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An experimental study on natural weathering of radionuclides from urban surfaces for aerosols deposited in wet and dry conditions

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Abstract – The European Model for Inhabited Areas (ERMIN) predicts long-term doses and other consequences following contamination of built-up areas by airborne radioactivity. Central to ERMIN are empirical models of the long-term behaviour of radionuclides on building and other surfaces in the environment. Experiments in which building materials were contaminated, and weathered outdoors for up to 1 year following contamination have enhanced the data on weathering from surfaces that can be used in ERMIN. New data have been obtained for the elements americium, cadmium, caesium, cobalt, iodine, ruthenium and strontium and for the surfaces brick, concrete and clay roof tiles, concrete slabs, glass, sandstone, tarmac and wood cladding. The results have highlighted some differences between the retention of aerosols on surfaces, particularly for glass. This paper presents a summary of the results and identifies the main differences in retention observed.

Keywords: remediation / decision support / surface retention

1 Introduction

The European Model for Inhabited Areas (ERMIN) predicts long-term doses and other consequences following contamination of built-up areas by airborne radioactivity. Central to ERMIN are empirical models of the long-term behaviour of radionuclides on building and other surfaces in the environment (Charnock, 2016). The weathering information currently used in ERMIN is largely based on caesium. Experiments in which building materials were contaminated and weathered outdoors for up to 1 year following contamination have been undertaken. The main aim was to look at how appropriate the data in ERMIN are for a range of surfaces and elements and to expand the pool of information available.

2 Experimental design

Building materials were contaminated with a range of radionuclides that represent groups of elements with different chemical properties that might be encountered following a radiological incident. The radionuclides chosen had gamma-ray emissions so they could be measured quickly by gamma spectrometry, enabling more experiments and repeated measurements to be made. Two parallel experiments were carried out: one set of surfaces contaminated using a simulated wet deposition and the other using a simulated dry deposition. PHE has the capability to generate droplets of solution containing soluble salts labelled with radionuclides. These were left to dry and deposit on the surface giving a simulated

dry deposition. It is noted that starting with a solution and allowing the droplets to dry on the surface implies a high degree of solubility. Wet deposition was simulated by adding a solution containing the radionuclide to the surface and allowing it to dry.

The run-off water from each surface was regularly collected until any further removal was not measurable. The percentage removal of the contaminants was calculated from the amount of contaminant in the run-off water as a percentage of the initial amount of contaminant applied, having been decay corrected. A rainfall gauge measured the amount of rainfall in each measurement period. Results from earlier PHE work (Brown *et al.*, 2011) have been added to the pool of data available to the PREPARE project. The experiments are summarised in Table 1.

3 Results and discussion of retention on surfaces

The results of PREPARE study are shown in Table 2. The main findings and observations from the experimental work are briefly described. Retention on porous surfaces (bricks, tiles, concrete and stone) was, in general, similar with a maximum removal of 50% and typical removals of 20–30% for 500–600 mm rainfall. An exception to this was observed for strontium, which was removed more quickly as well as all the contamination being removed (see Figure 1). This suggests that strontium remains more soluble in the surface matrix and is easily removed by rainfall.

There is limited evidence that dry deposition was removed somewhat quicker than wet deposition, and the total removed quantity over the experimental period (total of 527-mm

Table 1. Summary of main weathering experiments in the two studies.

Element	Surfaces	Deposition	Duration/total rainfall
<i>PREPARE study</i>			
Caesium, iodine, ruthenium	Framed glass, brick, clay roof tile, sandstone, tarmac, wood cladding	Wet, dry	1 year (Sep–Sep)/629 mm
<i>PHE study</i>			
Americium, cobalt, cadmium, strontium	Glass, brick, concrete roof tile, concrete slab	Wet	5 months (Nov–Apr)/527 mm

Table 2. Percentage of elements remaining on surfaces as a function of rainfall.

Date	Nov 2013				Dec			Jan 2014		Feb	Mar	Apr
	07	11	19	28	17	23	27	04	18	06	12	26
Cumulative rainfall (mm)	5	15	18	22	40	86	130	198	259	390	468	527
<i>Cs (wet deposition)</i>												
Wood	15	13	12	12	11	10	10	9	9	8	8	7
Glass	6	3	3	3	3	3	2	2	2	2	2	2
Sandstone	100	99	99	99	99	98	97	96	95	94	94	94
Tiled roof	97	96	96	96	94	93	92	90	89	86	85	83
Tarmac	100	99	99	99	99	99	99	99	99	98	98	98
Brick wall	100	100	100	100	99	97	95	93	92	90	88	88
<i>Cs (dry deposition)</i>												
Wood	13	11	11	11	9	8	8	8	7	7	6	5
Glass	10	10	10	9	9	9	9	9	9	9	9	9
Sandstone	97	95	94	94	93	90	88	85	83	81	79	78
Tiled roof	77	72	69	67	62	56	51	46	44	37	34	31
Tarmac	93	91	90	89	89	88	87	86	85	84	83	82
Brick wall	97	97	96	96	94	88	86	82	79	75	73	73
<i>Ru (wet deposition)</i>												
Wood	44	40	36	36	35	34	34	33	32	31	29	28
Glass	82	81	80	80	79	78	77	75	74	73	72	71
Sandstone	100	99	99	99	98	95	92	84	75	66	59	56
Tiled roof	95	94	93	93	92	91	90	89	89	88	87	86
Tarmac	96	95	95	94	94	94	94	94	93	93	93	92
Brick wall	100	100	100	100	100	99	99	98	98	97	97	97
<i>Ru (dry deposition)</i>												
Wood	53	51	49	49	44	42	41	38	35	31	25	22
Glass	31	31	30	30	29	29	28	28	27	26	25	24
Sandstone	94	92	90	90	88	84	82	78	76	72	69	68

Table 2. (continued).

Date	Nov 2013				Dec			Jan 2014		Feb	Mar	Apr
	07	11	19	28	17	23	27	04	18	06	12	26
Tiled roof	84	82	80	79	76	73	71	67	64	60	57	53
Tarmac	73	69	64	63	59	56	53	50	47	43	36	32
Brick wall	98	97	96	96	93	83	78	74	71	66	63	62
<i>I (wet deposition)</i>												
Wood	92	90	90	90	87	86	86	84	83	80	78	78
Glass	95	95	95	95	95	95	95	95	95	95	95	95
Sandstone	98	97	97	97	96	94	93	90	87	83	81	81
Tiled roof	85	78	76	75	72	71	70	68	67	64	62	62
Tarmac	95	91	90	89	87	86	86	85	83	82	80	79
Brick wall	100	100	100	100	99	97	96	93	90	87	87	87
<i>I (dry deposition)</i>												
Wood	70	64	63	62	54	51	51	44	41	41	41	41
Glass	90	90	90	90	90	90	90	90	90	90	90	90
Sandstone	90	85	84	83	81	78	77	71	68	67	67	67
Tiled roof	60	52	49	49	45	42	41	39	36	36	36	36
Tarmac	60	43	38	36	30	29	29	23	20	19	19	19
Brick wall	98	97	96	96	95	91	90	85	81	81	81	81

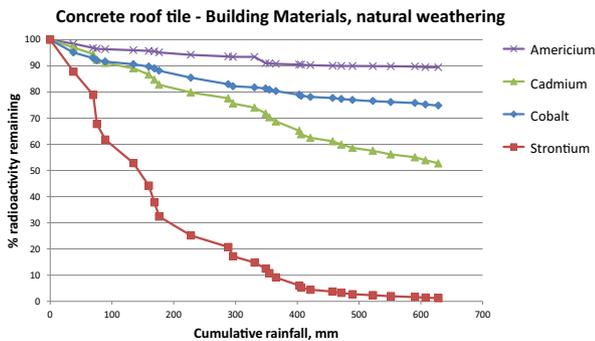


Figure 1. Weathering from concrete roof tile.

rainfall) was higher, although this was not consistent across the building materials. For clay roof tiles and tarmac, there was clearer evidence that the dry deposition, particularly of iodine and ruthenium, was higher by a factor of about 2–3 (60–70% removed compared to 10–20% removed for wet deposition). Figure 2 shows a comparison of weathering following simulated wet and dry deposition for brick wall and tarmac that shows the differences.

For glass and wood cladding, an initial high removal at the time of the first rainfall was observed, except for americium. Further experiments indicated that the short-term removal of elements from glass, and particularly americium, was

dependent both on the time between deposition and the first rain and the weather conditions over this period, hot weather appearing to effectively ‘bake’ the contamination onto the surface with the effect of significantly slowing down the rate of removal. There is some evidence that iodine and ruthenium were removed less from wood and glass than the other elements. The overall weathering from glass was also observed to be very variable and different between the PHE and PREPARE studies. A sheet of glass was used in the PREPARE experiments rather than glass in a window frame, the latter showing a much higher overall removal. It is difficult to further interpret these results, as the initial weather conditions, the amount of rain during the first rainfall event following deposition and possibly the type of glass; all appear to influence the amount and rate of weathering.

4 Conclusion

Undertaking experiments in real weather conditions has meant that interpretation of the results is not straightforward, as there are potentially many factors influencing the weathering removal and removal rate, as would be the case in reality. The overall rainfall in the 2 experimental periods was similar, although the rainfall pattern was different. The experiments show that, in general, there are not significant differences between the weathering of different elements

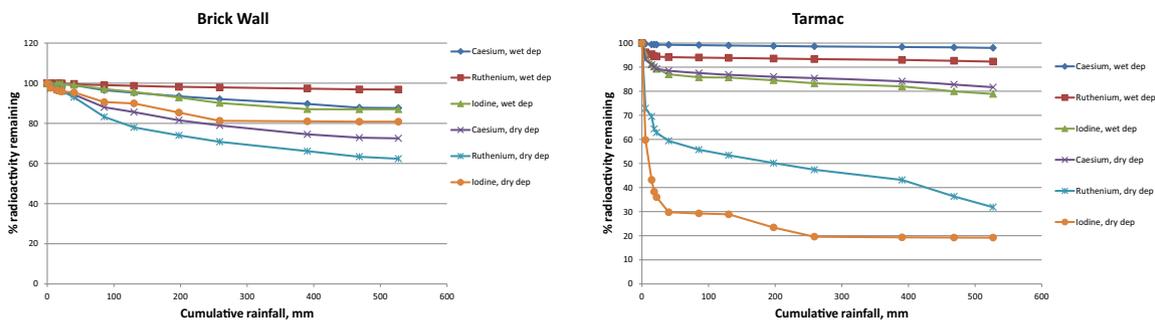


Figure 2. Weathering from brick wall and tarmac showing differences between elements.

from an individual surface with a few exceptions, notably strontium. There are differences, however, between surfaces, the notable one being glass, where removal is variable and not necessarily rapid, as might be expected. The results also indicate that there can be significant removal during the first rainfall event following deposition, and it may be appropriate to include this feature in the ERMIN model. The data will be used to derive weathering rates for aerosols for use in ERMIN.

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References

Brown J., *et al.* (2011) Development of experimental techniques to investigate the behaviour of chemical and radiological materials on urban surfaces. HPA Chemical Hazards and Poisons Report, Issue 20.
 Charnock T.W. (2016) Enhancement of the ERMIN urban dose and remediation model to account for physicochemical properties of contamination, *Radioprotection*, **51** (HS2), S105-S107.

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