Radioprotection, vol. **46**, n° 6 (2011) S265–S269

DOI: 10.1051/radiopro/20116593s

Fate and transport of radiocesium in urban building materials

A. Gusarov[1], N. Il'icheva[1], A. Konoplev[1], S.D. Lee[2], K. Maslova[1], V. Popov[1] and I. Stepina[1]

[1]*RPA "Typhoon", Obninsk, Kaluga region 249038, Russia*
[2]*National Homeland Security Research Center, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711, USA*

Abstract. Fine radioactive particulate dispersal in outdoor areas presents significant economic, social, environmental and public health concerns. The interactions of these radioactive particles with urban surfaces need to be well understood to develop optimized decontamination strategies. The major environmental factors influencing these interactions are relative air humidity, temperature and rain. The objective of this work is to investigate the fate and transport of water soluble cesium deposited on conventional urban building materials, especially concrete, brick, asphalt, limestone, and granite, under various environmental conditions (relative humidity (RH), and atmospheric precipitation). The data on the kinetics of ^{137}Cs desorption from building materials by the solution containing 100 mM of Ca^{2+} and 0.5 mM of K^{+} have shown that the shape of the ^{137}Cs desorption curves is similar for all building materials. There is a rapid initial decrease in the activity of the sorbed ^{137}Cs. The desorption rate first decreases quickly during 7–10 days and practically does not change later. The remaining ^{137}Cs in building materials that is not desorbed by the solution is 30-40% of the initially sorbed amount. For the building materials of interest, radiocaesium interception potentials (RIP). The RIP(K) value has been shown to range from 20 to 300 mM/kg and increase in the order: limestone > brick > concrete > granite > asphalt. The fine fraction of building materials (<0.125 mm) sorbs ^{137}Cs better than the coarse fraction (0.0125–0.25 mm). Based on RIP(K) value and measured concentration of cations the distribution coefficients K_d(^{137}Cs) were determined. The study of the mechanisms of radiocesium binding by different components of building materials, based on sequential extraction technique, shows that the highest ability to bind ^{137}Cs is characteristic of asphalt which retains 40.9±1.0 % of ^{137}Cs after all extractions. By the ability to bind ^{137}Cs with the residual fraction, the studied materials form the following sequence: concrete > limestone > granite > brick. Method to study radionuclides distribution in depth of building materials using layer-by-layer grinding has been developed.

1. INTRODUCTION

It is important to develop decontamination and clean-up technologies after incidents that involve radioactive particulates dispersed in an urban area. Fine radioactive particulate dispersal in outdoor areas presents significant economic, social, environmental and public health concerns. The interactions of these radioactive particles with urban surfaces need to be well understood to develop optimized decontamination strategies. An expanded understanding of these interactions under varied atmospheric conditions will aid in the development of more effective decontamination techniques and strategies [1, 2]. The objective of this work is to investigate fate and transport of water soluble cesium deposited on common urban building materials, especially concrete, brick, asphalt, limestone, and granite, under various environmental conditions (relative humidity (RH), and atmospheric precipitation).

2. MATERIALS AND METHODS

Building materials of asphalt, brick, granite, concrete and limestone have been studied. Material coupons have been powdered and three granulometric fractions have been obtained: <0.125; 0.125–0.25; >0.25 mm (Table 1). Principal physical properties of building materials under study are presented

Table 1. Distribution of building material powders by granulometric fractions.

Fraction, mm	Proportion %				
	Asphalt	Brick	Granite	Concrete	Limestone
<0.125	42.8	36.8	53.3	53.3	100
0.125–0.25	38.7	23.5	46.7	45.5	0
>0.25	18.5	39.7	0	1.2	0

Table 2. Selected physical properties of building materials.

	Porosity, cm^3/cm^3	Specific weight, g/cm^3	Hygroscopic moisture, %
Concrete	0.32	2.73	0.4
Granite	0.053	2.77	0.02
Limestone	0.174	2.72	0.03
Asphalt	0.194	2.71	0.095
Brick	0.311	2.77	0.07

Table 3. Elemental composition of Total content of building materials.

	Asphalt	Limestone	Brick	Granite	Concrete
	Macroelements, g/kg				
K	5.87	0.223	15.9	26.6	7.61
Na	23.2	0.543	20.9	40.9	4.55
Ca	26.3	530	10.7	124	29.4
Mg	21.8	8.56	30.6	3.27	11.5
Fe	43.0	0.655	37.5	9.03	27.6
Al	66.6	0.461	84.2	84.9	40.8

in Table 2. The ability of radionuclides to be sorbed largely depends on the chemical composition of building materials. Data on elemental composition of the building materials are shown in Table 3. Note the significant amount of calcium in limestone.

3. RESULTS AND DISCUSSION

The kinetics of ^{137}Cs sorption by the powdered building materials was studied in aqueous suspension at 20 °C for the interaction time 1, 7, 14 and 28 days. For concrete, limestone and brick the maximum degree of adsorption was seen after 24 hours of interaction, while for asphalt and granite it occurred after 6 days (Figure 1). In addition the ^{137}Cs sorption kinetics by building materials was determined using the dynamic method of investigating the ^{137}Cs sorption from the solution containing 100 мМ of Ca^{2+} and 0.5 мМ of K^+. Like in suspensions, sorption of ^{137}Cs by concrete and limestone from this solution becomes maximum in 24 hours of interaction. For asphalt and concrete the sorption equilibrium is established during about 3 days at 35 °C. Unlike the experiments with suspensions, the ^{137}Cs sorption equilibrium for brick sets in slower (15–20 days). The differences in the kinetics of ^{137}Cs sorption for the two methods can be explained by the fact that the studies were conducted at different temperature and under different conditions (no kinetic resistance in the film surrounding the sorbent particles in case of the limited volume method).

The data on the kinetics of ^{137}Cs desorption from building materials by the solution containing 100 мМ of Ca^{2+} and 0.5 мМ of K^+ have shown that the shape of the ^{137}Cs desorption curves is similar for all building materials. There is a rapid initial decrease in the activity of the sorbed ^{137}Cs. The desorption rate first decreases quickly during 7–10 days and practically does not change later. The remaining ^{137}Cs in building materials that is not desorbed by the solution is 30–40% of the initially sorbed amount.

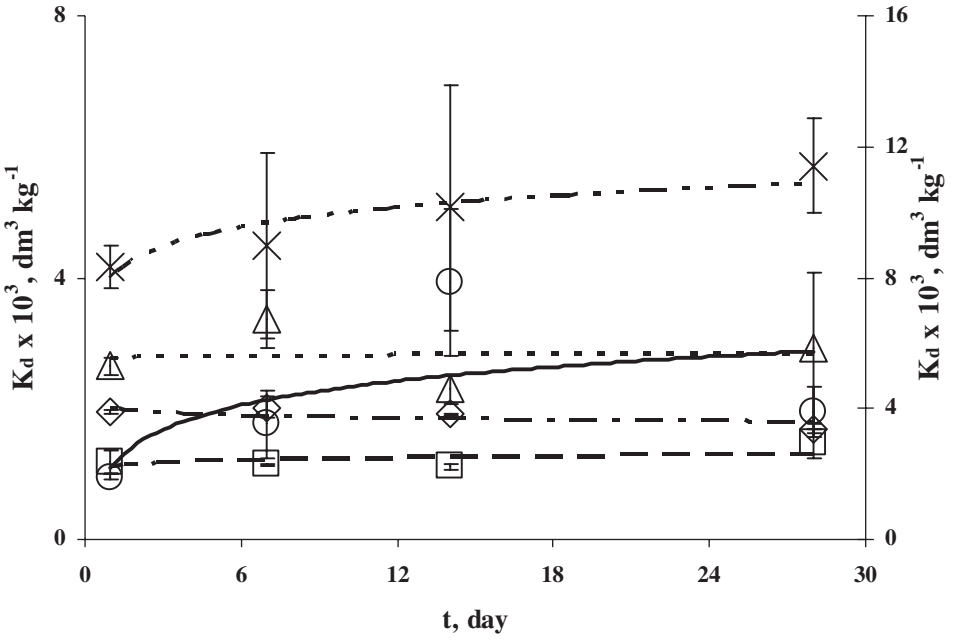

Figure 1. Kinetics of the distribution coefficients $K_d(^{137}Cs)$ in building materials at 20 °C: × -asphalt; △ -granite; ◇ -concrete; ○ -limestone; □ -brick.

Table 4. Radiocesium interception potentials RIP(K) and RIP(N) in the building materials per absolutely dried weight.

	RIP(K), mmol/kg		RIP(N), mmol/kg	
	<0.125	0.125–0.25	<0.125	0.125–0.25
Asphalt	280.44±16.3	135.80±10.7	29.45±0.58	20.77±0.078
Limestone	12.45±0.43	–	1.90±0.24	–
Granite	204.38±21.5	36.57±2.58	11.44±0.87	3.21±0.062
Concrete	185.35±5.10	68.57±1.83	195.43±1.51	44.56±0.054
Brick	14.93±3.03	9.62±1.25	5.35±0.57	2.36±0.22

For the building materials of interest, radiocesium interception potentials (RIP) were determined with respect to potassium ions RIP(K) and ammonium according to the methodology [3], as well as the selectivity coefficients values for exchange of potassium and ammonium ions K_c (K/N) on the selective sorption sites (Table 4). The RIP(K) value has been shown to range from 20 to 300 mM/kg and increase in the order: limestone > brick > concrete > granite > asphalt. The fine fraction of building materials (<0.125 mm) sorbs ^{137}Cs better than the coarse fraction (0.0125–0.25 mm). The extent to which sorption varies for different materials, however, is different: from 1.5 for granite to 15 for asphalt. The value of K_c(K/N) varies from 1.5–2.8 for concrete and brick to 7–18 for asphalt, limestone and granite, which suggests a possible impact of clay particles, such as illite, on the ^{137}Cs sorption. Based on RIP(K) value and measured concentration of cations the distribution coefficients $Kd(^{137}Cs)$ were determined.

Based on the desorption kinetics curves the mechanisms of the ^{137}Cs sorption by building materials have been estimated. The solution containing 100 мМ of Ca^{2+} and 0.5 mM of K+ desorbs about 60–70% of ^{137}Cs in 10–30 days. The study of the mechanisms of radiocesium binding by different components of building materials, based on sequential extraction technique, shows that the highest ability to bind ^{137}Cs is characteristic of asphalt which retains 40.9±1.0 % of ^{137}Cs after all extractions. By the ability

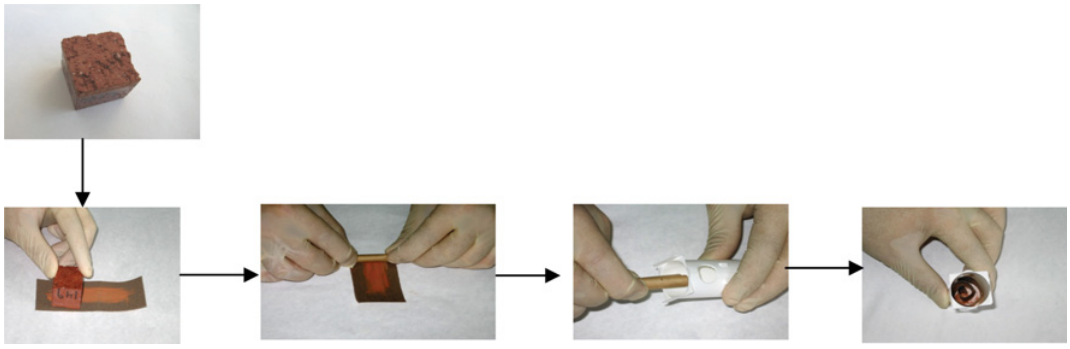

Figure 2. Sequence of steps for building material grinding.

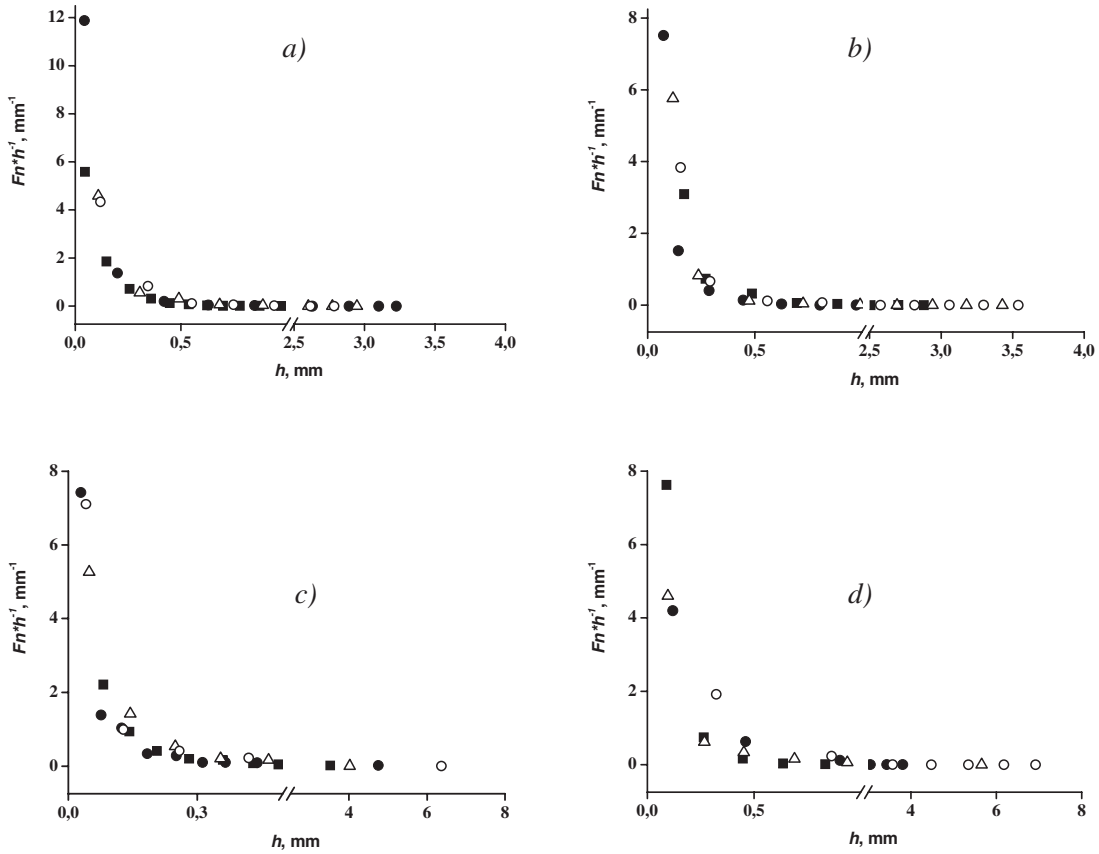

Figure 3. Depth distribution of ^{137}Cs in building materials as a function of interaction time (■ – 1 day, ◇ – 7 days, △ –14 days, ○ – 28 days) a) brick, humidity 30%; b) brick, humidity 85%; c) limestone, humidity 30%, d) limestone, humidity 85%.

to bind ^{137}Cs with the residual fraction, the studied materials form the following sequence: concrete > limestone > granite > brick.

The fractions of the exchangeable ^{137}Cs in the powdered building material samples after sorption during 24 hours from the solution with the potassium adsorption ratio 0.05 are as follows: 24.2±1, 0% for limestone, 8.9±1, 3% for granite, 38.4±1, 5% for concrete, 8.5±0, 3% for asphalt.

Effective method to study radionuclides distribution in depth of building materials using layer-by-layer grinding has been developed (Figure 2). Using the developed method, the ^{137}Cs distribution in depth of selected building materials at different air humidity and time at 20 °C was studied (Figure 3). More than 90% of applied activity in limestone is distributed in the surface layer to the depth of 1.5 mm. There have not been detected any obvious dependence on relative air humidity and incubation time. On the other hand, for ^{137}Cs distribution in the layer more than 0.2 mm a weak dependence of the profile on interaction time and relative air humidity can be seen. With respect to ^{137}Cs distribution in depth of brick and concrete, that there is no clear impact of incubation and relative air humidity on the shape of ^{137}Cs depth distribution curves. Nevertheless it needs to be emphasized that 95% of applied ^{137}Cs occurs in the layer 0–1.0 mm for brick and 0–0.5 mm for concrete.

Acknowledgment

The work was funded by US Environmental Protection Agency through Partner Project with International Science and Technology Center (ISTC Project #4007)

References

[1] Lee, C. P., Kuo, Y. M., Tsai, S. C., Wei, Y. Y., Teng, S. P., and Hsu, C. N. *Journal of Radioanalytical and Nuclear Chemistry*, **Volume 275** (2008) 343-349.

[2] Real, J., Persinb, F., and Camarasa-Claret, C. *Journal of Environmental Radioactivity* , **Volume 62** (2002) 1-15.

[3] Wauters J., Elsen A., Cremers A., Konoplev A.V., Bulgakov A.A. and Comans R.N.J. *Applied Geochemistry* **Volume 11** (1996) 589-594.