

Weathering of radiocaesium contamination on urban streets, walls and roofs

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Received 23 January 2001; received in revised form 24 September 2001; accepted 24 September 2001

Abstract

Recent investigations in Russia have emphasised the significance of dose contributions from contamination on urban streets and roof pavings, and, typically to a lesser extent, walls in the urban environment. The crucial factor determining the magnitude of these contributions is the retention of the contamination by the different types of urban surface. Since the Chernobyl accident, a series of long-term field studies has been carried out on urban streets, walls and roofs, to examine the weathering processes of ^{137}Cs on the various surface types. The derived time-functions are applied to estimate resultant long-term doses to inhabitants of an urban centre. The paper highlights the effect on caesium retention of surface material characteristics. © 2002 Elsevier Science Ltd. All rights reserved.

Keywords: Urban contamination; Radiocaesium; Chernobyl accident; In situ measurements; Weathering

1. Introduction

In nuclear accident consequence assessment models, such as COSYMA (Goossens & Kelly, 2000), calculations of external dose are often based on the assumption that the radiation field in inhabited areas will in the long run be governed by the radionuclide (essentially ^{137}Cs) deposition to grassed areas or areas of soil. This is in many cases true due to the slow downward migration of the caesium ion in soil. However, in urban centres, where the majority of the population resides, open, unpaved areas will often have limited dimensions, and the contamination on

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other types of surface may greatly influence long-term doses. After the Chernobyl accident, for instance, road surfaces and roof pavings were generally found to have received comparatively high levels of contamination, and could therefore be expected to have importance concerning dose. An analysis of the mobility of radiocaesium on the surfaces in question is essential to adequately estimate these dose contributions. This paper presents measurements made over the years that have passed since the accident, of the decrease in contamination level by weathering of the various contaminated man-made urban surfaces (streets, walls and roofs), and considerations are made in relation to mechanisms responsible for the fixation of caesium. A good understanding of the distribution and degree of fixation of caesium on such surfaces is also highly advantageous in connection with the planning of forced decontamination in urban areas, which may be carried out with great success, even more than a decade after the contamination occurred (Roed et al., 1998).

2. Materials and methods

2.1. Streets

A series of measurements has been made to examine the long-term behaviour of caesium deposited in urban streets. The measurements were made in the city of Gävle, Sweden (population 95,000), which received sufficiently high levels of contamination from the Chernobyl accident (ca. 100–200 kBq m⁻² of ¹³⁷Cs) to enable the use of germanium detector gamma spectrometry for in-situ measurements. However, a few months after the contamination had occurred, measurement times exceeding 1 hour were not unusual for obtaining results with a statistical uncertainty of some 10–20%. The spectrometer system was transported to the area in a mobile laboratory equipped with supporting electronics, electrical power and data handling facilities. In order to view defined areas, the detector was, in agreement with the methodology described by ICRU (1994), collimated with lead bricks assembled on a steel table, with the front of the detector end cap positioned 93 ± 0.5 cm above the ground. The thickness of the lead shielding, weighing 365 kg in total, was not less than 100 mm at any point, ensuring that contributions from extraneous radiation to the detected signal were minimal. However, when measuring surfaces with low levels in an area with adjacent surfaces with much higher levels, these contributions were not always totally insignificant, but were allowed for by subtraction after counting with lead in front of the detector end cap (Sinnaeve & Olast, 1991). Both asphalt and concrete street pavings were examined.

The collimated detector system was calibrated using four sheets of plywood (each measuring 1.5 m by 1.5 m), which had been contaminated uniformly with ¹³⁷Cs in paint. The plywood sheets were first under-coated so as to prevent penetration of the subsequently applied contaminated paint. For a series of calibration measurements, the contaminated plywood sheets were placed on the floor of a radiologically “clean” room, in positions together constituting an area of some 100 m². It was

found that more than 95% of the full energy counts from ^{137}Cs came from within the nearest 25 m² of surface.

2.2. Walls

During the campaigns in the Gävle area, in situ measurements were also made of the contamination levels on walls. The detector and collimator used for these measurements were similar to those described above for the street contamination measurements. However, here the front of the detector end cap was placed at a distance of 115 ± 0.5 cm from and parallel to the wall. The distance from the centre of the end cap to the ground was 93 ± 0.5 cm. Fig. 1 shows the applied lead shielding assembly for measurement on vertical surfaces. Extra lead bricks placed on the surface of the steel table ensured that the full energy count contribution from the usually much more contaminated underlying horizontal surface was negligible.

Here again, the four plywood sheets were applied for detector system calibration, mounted on top of each other on a scaffold tower. This scaffold tower was moved sideways for each of a series of measurements, which together gave information on the gamma signal that would be detected from a large wall. It was found that about 80% of the counts came from an area of only some 4 m², whereas 95% came from the 12 m² closest to the detector.

2.3. Roofs

Since the Chernobyl accident occurred in 1986, a long-term experiment has been conducted at Risø National Laboratory to investigate the weathering effect on Chernobyl fallout contamination levels on various different types of roofing materials. The roof materials examined were a corrugated eternit (natural, fibre-cement and resin slate) paving with a slope of 45°, a corrugated eternit paving with a slope of 30°, a red clay tile paving with a slope of 45°, a concrete tile paving with a slope of 45°, a silicon-treated eternit paving with a slope of 45°, and a silicon-treated eternit paving with a slope of 30°. At various time intervals, the roof pavings were

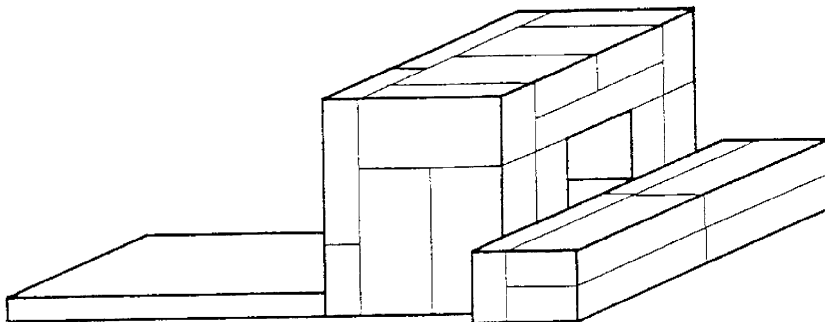


Fig. 1. Lead shielding design for contamination measurements on vertical urban surfaces.

dismounted from the roof for measurement in a gamma spectrometer in a room with low background radioactivity. Here, series of 18 tiles of each type were mounted for measurement on a rack. Tiles from the top, middle and bottom of both the east and west facing parts of the roofs were weighted equally in the evaluation of contamination level.

3. Results

3.1. Streets

The results of in-situ measurements of the decline in the ^{137}Cs contamination level on streets and pavements in the central part of the town of Gävle, Sweden are shown in Fig. 2, relative to the contamination level on a grassed reference surface. Statistical uncertainties on individual measurements ranged between 5 and 20%. The variation between data recorded at the same time is, however, greatly dominated by variations in traffic in different areas of the city. As can be seen, less than 2% of the caesium initially deposited on these surfaces after the Chernobyl accident was left after a decade had passed. Also shown in this figure is a modelling fit according to the expression:

$$C_r(t) = 50\% \cdot (A_1 \cdot \exp(-\lambda_1 \cdot t) + A_2 \cdot \exp(-\lambda_2 \cdot t)),$$

in which $C_r(t)$ is the street contamination level at time t , relative to the contamination

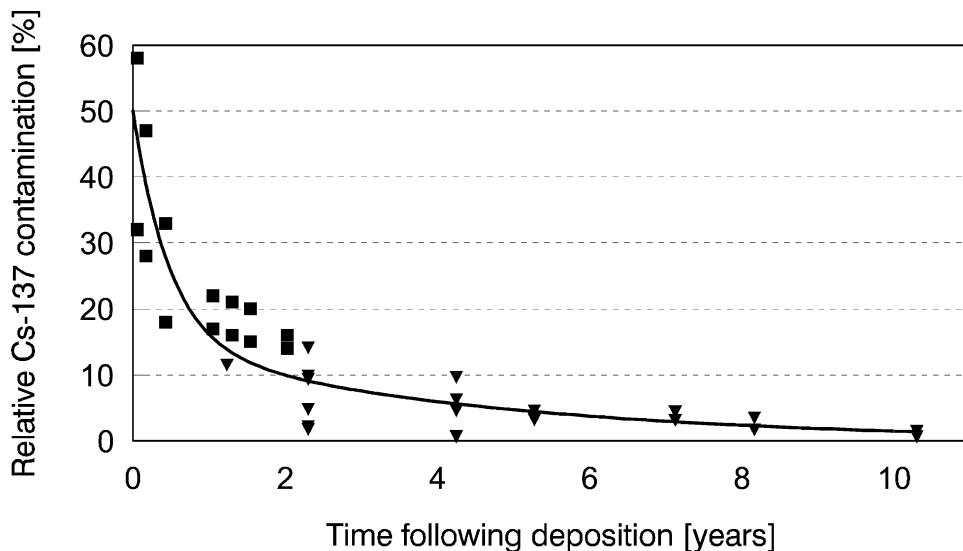


Fig. 2. Weathering of Chernobyl ^{137}Cs on streets and pavements in Gävle. The relative contamination levels are given in percentages of the level on a corresponding reference surface (a cut lawn). Measurements marked with squares were carried out by Karlberg (1988).

level on a grassed reference surface in the area, and A_1 and A_2 are equal to respectively 0.7 and 0.3, whereas λ_1 and λ_2 are rate constants corresponding to half-lives of respectively 120 days and 3 years.

The contamination levels on asphalt surfaces in Gävle are now in most places below the detection limit (ca. 0.2 kBq m⁻², corresponding to about 1% of the contamination level on a lawn). Contamination can now only be registered in areas of relatively little traffic, such as parking lots.

3.2. Walls

The results of the measurements on walls in Gävle are shown in Table 1. As can be seen, the natural decrease in contamination level on walls is very slow compared with that on streets. The recorded contamination levels vary greatly between locations, partly due to different conditions at deposition. For instance, the north-facing yellow clay brick wall was totally shielded against rain during the deposition and thus received only dry deposition. Some samples taken in 1991 from a plastered wall in Gävle at a height of 40 cm showed several times higher levels of both ¹³⁷Cs and ⁴⁰K than did samples taken at heights of 80 or 120 cm. This indicates that soil (or street dust) splash may significantly influence the contamination level on the lower part of the wall. Generally, the half-life of the weathering process appears to be of the order of 5–10 years. The influence of material differences on this time-factor is indistinguishable from the data set.

3.3. Roofs

Table 2 shows the results of the weathering measurements on different roof pavings contaminated at Risø by Chernobyl fallout in rain. Measurements of ¹³⁷Cs in precipitation at Risø confirm that long term deposition of ¹³⁷Cs is negligible compared to the deposition over the first few weeks after the Chernobyl accident.

These measurements show that a widely varying fraction of the contamination initially deposited on the different types of roofing material was removed with run-off water during the deposition phase (Roed, 1987). Corrugated eternit and clay tiles retained most of the contamination, both during the deposition phase and over longer periods of time, whereas the silicon treated eternit surfaces retained the least. The roof surfaces with the greater slope (45°) retain more caesium than those with the lesser slope (30°). The reason for this is presumably that the rainwater runs over a longer distance, offering more ion-exchange sites. It would appear that the natural reduction (by weathering and radioactive decay) in caesium contamination level on roofs can generally be described by a function of the type:

$$C(t) = 0.5 \cdot C_0 \cdot \exp(-t \cdot \ln(2)/T^{1/2}) \cdot (\exp(-\lambda_1 \cdot t) + \exp(-\lambda_2 \cdot t)),$$

where $C(t)$ is the contamination level at time t , C_0 is the contamination level at time 0, $T^{1/2}$ is the radiological half-life of ¹³⁷Cs (30.07 y) and λ_1 and λ_2 are rate constants corresponding to half-lives of respectively some 1–4 and 25–50 years.

Table 1
Measured ^{137}Cs contamination levels (kBq m^{-2}) on various types of walls in Gävle, Sweden, where it rained as the contaminated cloud from Chernobyl passed by^a

Wall type	Orientation	1987	1988	1991	1993	1994	1996
Plastered	South-east		0.78±11%			0.53±12%	
Plastered	South-west		0.55±16%			0.21±27%	
Red clay	South	1.93±9%	1.65±9%				
Red clay	North		3.93±7%	1.85±7%	2.13±15%	1.89±7%	1.36±10%
Yellow clay	North		0.42±15%			0.14±36%	
Yellow clay	South		1.06±11%	0.80±10%	0.76±10%		
Plastered	North	0.99±10%		0.98±10%			
Plastered	East		1.14±10%			0.59±12%	

^a The contamination level on grassed reference surfaces in the area was in the range 150–200 kBq m^{-2} .

Table 2

Measured ^{137}Cs contamination levels on various types of roofs at Risø (as percentages of the initial contamination, 760 Bq m^{-2} , as measured from a 10 m^2 rain sampler)^a

Roof type	7 days	100 days	210 days	390 days	570 days	750 days	5300 days
Corrugated eternit — 45°	87	81	79	76	73	68	41
Corrugated eternit — 30°	80	75	72	69	67	64	35
Red clay tile — 45°	68	62	59	55	48	47	25
Concrete tile — 45°	59	49	45	40	35	30	24
Silicon treat. eternit — 45°	21	15	13	12	12	11	7
Silicon treat. eternit — 30°	19	14	11	10	9	9	5

^a Statistical uncertainties on the measurements ranged between 2% for the most contaminated materials, and 10% for the least contaminated.

4. Discussion

4.1. Streets

The measurements made on streets over the first 2 years following the Chernobyl accident are in good agreement with a series of measurements made in Bavaria, Germany over the same period (Roed & Jacob, 1990). The rather rapid decrease in contamination level on streets ties in with the results of a laboratory experiment (Andersson, Roed, Paretzke, & Tschiersch, 1995), which showed that any caesium present on asphalt pavings is largely confined to a thin surface dust layer, and would inevitably be removed over a relatively short period by mechanical impact (e.g., through traffic). After the first few years, the rate of removal of caesium from these surfaces decreases markedly. A likely explanation for this may be that the contamination levels on the streets are at this point so low compared with e.g., those of adjacent soil areas that the transfer of contaminated soil particles to the streets compensates for much of the contaminant loss from the streets through weathering.

Using the urban dose model URGENT, which is based on the above time-series data, Andersson et al. (1995) estimated the typical dose contribution received by inhabitants of an urban centre (multistorey building environment) over 10 years from streets in an area contaminated by 1 MBq m^{-2} of ^{137}Cs . Here it was assumed that inhabitants spend 15% of their time on the various outdoor surfaces (mainly grass). In some densely populated urban centres, however, most of this time will be spent on streets or other paved surfaces. According to the model estimate, the 10-year dose contribution from these surfaces would then amount to about 3–4 mSv.

In special cases, where there is very little traffic, street surfaces may, however, remain highly contaminated over longer periods. An example of this is the deserted, heavily contaminated town of Pripjat, only about 3 km away from the Chernobyl power plant (Roed & Andersson, 1996). Here, contamination levels on some streets were in 1993 measured to be as high as 300 kBq m^{-2} . Based on the total level of contamination per unit of area in the adjacent soil and the assumption of dry depo-

sition, the initial contamination level on the roads was estimated to be ca. 1500 kBq m⁻². This means that about one-fifth of the deposited caesium had remained on the road surface after 7 years. This is some 4 times as much as was measured in Gävle, for instance. If the depletion processes in Gävle and Pripjat had been the same, the remaining contaminant fraction in Pripjat would in the long run be expected to be the smallest, since the contamination seemed easier to remove by forced decontamination in Pripjat. The explanation for this may be that the contamination in Pripjat, very near the Chernobyl power plant, took place with large (insoluble) core fragment particles.

4.2. Walls

Normally, the contribution to dose from contamination on walls will not be great, due to the vertical orientation of these surfaces. Deposition of contaminants in precipitation will only be significant in rare cases with strong winds and for all particle sizes measured in air after the Chernobyl accident, dry deposition velocities to horizontal surfaces will be much greater than those to vertical surfaces (Underwood, 1987). Measurements made shortly after the accident occurred in 1986 showed that the dry deposition velocity to walls of the ca. 1 µm AMAD caesium contaminant aerosol was about one-tenth of that to a cut lawn (Roed, 1990). With the exception of the near-zone around the Chernobyl power plant, wet deposition led to much higher contamination levels than did dry deposition. Wet deposition, though widely varying, was found to lead to contamination levels on walls, which were typically only a few per cent of those measured on lawns in the same areas (Roed, 1990).

However, as can be seen from the time-series of measurements shown in Table 1, the natural decrease in contamination level on walls is in general very slow, since the wearing of the walls by natural weathering processes is very limited. For comparison with the Gävle results, it can be mentioned that in-situ measurements made in 1993 in the town of Pripjat showed that sandstone and clay brick walls were then contaminated by ca. 200–300 kBq m⁻² of ¹³⁷Cs (Roed & Andersson, 1996). If it is assumed that the deposition pattern in this area was the same as was observed in many other areas of Europe after the Chernobyl accident, the contamination level on walls should as mentioned above be about one-tenth of that on a grassed lawn. As the total ¹³⁷Cs ground contamination in Pripjat was found to be about 3 MBq m⁻², weathering processes must have had little effect on the contamination level on these walls.

Still, estimates using the URGENT model indicate that doses received from contamination on walls over the first decade after deposition will in most cases be at least a factor of 2 smaller than those received from, for instance, the contamination on roads or roofs in the same housing area.

4.3. Roofs

The results presented for roofs are consistent with measurement data obtained in Germany over the first 2 years after the Chernobyl accident (Jacob, Meckbach, &

Müller, 1987; Roed & Jacob, 1990), and the generally high retention of ^{137}Cs by clay tiles and slates over the earliest months agrees with the findings of Sandalls and Gaudern (1988) on wet-contaminated roofs in West Cumbria, UK. The results are also in reasonably good agreement with the results of another series of measurements carried out at Risø. Here clay roof tiles that had been contaminated by Chernobyl fallout in Gävle were brought to Risø after 4 years of exposure to wind and weather (Andersson et al., 1995). The contamination level at this stage was assessed and the tiles were then exposed to Danish weather after having been placed on a specially constructed scaffold. A decrease in the radiocaesium contamination level by between 28 and 35% was recorded over the following 19 months. In a pre-Chernobyl experiment, Wilkins (1987) found that the removal of caesium from roof tiles appeared only to occur during periods of rainfall.

Estimates using the URGENT model (Andersson, 1996), in which weathering parameters are to some extent based on these results, also show that doses received from contaminated roofs may be significant. To people living in single storey buildings or on the top floor of multi-storey buildings in an urban centre with a ground contamination level of 1 MBq m^{-2} , the dose contribution over 10 years from contamination on a clay, concrete or slate roof may under normal conditions be expected to be 3–7 mSv (Andersson et al., 1995). However, measurements made in the Novozybkov area of Russia in 1995 and 1997 (Fogh et al., 1999; Roed et al., 1998) showed that under special conditions, where slate roofs are overgrown with algae and fungi and nothing is done to clean the roof, a much greater part of the initially deposited caesium may be retained. Normally, it would be expected that only ca. 5–10% of the initially deposited radiocaesium would be left on the roof after 11 years. However, if it is assumed that the deposition pattern on the various surfaces in the investigated Russian area was in-line with what was observed in other locations, to which contamination occurred in rain, it could be calculated that about one-third of the contamination had in fact remained on the roof. This phenomenon may be ascribed to the large and rough surface of the algae that covered the asbestos roof, acting as a contaminant sink and offering a great number of ion-exchange sites. It is known that metal ions may become strongly bound in various types of algae (Lujan, Darnall, Stark, Rayson, & Gardeatorresdey, 1994). Cleaning the roof, including removal of organic litter and algae, even at this point reduced the dose rate measured inside the house by about 25%. In comparison, removal of the contaminated topsoil from an area of 20 m by 20 m around this suburban house only reduced the dose rate by ca. 30%. If the open soil area around the house had been smaller, the fractional dose rate reduction by treating the roof would have been even greater.

4.4. General observations

With the possible exception of street pavings, the above results show that radiocaesium contamination is strongly bound and to a great extent retained over long periods of time after its deposition on man-made urban surfaces. This is also the case in soil, where a large fraction of the Chernobyl fallout radiocaesium is still often present in the top few centimetres of the vertical soil profile. It has previously been reported

(e.g., Sawhney, 1967; Nommik & Vahtras, 1982; Andersson & Roed, 1994) that the strong selective fixation of radiocaesium in soil is closely related to the presence in the soil of three-layer micaceous minerals (particularly illite). It is believed that the fixation occurs at frayed edges near the end-surfaces of the mica crystal unit layers.

A wide range of urban construction materials contain illite. For instance, illite is sometimes added to cement-based materials to make stronger and more ductile products (Moukwa, Lewis, Shah, & Ouyang, 1993). Also the mineral tricalcium disilicate hydrate (tobermorite) is sometimes applied for this purpose (Sato, Hashida, Takhashi, & Yamasaki, 1997), and this mineral, which has a layer structure similar to that of illite, has also been reported to exhibit some selectivity for caesium in solutions containing competing cations (Shrivastava & Shrivastava, 1998; Ma, Brown, & Komarneni, 1996). Illite also occurs naturally in limestone applied for producing cement (Sprung & Siebel, 1991), and is usually present in the materials applied to make clay tiles and bricks (Andersson et al., 1995; Strazzera, Dondi, & Marsigli, 1997). Further, de Preter (1990) found that the number of highly selective caesium sorption sites on concrete and eternit-like slate products and even street dust, which to some extent originates from erosion and weathering of urban surfaces, did not differ greatly from what was found on micaceous tile samples, indicating a content of minerals with similar characteristics. The measurement data above shows that clay, concrete and eternit roof tiles for instance, which may all contain illite, retain much caesium over long periods, as opposed to silicon-treated tiles, which would also have a lower open porosity. Especially on those surfaces which are subjected to significant abrasion (i.e. roof tiles rather than bricks), it might therefore be expected that material characteristics, and possibly the content of illite or similar minerals in particular, play a leading role in the weathering process and thereby may govern long-term doses. This can explain the differences shown in Table 2 in the weathering of ^{137}Cs on roof pavings of different materials. However, some urban construction materials are fired, and during the firing process, illite undergoes some changes (Wahl, 1965). Generally, detrital illitic structures will persist until about 1100°C (Cole & Segnit, 1963), but an amorphous vitrification of micaceous materials will set in at approximately 1200°C.

Andersson (1991) demonstrated the impact of firing temperature on the caesium fixation on micaceous urban surfaces. Here, identical “kiln-ready” clay tile samples, in which illite was found to be the predominant micaceous mineral, were fired at respectively 700°C and 1100°C. This represents the lower part of the temperature range at which clay tiles are usually fired. Clay bricks, however, are normally fired at much higher temperatures (ca. 1500°C). The open porosities of the samples were similar, but the specific surface area (BET) of the tiles fired at the lower temperature was about 100 times greater, indicating substantial structural differences. It was found that caesium was much more strongly bound to the tile fired at the lower temperature. Andersson (1991) also demonstrated that the number of strong (virtually irreversible) caesium fixation sites on an ordinary clay tile from a roof in Oxfordshire was great compared with the typical amounts of naturally occurring caesium depositing in British rainwater. In fact, it would take thousands of years just to fill up these sites, which are the ultimate destinations of adsorbed caesium. This means

that even tiles that have been exposed to the weather for centuries can still be expected to have a marked capacity for absorbing and retaining fallout caesium.

5. Conclusions

Long-term field measurement series were made in areas of Sweden and Denmark contaminated by the Chernobyl accident, to examine the impact of weathering on contamination levels on urban streets, walls and roofs. Empirical weathering time-functions of caesium on various types of urban surface were derived from the field investigations, and variation due to material parameter differences was discussed. Based on these time-functions, it is estimated that dose contributions over 10 years from contaminated roofs and from road pavings are to some urban inhabitants of the order of ca. 3–7 mSv in an urban area contaminated by 1 MBq m⁻² of ¹³⁷Cs. As the Chernobyl accident led to much higher contamination levels than this in some areas of the former Soviet Union, the dose contributions from these contaminated urban surfaces may be highly significant. Contamination on walls is expected to be responsible for a somewhat smaller dose contribution. In special cases, for instance where there is very little traffic on roads, or roofs are covered with algae, the caesium retention has been found to be significantly greater than what has mostly been observed.

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