the mixture need to be considered in the screening. Public exposure from concrete needs to be assessed separately if the Cs-137 activity concentration in concrete exceeds 50 Bq kg^{-1} [25]. In addition to the screening index of buildings, there are other similarly structured screening tools in the Finnish regulations for the gamma radiation emitted from materials used in the construction of roads, streets and yards, and for other earthworks [25].

2. Materials and methods

Incineration residue samples were collected by the plant operators from nine different waste incineration plants around Finland (table 1, figure 1) and sent to the STUK laboratory. During the time of sampling there was a total of 11 operational waste incineration plants in Finland. In one location there were separate boilers for hazardous waste (plant 8) and municipal waste (plant 9), and these were counted as separate facilities because of the different fuel types (table 1). Six of the studied plants incinerated mainly MSW. Additionally, some of these facilities may also incinerate smaller side streams of wastewater sludge, waste wood, medical waste, industrial waste or hazardous waste, so the waste content can vary also temporally at one facility. Plant 7 was a smaller pilot plant for incineration of wastewater sludge, and plant 1 incinerated solid recovered fuel including waste wood (table 1).

Plant operators were asked to send samples from different ash and slag material batches that are the final residues of the incineration process. Two temporally different samples were requested for each ash and slag sample type: one that would represent a shorter period batch (1 week to 1 month) and another a longer period combined sample (6 months to 1 year). Altogether we received 63 samples of 0.5–1 litre volume. For the purposes of data analysis, APC residue, fly ash and boiler ash samples were grouped together as a fly ash residues group (fly ash, 39 samples), because all these materials are from the flue gases and all these three fractions are not consistently forming or reported from every facility. Respectively, bottom slag and bottom ash were grouped together as a bottom ash residues group (bottom ash, 21 samples) representing the materials remaining at the grate after combustion. Additionally, one facility had a separate gas fired power boiler out of which three residue samples were analysed. The concentrations in these three samples were below or close to the limits of detection for all nuclides and they are not considered further in the results, leaving 60 samples for statistical analysis.

Samples were dried at 105 °C and all concentrations were calculated to dry weight. For gamma-ray measurements, the dried samples were compressed into 100 ml cylindrical containers (STUK 'T-jar'). Containers were packed in airtight bags that were heat sealed under vacuum. Samples were then let to stand for 3 weeks before gamma-ray measurements so that radon progeny would reach secular equilibrium. Some of the samples were also chosen to be additionally measured immediately after sealing, in order to analyse possible short-lived artificial radionuclides in the residues.

Gamma-ray measurements were made at the STUK laboratory with multiple Canberra and Ortec high-purity germanium (HPGe) detectors of the type suitable for low energies (e.g. Canberra BEGe with a carbon epoxy window). These detectors are cooled with liquid nitrogen and data were collected with digital multi-channel analysers such as Ortec DSPEC 502. The laboratory has a low-background environment, and the detectors are housed in low-background lead shielding with 15 cm of lead in most cases. The software used for peak search, peak area determination, baseline and background subtraction, nuclide identification and activity calculations including coincidence corrections was UniSampo–Shaman [29–31]. Efficiency calibrations were made using certified reference sources of 20 mm thickness for one geometry (STUK 100 ml 'T-jar'). Corrections for other sample geometries, sample heights, density and chemical compositions were made using efficiency transfer software Efftran [32] in combination with UniSampo–Shaman and the STUK laboratory information system [33].

In the gamma-ray analysis Pb-210 was determined from the 46.5 keV peak, Cs-137 from the 661.7 keV peak and K-40 from the 1460.8 keV peak. Ra-226 was determined as a weighted average of the radon progeny Bi-214 and Pb-214 (295, 351, 609, 1120 and 1764 keV peaks). Th-232 was determined as a weighted average of Ac-228 and Tl-208 (338, 583, 911 and 2614.5 keV peaks, corrected for branching at Bi-212) since these decay products representing Ra-228 and Th-228 were in or close to equilibrium. U-238 was determined from the 1001 keV peak of Pa-234 m, or the 63 keV peak of Th-234, if Pa-234 m was below the level of minimum detectable concentration (MDC). Additionally, for the subset of samples that were analysed with inductively coupled mass spectrometry (ICP-MS; $N = 27$), the ICP-MS-result for U-238 was selected (see [33] for a comparison of gamma-ray spectrometry with ICP-MS).

U-238 analyses were performed using ICP-MS for a subset of the ash samples ($N = 27$) at the STUK laboratory. Approximately 0.5 g of ash samples were accurately weighed and treated with aqua regia (3:1 HCl:HNO_3) overnight on ice and then digested with the UltraWAVE ECR microwave digestion system

Table 1. Information on the waste incineration plants included in the study, Information is from the annual reports of the plant operators, from a joint website of the environmental administration in Finland.

	Facility 1	Facility 2	Facility 3	Facility 4	Facility 5	Facility 6	Facility 7	Facility 8	Facility 9
Power (MW)	160	36	53	63	66	2 x 58	3	20	55
Technology	Gasification-based	Grate	Grate	Grate	Grate	Grate	Pilot	Grate and rotary kilns	Grate
Incinerated waste mainly (%)	SRF (40%) waste wood (60%)	MSW (97%) industrial (3%)	MSW (99%)	MSW	MSW (91%)	MSW	Wastewater sludge	Hazardous waste (100%)	Municipal (30%–50%), industrial (50%–70%)
Capacity (t a^{-1}) 2021	300 000	100 000	195 000	200 000	180 000	420 000	10 000	75 000	175 000
Utilised waste 2021 (t)	96 000 (SRF); 141 000 (wood)	90 769 (MSW); 2829 (industrial)	153 919	200 236	168 107	1: 189 427 2: 186 732		60 977	152 348
Incineration residues analysed	Bottom ash, fly ash, (gas boiler APC residue)	Slag; bottom ash, APC residue	Boiler ash, APC residue (no slag sample)	Slag, boiler ash, APC residue	Bottom ash, boiler ash, fly ash, APC residue	Bottom ash, fly ash, APC residue	Fly ash, APC residue	Slag, fly ash	Slag, boiler ash, fly ash
Cs-137 fallout zone (figure 1)	IV	IV	I	III	V	II	I	III	III

Notes: APC, air pollution control; MSW, municipal solid waste; SRF, solid recovered fuel.

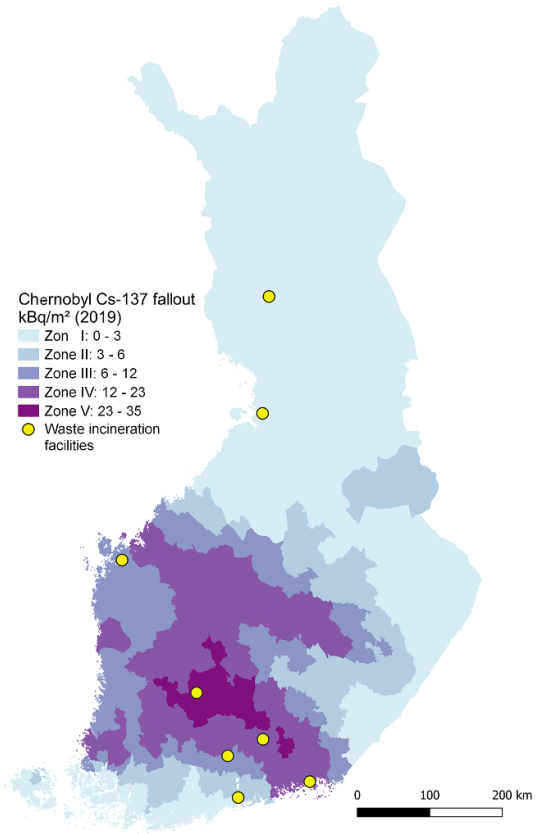

Figure 1. Locations of the waste incineration plants compared with the Cs-137 fallout zones in Finland [26] from the Chernobyl accident in 1986. Activities are shown as 2019 levels, which represents approximately half of the activity compared with that at the time of the fallout in 1986.

(Milestone, Italy) using a temperature of 165 °C for 20 min following 190 °C for 10 min. After digestion the samples were diluted with ultrapure distilled water to 100 ml and filtered to avoid insoluble solids. For ICP-MS analysis the samples obtained from microwave dissolution were further diluted to 1:100 with 1% nitric acid. Analyses of U-238 concentrations were performed by ICP-MS (iCAP Q, Thermo Fisher Scientific, USA) using calibration standards of 0, 1, 10 and 100 μg l^{-1} for U-238 (Romil Ltd, UK) to generate the calibration curve. Bismuth was used as an internal standard to compensate for matrix effects (Romil Ltd, UK).

3. Results and discussion

Statistics for the fly ash and bottom ash groups are shown in table 2. Cs-137, K-40, Ra-226 and Pb-210 were detected in all measured samples, and Th-232 and U-238 in all but one of the samples. Am-241 was detected in 23 out of 60 samples. The artificial nuclides I-131 and Lu-177 were detected in a few samples (see section 3.2). Overall, the median activity concentrations of natural radionuclides and Cs-137 are significantly smaller in both ash groups than the exemption levels in Finland, by a factor varying from 10 to 180. Also the maximum values of all measured radionuclides are smaller than the exemption levels. The median concentrations of Cs-137 and Pb-210 are higher in fly ash (36 and 102 Bq kg^{-1}, respectively) by a factor of 4–5 when compared with bottom ash (7 and 23 Bq kg^{-1}), whereas for Th-232 the median concentration in bottom ash (31 Bq kg^{-1}) is higher than in fly ash (14 Bq kg^{-1}) by a factor of 2.2. For K-40, Ra-226 and U-238 the median concentrations in bottom ash (408, 33 and 35 Bq kg^{-1}) and fly ash (680, 21 and 26 Bq kg^{-1}) are relatively close, being within a factor of approximately 1.5. For most measured nuclides the maximum concentrations are found in the fly ash group (K-40, 1532 Bq kg^{-1}; Pb-210, 801 Bq kg^{-1}; U-238, 318 Bq kg^{-1}; Th-232, 64 Bq kg^{-1}; Cs-137, 152 Bq kg^{-1}; Am-241, 59 Bq kg^{-1}), except for Ra-226 where the maximum concentration (48 Bq kg^{-1}) was found in bottom ash. The measured activity concentrations are shown by facility in table 3.

Activity distributions of Cs-137 and Ra-226 are shown in figures 2 and 3, respectively. Cs-137 is an example of a radionuclide that is concentrated more in the fly ash (figure 2(A)), and a similar pattern is exhibited by Pb-210. The concentration of Cs-137 is variable between individual facilities and spans more

Table 2. Basic statistics of Cs-137, Am-241 and natural radionuclide activity concentrations (Bq kg^{-1} dw) in residues from waste incineration plants in Finland. Activity concentrations of Lu-177 and I-131 are described in the text.

	Activity concentration (Bq kg^{-1} dw)											
Bq kg^{-1} dw	Fly ash group (fly ash + boiler ash + APC residue)						Bottom ash group (bottom ash + slag)					
Nuclide	Mean	sd	Median	Min	Max	*N*	Mean	sd	Median	Min	Max	*N*
Cs-137	45	33	36	0.7	152	39	9.0	14	6.6	1.2	68	21
K-40	700	286	680	226	1532	39	450	158	408	263	1035	21
U-238	38	51	26	10	318	38	36	9.4	35	21	64	21
Ra-226	24	10	21	4.6	46	39	33	8.0	33	21	48	21
Pb-210	125	125	102	18	801	39	27	29	23	6.6	149	21
Th-232	18	15	14	2.5	64	38	30	5.2	31	20	38	21
Am-241	5.3	16	0.8	0.4	59	13	2.3	4.2	0.9	0.6	14	10

Notes: APC, air pollution control; dw, dry weight; *N*, number of samples where nuclide concentration > minimum detectable concentration; sd, standard deviation.

than two orders of magnitude (figure 2(B)). In contrast, there is little difference between fly ash and bottom ash Ra-226 concentrations (figure 3(A)) and there are also no large differences between different facilities for Ra-226 (figure 3(B)).

3.1. Naturally occurring radionuclides and Cs-137

The activity concentrations of natural radionuclides obtained in this study are of the same level as in the other available studies on MSWI residues from the United States, Japan and Germany [5–8]. In the Japanese plant, Cs-137 was also analysed [7] and the median Cs-137 concentration in fly ash was ten times higher in Finland. The difference in Cs-137 arises from the Chernobyl fallout (figure 1) which is discussed in section 3.1.1.

Higher concentrations of Pb-210 and Cs-137 in the fly ash group versus bottom ash are typical of high-temperature combustion processes such as burning of coal or biofuels, reflecting the volatility of lead and caesium at relatively lower temperatures when compared with uranium, thorium and radium. The more volatile components such as polonium, lead and caesium are enriched in the fly ash fractions when compared with the bottom ash residues (e.g. [5, 34–36]). This same feature is visible in the results of this study, although the concentrations of Pb-210 and Cs-137 in MSWI residues (median 107 and 37 Bq kg^{-1} in fly ash) are significantly smaller than in residues from bioenergy production in Finland [27, 37], where the median Pb-210 and Cs-137 concentrations in fly ash were 570 and 1300 Bq kg^{-1}, respectively [27].

3.1.1. Cs-137 and the fallout zones

It is known that Cs-137 is enriched in the bark of, for example, pine, spruce and birch, when compared with the wood itself [21, 38, 39]. Waste incineration plants include wood in some cases, either as industrial waste or other types of wood waste (table 1), and MSW can also include wood. A previous study on ash from bioenergy production in Finland showed that significantly higher Cs-137 concentrations in fly ash were found in the regions with the highest original fallout levels [27]. For waste incineration facilities included in this study, there is no evident increase in Cs-137 concentration in the fly ash with increasing percentage of wood in the fuel (tables 1 and 3), but there is a clear increase of Cs-137 concentration according to the fallout zone of the facility, which is in contrast with nuclides such as Ra-226 (figure 4). The results of Cs-137 for both MSWI fly ash from this study and biofuel fly and grate ash [27] are shown in figure 5. The increasing trend of Cs-137 concentration with the fallout zone is very similar for both fuel types but the median concentration of Cs-137 in biofuel ashes is systematically one order of magnitude larger. This is perhaps understandable considering the difference in the fuels, as in this study the highest percentage of wood in fuel was 60%, and wood that has been used in industrial construction rarely contains significant amounts of bark. In the previous study on ash from bioenergy production in Finland [27], bioenergy facilities burned 70%–100% wood. It was found that increasing the amount of peat in the fuel would reduce the concentration of Cs-137 in the fly ash, but the highest Cs-137 concentrations were found in the facilities that used 100% wood. In bioenergy production it is typical to burn woodchips, which also contain significant amounts of bark. Also, the left-over bark from sawmills is used in bioenergy production. Therefore, a combination of original regional fallout levels and the amount of bark in fuel is suggested as a possible explanation for the different Cs-137 concentration levels in fly ash from waste incineration versus bioenergy production (figure 5). The existence of the trend with fallout zones also for MSWI fly ash suggests that despite the centralisation of production and logistics for many household goods there is still a local component in MSW which is preserving some of the Cs-137 signature of the local fallout zone.

Table 3. Activity concentrations (Bq kg^{-1} dw) of Cs-137, Am-241 and natural radionuclides in waste incineration fly ash and bottom ash residues by facility (facility numbers correspond to table 1). Fly ash group comprises fly ash + boiler ash + air pollution control; bottom ash group comprises bottom ash + slag; *N*, number of samples where nuclide concentration > minimum detectable concentration .

		Activity concentration (Bq kg^{-1} dw)																			
		Facility 1				Facility 2				Facility 3				Facility 4				Facility 5			
Bq kg $^{-1}$ dw	Nuclide	Median	Min	Max	*N*	Median	Min	Max	*N*	Median	Min	Max	*N*	Median	Min	Max	*N*	Median	Min	Max	*N*
Fly ash group	Cs-137	36	25	36	3	72	52	85	5	19	15	22	6	53	37	95	4	96	50	152	4
	K-40	254	226	268	3	857	757	1101	5	625	564	680	6	699	416	1138	4	766	312	1393	4
	U-238	19	13	27	3	22	12	52	5	21	15	22	6	26	10	30	3	30	14	38	4
	Ra-226	21	18	22	3	19	14	46	5	22	16	29	6	22	4.6	33	4	25	17	37	4
	Pb-210	62	48	64	3	112	79	284	5	64	55	101	6	89	18	112	4	93	70	207	4
	Th-232	13	12	13	3	6.3	3.9	35	5	15	7.9	24	6	17	2.6	27	4	4.0	3.2	31	3
	Am-241	0.8	0.7	1.6	3	0.5	0.4	0.5	2	30	0.8	59	2	0.6	0.4	0.8	2	1.2	0.7	1.7	2
Bottom ash group	Cs-137	1.5	1.2	1.6	3	9.4	7.7	68	4					11	8.7	14	3	10	9.0	10	2
	K-40	352	263	408	3	426	399	1035	4					458	373	466	3	308	298	317	2
	U-238	40	29	47	3	40	33	64	4					36	23	47	3	32	29	35	2
	Ra-226	23	21	24	3	39	33	45	4					29	28	31	3	27	24	30	2
	Pb-210	14	12	15	3	29	27	149	4					27	23	29	3	16	13	18	2
	Th-232	22	20	24	3	36	33	37	4					35	31	38	3	27	25	30	2
	Am-241	1.0	0.7	1.2	3	7.4	0.8	14	2					0.7	0.7	0.7	1	0.6	0.6	0.6	1

		Facility 6				Facility 7				Facility 8				Facility 9			
	Nuclide	Median	Min	Max	*N*	Median	Min	Max	*N*	Median	Min	Max	*N*	Median	Min	Max	*N*
Fly ash group	Cs-137	26	21	66	5	0.8	0.7	1.0	3	23	4.9	58	3	57	30	80	6
	K-40	915	517	1532	5	344	304	526	3	719	635	754	3	754	594	880	6
	U-238	17	10	40	5	63	61	83	3	39	12	318	3	37	16	104	6
	Ra-226	20	4.8	28	5	38	32	42	3	17	14	37	3	31	21	37	6
	Pb-210	116	107	801	5	56	41	58	3	132	65	224	3	147	115	243	6
	Th-232	16	2.5	24	5	59	48	64	3	5.0	3.2	16	3	20	10	27	6
	Am-241	1.2	1.2	1.2	1									0.7	0.7	0.7	1
Bottom ash group	Cs-137	3.4	1.9	4.2	3					2.0	1.8	2.4	3	6.6	5.5	10	3
	K-40	385	376	449	3					585	488	609	3	434	402	494	3
	U-238	31	29	35	3					37	21	39	3	35	27	38	3
	Ra-226	29	26	33	3					47	42	48	3	35	33	35	3
	Pb-210	26	20	36	3					9	7	13	3	24	20	28	3
	Th-232	27	24	30	3					34	32	35	3	31	28	33	3
	Am-241	0.9	0.9	0.9	1					2.9	2.9	2.9	1	0.6	0.6	0.6	1

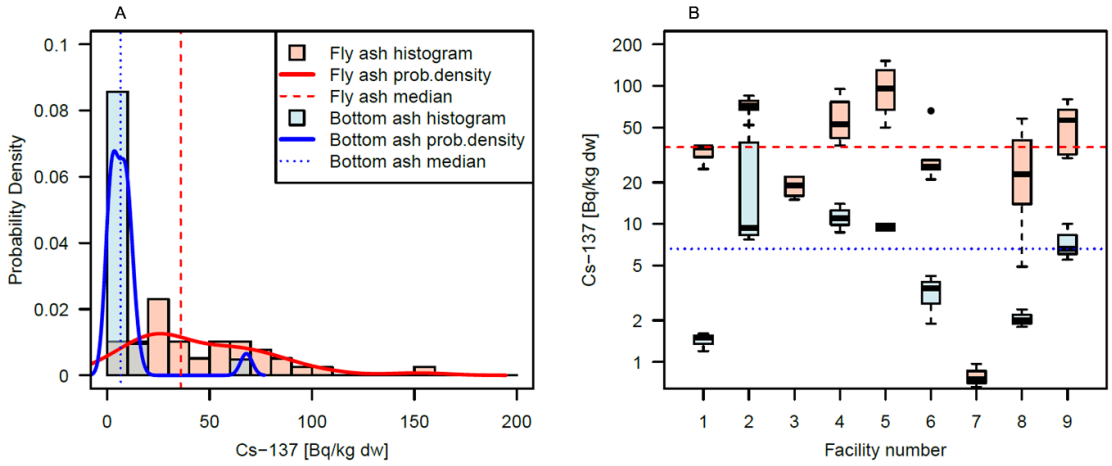

Figure 2. The distribution of Cs-137 activity concentration in the fly ash and bottom ash residue groups from nine waste incineration facilities in Finland shown as a combined dataset (A) and according to each facility (B). The facilities are described in table 1 and the statistics in tables 2 and 3. Boxplot (B) shows median (horizontal black bars), interquartile range (coloured boxes), minimum and maximum (whiskers) and outliers (black dots).

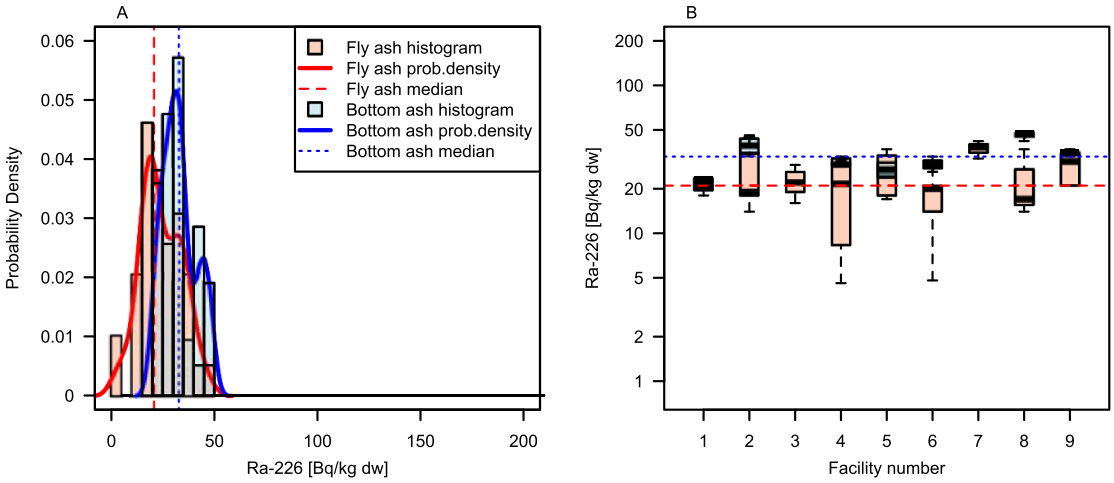

Figure 3. The distribution of Ra-226 activity concentration in the fly ash and bottom ash residue groups from nine waste incineration facilities in Finland shown as a combined dataset (A) and according to each facility (B). The facilities are described in table 1 and the statistics in tables 2 and 3. Boxplot symbols as in figure 2.

3.2. Artificial radionuclides other than Cs-137

Besides Cs-137, other artificial radionuclides, I-131 (3.6–36 Bq kg^{-1}, three samples), Lu-177 (3.6–15 Bq kg^{-1}, two samples) and Am-241 (0.4–60 Bq kg^{-1}, 23 samples) were also detected in the ash samples. I-131 and Lu-177 were detected only in fly ash, but Am-241 was also detected in bottom ash. I-131 and Lu-177 are commonly used in hospitals in radiotherapy treatments of cancer patients and can possibly end up in MSW among hospital waste or waste from patients' households. These nuclides are also continuously detected from sewage sludge that might end up in the incineration facilities in some localities. In Helsinki, STUK regularly monitors the sewage sludge from Viikinmäki wastewater treatment plant as a part of its environmental radiation surveillance programme. The activity concentrations of these nuclides in dried sludge in 2021 were 210–430 Bq kg^{-1} for I-131 and 950–1700 Bq kg^{-1} for Lu-177 [24].

Since Am-241 was detected in almost half of the samples in multiple locations, a mass balance calculation was made to investigate whether it is plausible that the Am-241 found in the waste incineration residues in this study could be from 33 kBq household smoke alarms. These alarms are exempted by the radiation legislation but they should be taken into recycling and not end up in MSW. However, these alarms can end up in municipal waste when not disposed of appropriately. For MSW grate incineration, bottom ash (bottom ash and bottom slag) typically represents 16%–25% by mass of the original waste on a wet basis, and fly ash (boiler ash, fly ash and APC waste combined) 3%–15% [13, 14]. For approximate mass balance calculations mass fractions of 20% and 10% can be used for bottom ash and fly ash groups, respectively. If the amount of

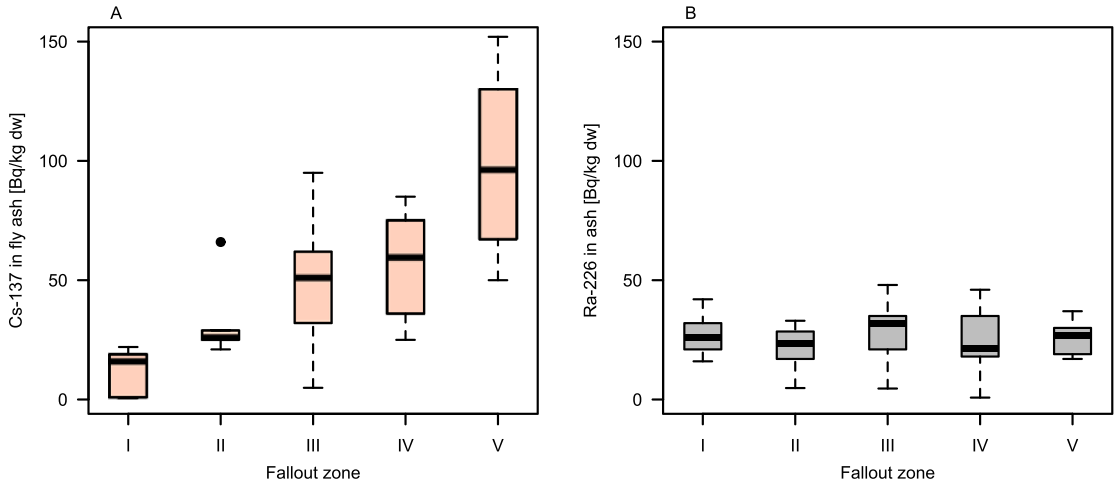

Figure 4. Activity concentrations of Cs-137 in fly ash (A) and Ra-226 in ash (B) from waste incinerations plants as a function of the Chernobyl nuclear accident Cs-137 fallout zones in Finland (figure 1). Ra-226 results from both fly ash and bottom ash groups (table 2) are combined since there was no significant difference in their distributions (figure 3). Cs-137 concentration in fly ash increases as the amount of regional fallout Cs-137 in the environment increases. The trend of Ra-226 (no significant change with fallout zone) is shared with K-40, Pb-210, U-238 and Th-232. Boxplot symbols as in figure 2.

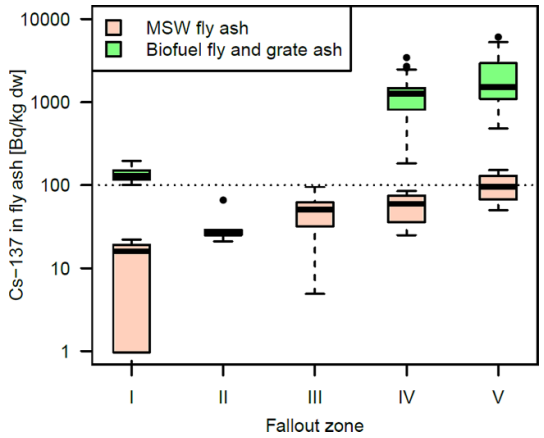

Figure 5. Cs-137 activity concentration in municipal solid waste (MSW) incineration fly ash compared with fly and grate ash from bioenergy production in Finland [27], according to the Chernobyl Cs-137 fallout zones in figure 1. The dotted horizontal line shows the general clearance level of 0.1 kBq kg^{-1} for Cs-137 from directive 2013/59/Euratom (in Finland, exemption from exposure assessment is when fallout Cs-137 is <1 kBq kg^{-1}, excluding building materials). Boxplot symbols as in figure 2.

incinerated waste was 300 000 t a^{-1} in fresh weight (table 1), 60 000 t a^{-1} of bottom ash and 30 000 t a^{-1} of fly ash could be formed. Since the median activity concentration for Am-241 is approximately 1 Bq kg^{-1} for both ash types (table 2), these can be combined to a total of 90 000 t a^{-1} of residues with 1 Bq kg^{-1} of Am-241. Only two samples showed higher concentrations than the average, but they are from 1 week and 1 month samples representing short 'spikes' compared with the long-term level of 1 Bq kg^{-1} (figure 6). To reach 1 Bq kg^{-1} activity concentration in 90 000 t a^{-1} of ash from 33 kBq smoke alarms would require more than 2700 items per year at one MSWI plant. This does not seem plausible since the recycling of appliances has been in place for a long time already. The highest Am-241 value from a 1 week sample (figure 6) could be from, for example, industrial smoke alarms but some other sources are probably also contributing to the general Am-241 level in the residues. For example, waste containing Am-241 from radiation practices can be appropriately disposed of as mixed waste and end up in incineration, if the activity is lower than the exemption level (10 kBq and 1 kBq kg^{-1} Am-241 for <1000 kg of material). It is also known that a small amount of Am-241 can be found in the environment due to the fallout from atmospheric weapons testing [40]. For example, the measurement database of STUK contains results of ash samples from peat combustion. These samples contain 1–3 Bq kg^{-1} of Am-241, when the measurement has been made with a small enough MDC of <1 Bq kg^{-1}. A similar value of a few Bq kg^{-1} can be typically measured in sediments from the 1950s to 1980s [41]. It is unclear how much of this fallout Am-241 could end up in MSW but at least a small contribution is possible from, for example garden waste containing peat or soil. The exact origin

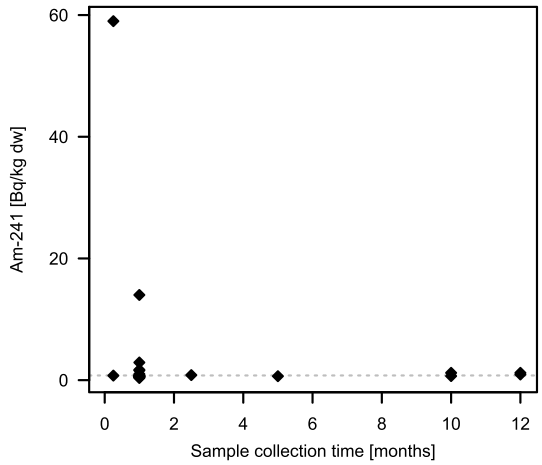

Figure 6. Am-241 activity concentration in residues from waste incineration plants in Finland as a function of the sample collection time. A 12 month sample means that the sample is a composite sample representing the average ash from 1 year. The highest Am-241 concentration is seen in a 1 week sample and the concentrations approach the overall median of 1 Bq kg^{-1} after 1 month collection times.

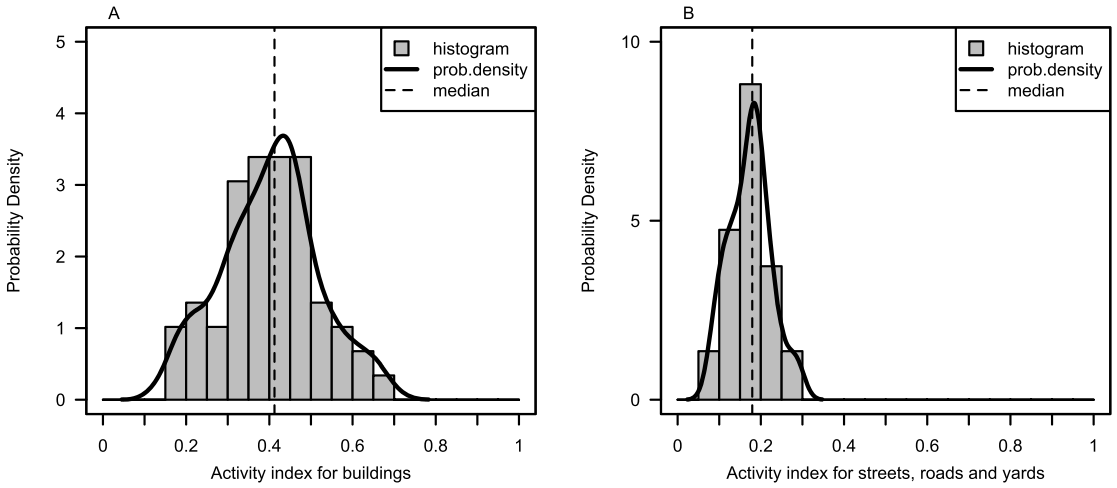

Figure 7. Screening index values for the external gamma radiation emitted by waste incineration residues in Finland if they were used as building materials in the construction of buildings (A), or for roads, streets and yards (B). An index value <1 indicates that material can be used without further exposure assessment. Index formulae can be found in [25].

of the Am-241 in MSWI residues remains unclear but the measured level is of no concern in terms of radiation protection.

3.3. Screening of building materials

For the purposes of assessing radiation risk of waste incineration residues as part of building materials, the screening index values for the ashes of this study were calculated, and the results are shown in figure 7 for building construction and road construction. All the analysed residues have index values <1 which indicates that they could be used as building materials without further exposure assessment. The same is also true from the point of view of Cs-137 in concrete for buildings, as the maximum Cs-137 concentration in residues in this study is 150 Bq kg^{-1}, and in the STUK guidance it is calculated that for typical amounts of ash mixed into concrete, the Cs-137 activity concentration in ash could be up to 300 Bq kg^{-1} before the exposure assessment limit of 50 Bq kg^{-1} in the final concrete would be reached. It is important to remember that this evaluation is only from the point of view of radiation and the waste incineration ashes could contain chemical hazards such as heavy metals which could prevent their use as building materials. However, for the purposes of facilitating waste reduction and the circular economy, it is also important to state that radioactivity is not limiting the potential beneficial use of MSWI residues.

3.4. Effects from incinerated waste type

Nearly all the operational waste incineration plants in Finland were included in this study (9 out of 11). Most of the plants used MSW for fuel, therefore the results can be considered at least nationally representative for MSWI residues. The other fuel types, wood, hazardous waste, industrial waste and wastewater sludge were much less commonly used (table 1). It is still worthwhile to investigate whether there are any compositional differences in the residues which would be correlated to the fuel type. When comparing the reported fuel types with the activity concentrations in ash, the most significant differences arose in the case of wastewater sludge. The ashes from wastewater sludge had the lowest Cs-137 concentrations and highest U-238 and Th-232 concentrations among the studied residues. The mean Cs-137 concentration from wastewater sludge fly ash was 1 Bq kg^{-1} whereas for fly ash from other fuel types it was 50 Bq kg^{-1}. The mean U-238 and Th-232 concentrations were 70 and 60 Bq kg^{-1} for wastewater sludge ashes, and 35 and 20 Bq kg^{-1} for ashes from other fuel types, respectively. Also, there were no detections of Am-241 from the wastewater sludge ashes. However, the number of wastewater sludge ash samples was small ($n = 3$, tables 1 and 3) so not too much significance can be attributed to the non-detection of Am-241, as it could also be a result of the variability of MDCs between the different detectors used. For Ra-226, K-40 and Pb-210, the concentrations in wastewater sludge ash were within the variability of the MSWI residue compositions. No clear correlations were detected in the activity concentrations compared with fuel type for industrial waste, hazardous waste or wood, but the number of samples for these categories was too low for meaningful comparisons.

4. Conclusions

Based on the radiological characterisation of 60 residue samples (bottom slag, bottom ash, boiler ash, fly ash and APC residue) from nine waste incineration facilities in Finland, it is concluded that the contents of natural and artificial radionuclides in the studied waste incineration residues are low from the radiation protection point of view. This is the case even for the geographical areas which received up to 80 kBq m^{-2} of Cs-137 fallout in 1986. Therefore, there are no restrictions to the reuse, recycling or utilisation of these residues originating from radioactivity. However, this conclusion is only generally applicable to incinerated municipal waste, as most of the studied facilities used MSW as fuel, and only restricted materials are meant to be disposed of as municipal waste. In contrast, the composition of hazardous waste can potentially be highly variable depending on the origin of the waste. If hazardous waste containing significant amounts of natural or artificial radionuclides is incinerated, there could be a need for radiation protection measures when handling the residues, as radionuclides are concentrated in the incineration residues compared with the original waste.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgments

This work has been made as part of the Finnish national programme for the monitoring of radioactivity in the environment. The authors acknowledge the internal support of the Radiation and Nuclear Safety Authority—STUK, Finland, which enabled the measurements of samples and the analysis of results. Energiateollisuus ry is acknowledged for the contacts of the waste incineration facilities.

Conflict of interest

The authors declare no conflicts of interest.

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