WEATHERING OF RADIONUCLIDES DEPOSITED IN INHABITED AREAS

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ABSTRACT

When determining the long-term consequences of an accidental deposition of radionuclides from a nuclear power plant in an inhabited area it is essential to be able to predict the migration with time of the deposited radiocaesium.

Through the years that have passed since the Chernobyl accident occurred in 1986, the weathering effects on deposited radiocaesium on different types of surface in urban, suburban and industrial areas have been followed through six measurement campaigns to the Gävle area of Sweden. The weathering effects after the Chernobyl accident were also investigated in towns in the Ukraine and in Russia.

The radiocaesium level on asphalt and concrete pavements was found to decrease rather rapidly. It was found that the weathering effects over the first decade could be described by a double exponential function. Similar analytical functions were derived for the other urban surfaces. However, the weathering half-lives of radiocaesium on walls and roofs of buildings were found to be much longer.

Even in urban centres, the largest contribution to the dose-rate immediately after deposition often comes from the open grassed areas and areas of soil. As the dose-rate from such surfaces usually decreases slowly, depending on the soil type, the relative importance of these surfaces will often increase with time.

After a decade, the dose-rate from horizontal pavements will decrease by a factor of 10 or more, but the dose-rate from an area of soil or a roof may only be halved. Correspondingly, the dose-rate from a wall decreases by only 10-20 %.

INTRODUCTION

Typical relationships have been identified between the contamination levels on different types of urban surface shortly after the Chernobyl accident (1). These relationships change considerably with time, due to various 'weathering' processes (depletion of the retained material by such effects as wind, precipitation and mechanical impact of human activity) in the urban environment. An essential requirement for the process towards an estimation of the long-term consequences of an accidental deposit of radionuclides is therefore an adequate knowledge of the effect of weathering. For this purpose, investigations have been conducted in urban environments which received either a wet or a dry deposit of ^{137}Cs after the Chernobyl accident.

URBAN WEATHERING MEASUREMENTS IN SWEDEN

Since the Chernobyl accident, the weathering effects on different types of surface in urban, suburban and industrial areas have been followed in the Gävle area in Sweden, which received a high level of contamination with a heavy rainshower shortly after the accident. In situ measurements with germanium detectors shielded/collimated with lead were made in the centre of Gävle, in a light industrial area about 1 km east of the town centre and in a suburban/rural area about 13 km to the north-east of the town centre.

The radiocaesium levels were measured on various kinds of surface, including walls, grassed areas, pavements, walkways and roads. Six measurement campaigns were conducted in the Gävle area in the period 1987-1994. Table 1 shows a representative selection of the measured data.

Table 1. ^{137}Cs levels measured in Gävle, Sweden in 1987, 1988, 1990, 1991, 1993 and 1994

Location	Surface	Material	Orientation	^{137}Cs levels measured in Gävle, Sweden					
				1987	1988	1990	1991	1993	1994
City hall	wall	plaster	south-east	-	0.78 ± 11%	-	-	-	0.53 ± 12%
City hall	wall	plaster	south-west	-	0.55 ± 16%	-	-	-	0.21 ± 27%
City hall	pavement	flagstone	middle	-	9.71 ± 6%	4.26 ± 7%	3.13 ± 8%	3.10 ± 9%	-
City hall	pavement	flagstone	west	-	6.40 ± 6%	3.15 ± 7%	2.17 ± 8%	2.12 ± 9%	2.12 ± 6%
City hall	road	asphalt		-	1.49 ± 9%	0.44 ± 17%	-	-	-
Gevalia	wall	red brick	south, washing	1.93 ± 9%	1.65 ± 9%	-	-	-	-
Gevalia	wall	red brick	north, washing	-	3.93 ± 7%	-	1.85 ± 7%	2.13 ± 15%	1.89 ± 7%
Fire station	wall	yellow brick	north, dry deposition	-	0.42 ± 15%	-	-	-	0.14 ± 36%
Transformer substation	wall	yellow brick	south, washing	-	1.06 ± 11%	-	0.80 ± 10%	0.76 ± 10%	-
Magasin St.	wall	plaster	north, washing	0.99 ± 10%	-	-	0.98 ± 10 %	-	-
General Food	wall	plaster	east	-	1.14 ± 10%	-	-	-	0.59 ± 12%
Fire station	pavement	asphalt+ flagstone		-	-	3.21 ± 8%	2.38 ± 8%	-	-
Gevalia	car park	asphalt		3.17 ± 7%	3.28 ± 7%	-	-	-	-
Gevalia	cross-roads	asphalt		-	1.19 ± 9%	-	-	-	-

Although the first campaign was made in 1987, a comparison with other measurements made in the same area in May-October 1986 (2) made it possible to approximately relate the measured results to the initial radiocaesium deposition in the area.

The dose-rate reductions which were recorded in Gävle on grassed areas were found to be greatly dependent on the soil type, and the investigated soils displayed many different shapes of vertical profile of ^{137}Cs. Generally, the dose-rate from areas of soil was found to decrease by 40-60 % through the first 8½ years after deposition.

As can be seen from the table, very little, if any decrease in the levels of radiocaesium contamination on walls of buildings was identifiable 8½ years after the deposition. In one case, the level has actually increased at one point. This change is, however, not highly significant, but may have been caused by a wash-down of radioactive substances from the upper parts of the wall. Anyway, the measurement at this site the following year showed the expected decrease. Although it is difficult to distinguish between the weathering rates on the walls in Gävle previous efforts have shown that radiocaesium is retained most effectively by micaceous construction materials, especially those that have been fired at comparatively low temperatures, where the small openings in the mica structure are intact. A yellow brick wall of the fire station has a very low contamination level compared with the other walls because it was exposed to only dry deposition as the structure of the building prevents the wall from precipitation.

In contrast to the situation on walls, the levels of radiocaesium on asphalt surfaces have now decreased so much that less than 10 % of what was measured in 1988 is left. A comparison with the results of Karlberg and Sundblad (2) shows that this means that less than 2 percent of the initially deposited radiocaesium is now left on the road. These levels are now generally below the detection limit. This ties in with the results of laboratory investigations, which have shown that only a very little fraction of a caesium contamination is associated with the bitumen fraction of the asphalt. A large fraction of the caesium has been found to be associated with the more mobile street-dust, which usually contains micaceous substances weathered off various surfaces.

As for the concrete paved surfaces, the remaining 10 % of the initially deposited radiocaesium seems very firmly fixed. No significant decrease has been recorded over the latest 3 years. The weathering on horizontal hard surfaces was generally found to be faster in the more heavily trafficked spots.

Clay roof tiles which were contaminated due to the Chernobyl accident and exposed to the wind and weather in Gävle for four years were collected and brought to Risø, where the contamination level at this stage was assessed. The tiles were subsequently exposed to Danish weather after having been placed on a specially constructed scaffold. A decrease in the radiocaesium contamination level of between 28 and 35 % was recorded over the following 19 months period.

This is in reasonably good agreement with measurements on different types of roof contaminated by Chernobyl fallout at Risø, from which a semi-empirical model was derived (3). The weathering processes on both roofs, walls and horizontal pavings were found to have a slow and a fast component and could be adequately described over the first decade by two-component exponential functions. Some typical values of the parameters are given in Table 2.

Table 2. Typical weathering parameter values on different types of urban surface (slow and fast component)

Surface:	Fraction	Half-life	Fraction	Half-life
Roofs	0.7	$7.0 \cdot 10^3$ days	0.3	$3.5 \cdot 10^2$ days
Pavings	0.5	$7.0 \cdot 10^2$ days	0.5	$7.0 \cdot 10^1$ days
Walls	0.9	$2.0 \cdot 10^4$ days	0.1	$7.0 \cdot 10^1$ days

URBAN WEATHERING MEASUREMENTS IN THE FORMER USSR

Measurements of dry-deposited ^{137}Cs in the Pripyat area in the summer of 1993 with shielded germanium detectors showed that the contamination level on sandstone walls was in the range of 199-350 kBq/m^2 (the decrease since 1986 must have been very small), compared with 0.9-28 kBq/m^2 in other towns, where deposition occurred with rain. On the 'impervious' horizontal surfaces, such as roads and pavements, the caesium contamination level was found to be in the order of 30-350 kBq/m^2 in Pripyat, with the highest levels on concrete pavements and the lowest on asphalt.

From the contamination level and distribution of radiocaesium in local soil it was possible to estimate the caesium contamination level on pavings in the area shortly after the accident to about 1.5 MBq/m^2. This means that a much larger fraction remains on the paved surfaces in Pripyat than in Gävle. However, there has been very little traffic in Pripyat since the Chernobyl accident. If the weathering processes in the two towns had been the same, the remaining fractions in Pripyat would probably have been the smallest at this stage. Anyway, it was easier to remove caesium in Pripyat by forced decontamination trials than in other towns. The reason for this is believed to be that the contamination in Pripyat (near Chernobyl) took place with large (insoluble) core fragment particles.

CONCLUSIONS

The effects of weathering in towns and industrial areas have been investigated through field measurements in areas of Sweden and Russia, which were contaminated with ^{137}Cs after the Chernobyl accident. The radiocaesium levels on road pavings were found to have been halved after less than one year, and after about 3 years only about 10 % remains. The dose-rate contributions from roofs and soil areas were approximately halved in the first 8½ years after deposition, while measurements on walls showed little or no decrease in contamination level. Analytical expressions for the weathering processes on roofs, walls and pavings were derived and estimates of typical parameters were made. Measurements in Pripyat indicated that the impact of human activity on the decrease of the contamination levels can be great.

REFERENCES

1) J. Roed, Deposition and removal of radioactive substances, ISBN 87 7303 514 9 (1990).
2) O. Karlberg and B. Sundblad, A study of weathering effects on deposited activity in the Studsvik and Gävle area, Studsvik Technical Note NP86/78 (1986).
3) J. Roed, Modelling of run-off and weathering processes, presented at the MARIA workshop at Kernforschungszentrum Karlsruhe, Oct. 17-21 (1988).