

# Dirty bomb fallout

*Lawrence Livermore National Laboratory has studied the deposition of caesium on urban concrete after detonation of a mock radiological dispersal device.*

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**A**t present, there is a significant need to develop decontamination agents that can be used effectively after detonation of a radiological dispersal device (RDD) in an urban environment. There is also a need for development of reproducible test surfaces to be used to determine the efficacy of the agent being developed.

Researchers at America's Lawrence Livermore National Laboratory (LLNL), under the auspices of the US Department of Energy (DoE), conducted a field study to evaluate the deposition of an explosively dispersed radionuclide surrogate CsCl on grime-bearing and non-grime-bearing urban surfaces. The goal was to investigate preparation and contamination of urban surfaces that closely mimic what one would expect to encounter following RDD detonation. Migration of Cs into concrete surfaces was investigated in detail.

Many non-proliferation, security and response organisations that have modelled RDD scenarios use Cs-137, as well as Co-60, Sr-90, Am-241 as the most likely RDD agents. Cs-137 is an isotope of concern for possible use in an RDD due to its potential availability resulting from its widespread legitimate use in construction, geotechnical and medical industrial devices. In some Cs-containing instruments the Cs-137 is present as the highly dispersible and water soluble salt, CsCl. In this form Cs is able to rapidly disperse in the environment, as witnessed in the 1987 Goiânia accident in Brazil, when inadvertent dispersal of a radiotherapy source resulted in fatalities and injuries.

## EXPLOSIVE PREPARATION

At LLNL's contained firing facility a mock RDD made using C-4 plastic explosive and non-radioactive CsCl was detonated. Prepared concrete samples were made using four different conditioning regimes to mimic a

range of conditions that may be encountered during an RDD event. This sample set included dry, wet, carbonated and non-carbonated cores with and without the application of urban grime. In addition, intact concrete samples removed from an urban surface were placed inside the firing facility chamber. The samples were placed inside the firing chamber at three different distances from the mock RDD device. Following detonation, the samples were removed from the firing chamber and selected cores were characterised by laser ablation and scanning electron microscopy.

Two types of concrete samples were placed in the chamber: lab-prepared 3.8cm diameter by 7.6cm-long cylindrical cores and 12.8x12.8x5.1cm concrete blocks retrieved intact from an urban transit system subway tunnel. Lab-prepared cores were used in order to generate a large number of samples with consistent, measurable physical characteristics that could be pre-conditioned in a controlled environment. A total of 300 concrete cores were prepared over a period of three months.

Upon removal from the moulds, the concrete cores were placed in a 19-litre bucket of 3g/litre Ca(OH)<sub>2</sub> solution to soak for a period of seven days. After the cores were removed from the Ca(OH)<sub>2</sub> solution, they were placed in an empty bucket and thoroughly rinsed with tap water. After rinsing, five cores were removed from each pour, patted dry with a paper towel and weighed. All of the cores were then placed in an oven that was pre-heated to 60°C and dried for a period of 48 hours. Following this initial conditioning period, the cores were further conditioned using one of the following methods: grime addition with accelerated carbonation, grime addition without carbonation, wet conditioning without grime or dry conditioning without grime.

The objective of grime addition without carbonation was to prepare concrete samples with an attached grime layer similar to that found in urban environments. Two different grime materials were used to condition cores.

Some 35 cores were conditioned with National Institute of Standards and Technology (NIST) urban dust (standard reference material #1649a) and 35 cores were conditioned with grime material that was collected from an urban transit system.

The transit system grime material was collected from a vent inside of a transit station and was comprised of predominately Fe (40%), S (22%), Ca (15%) and Si (10%). Full characterisation of this material is still in progress. The NIST urban dust was a well-characterised material comprised of inorganic C (18%), Fe (3%), S (3%), Pb (1%), Mg (1%), and smaller amounts (<1%) of polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs) and pesticides. A 1:1(wt) grime to DI H<sub>2</sub>O slurry was prepared in a small glass beaker and 1ml of slurry was applied to the core surface using a 10ml disposable plastic syringe.

Control cores were prepared by adding 10ml of DI H<sub>2</sub>O to the core surface. The grimed cores were placed in an environmental chamber which held an open container of saturated Mg(NO<sub>3</sub>)<sub>2</sub>.6H<sub>2</sub>O solution. The goal was to maintain a relative humidity of ~53% during core conditioning. The cores were maintained under these conditions for a total of 21 days prior to placement in the contained firing facility chamber.

For the accelerated carbonation conditioning batch of cores, our goal was to prepare concrete cores that would have a measurable carbonation layer to more closely mimic aged concrete found in urban environments. A total of 60 cores (20 NIST, 40 transit grime) were subjected to two applica-



tions of grime and two five-day carbonating conditioning periods. Grime slurry was prepared with 0.562g grime in 22.5g DI H<sub>2</sub>O. Following grime addition, the cores were placed in a vacuum dessicator that contained a saturated KI solution beneath the perforated plate. After the lid was placed on the desiccator, the air in the chamber was evacuated and the headspace was purged with a mixture of 50%N<sub>2</sub>/50%CO<sub>2</sub>.

Phenolphthalein testing of these cores indicated that no measurable carbonation layer was formed on this set of samples as expected. We believe this is the result of insufficient CO<sub>2</sub> and too little carbonation time. A follow-on series of experiments are planned to develop a more effective accelerated carbonation method.

After the 48-hour drying period of the basic cores was completed, a total of 50 cores were subjected to wet conditioning with no grime. After the cores were removed from the oven, they were submerged in a container of tap water for a period of six days. After the soaking period, the cores removed from the container and placed in a dessicator with a tray of water placed beneath the perforated plate. These cores were held in the dessicator until placement in the sample holder, two hours prior to device detonation.

After the 48-hour drying period of the basic cores was completed, a total of 50 cores were subjected to dry conditioning without grime addition. After the initial 48-hour drying period had elapsed these cores were dried at 60°C for an additional 72 hours. The cores remained in the drying oven until the day they were placed inside the samples holders, one day before device detonation.

## FIRING SETUP

The contained firing facility is an approximately 2100m<sup>3</sup> firing chamber (17x15x9m) designed to safely contain detonations of up to 60kg of high explosives without release of hazardous materials to the environment. The primary mission of the facility is to support hydrodynamic testing of both nuclear and conventional weapons systems under the DoE's stockpile stewardship programme.

The project team had access to the firing chamber for a total of six days. On day one, the chamber was set up and all prepared samples were set up, with the exception of the wet conditioned samples. On day two the wet conditioned samples were placed in the chamber and the mock RDD was

detonated. On days three, four and five, the chamber remained sealed with samples inside. On day six, the chamber was purged and the samples retrieved.

In the firing chamber, three 2x2.5x0.05m steel blast shields were set out at distances of 3, 6, and 9m from the mock RDD to serve as immobile surfaces to attach the sample holders to. The sample holders were constructed of high-density aircraft foam that was machined to secure the samples during detonation over-pressure and facilitate sample removal without disturbing the sample face containing the explosively deposited Cs. The sample holders contained several of each form (wet, dry, grimed) of laboratory prepared samples as well as field-collected and analysis-specific (for example, elipsometry) prepared media. The sample holders were attached to the blast shields with metal bands. The vertical sample surfaces were arranged to maximise exposure to the mock RDD. Floor sample holders that contained similar samples were placed behind the blast shields and attached to 1x1x0.05m steel plates to assess horizontally deposited 'fallout' Cs compared to explosively deposited Cs.

Most controlled firing experiments are designed with the high explosive (HE) placed over the 150mm thick steel shot anvil in the centre of the chamber floor. Chamber dimensions in this configuration would not allow us to place samples at 9m from point of detonation.

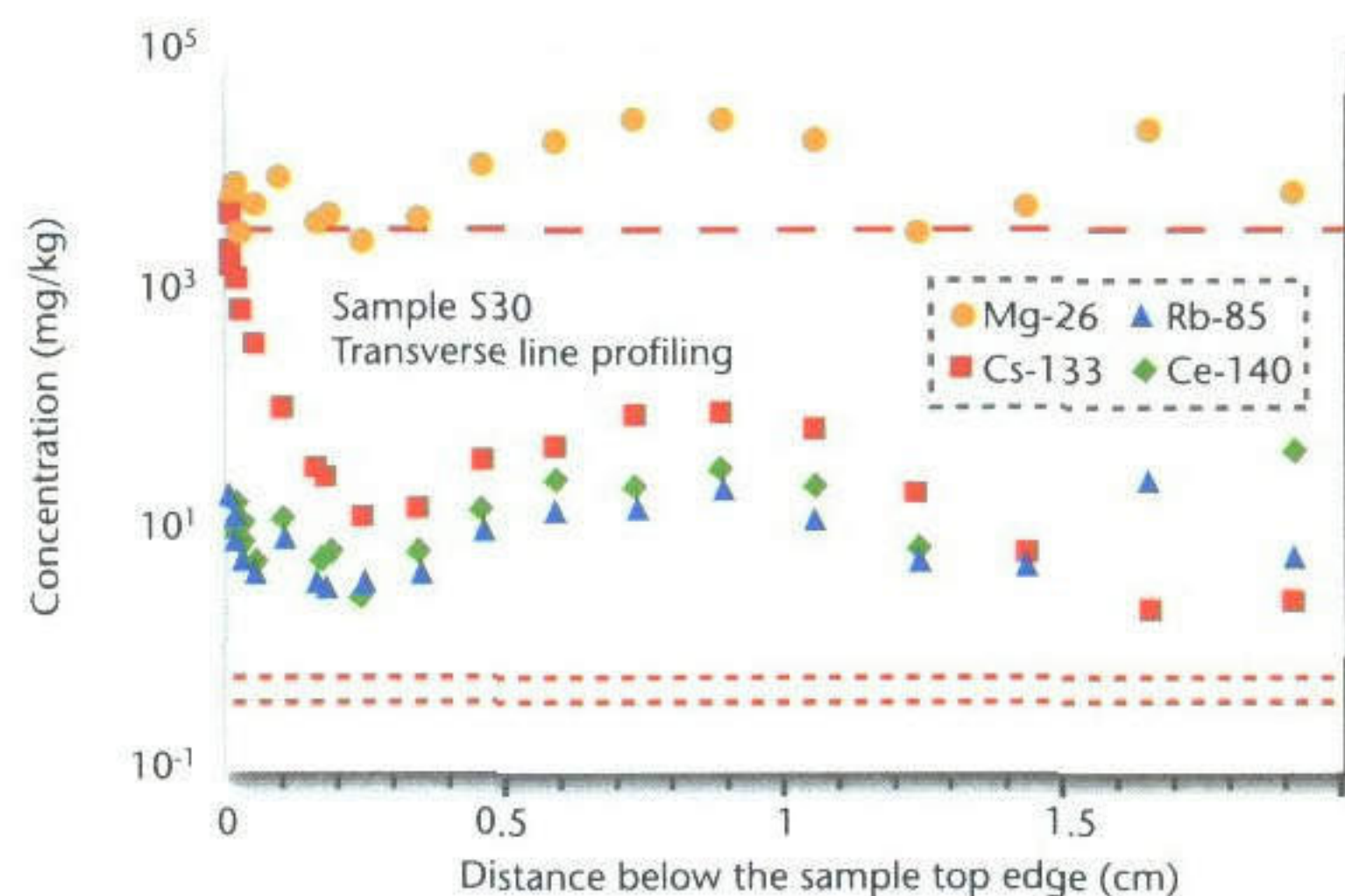
Due to the small amount of HE (1.5kg C-4) in the mock RDD, we were able to position the mock RDD in a corner of the chamber over a 50mm steel plate. Some 1kg of non-radioactive CsCl was placed in the mock RDD as a surrogate for Cs-137. The mock RDD was suspended from an aluminium A-frame at a height of 1m and aligned with the sample holders attached to the blast shields.

## RESULTS

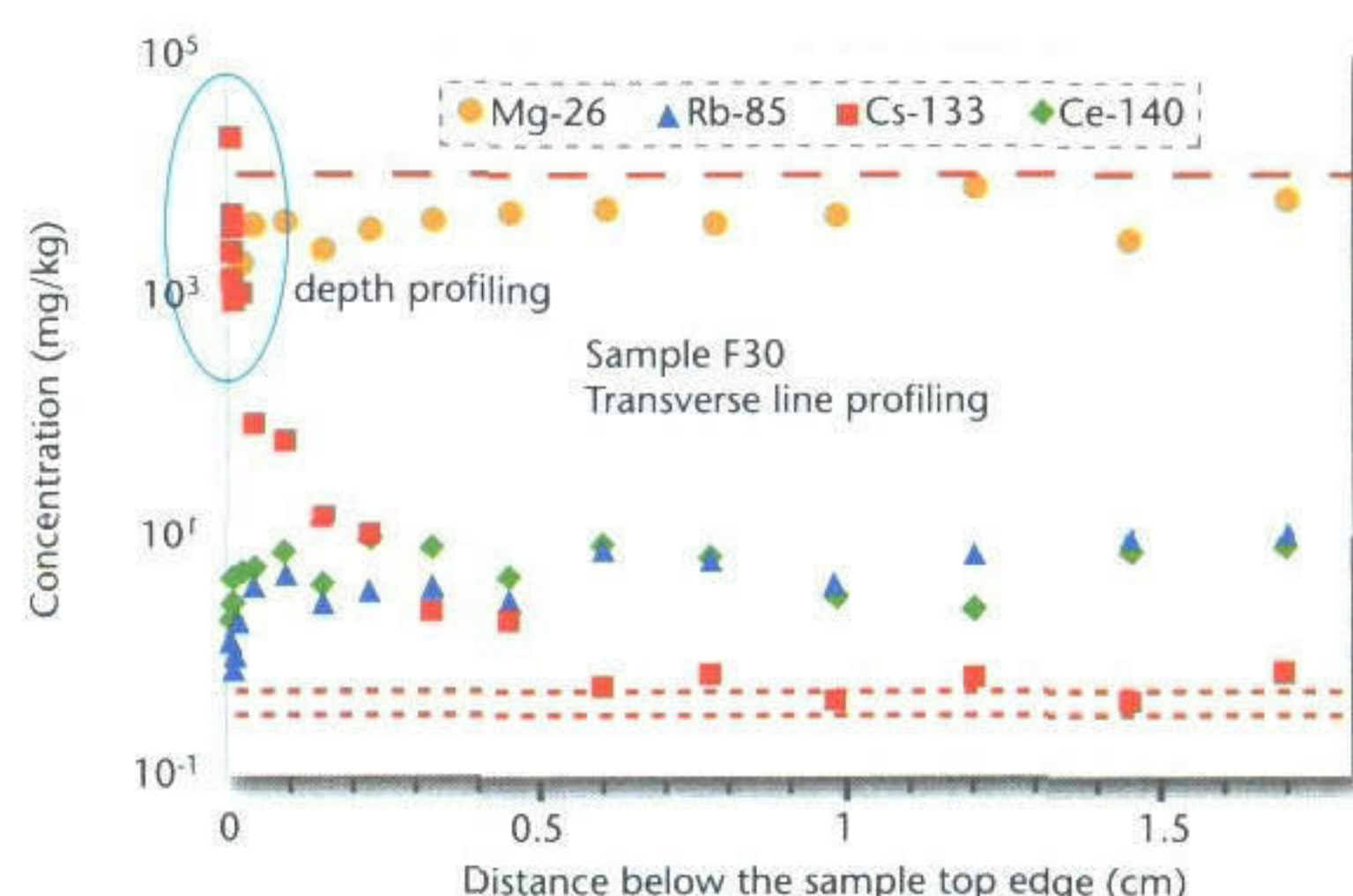
Sample characterisation at LLNL used several different techniques including laser ablation with inductively coupled plasma spectrophotometry (LA-ICP), scanning electron microscopy (SEM), elipsometry, and ion microprobe.

Preliminary results suggest that significant migration of Cs into concrete surfaces may occur following explosive deposition of Cs on the surface (see data in the Figures shown, determined by LA-ICP). The transverse concentrations were measured by splitting the samples vertically and taking a series of

Cs deposition with depth for an urban sample placed on a blast shield located 9m from mock device



Cs deposition with depth for an urban sample placed on the floor 9m from mock device



In both Figures, the upper dashed line represents Cs concentration on the sample surface. The lower double dashed lines represent Cs concentration in control samples (not placed in chamber) plus one standard deviation.

laser ablation measurements down the fractured surface.

It appears that Cs migration into the floor sample is evident to a depth of 0.5cm while in the sample secured to the blast shield, the Cs migration extends at least 1.5cm below the concrete surface.

It should be noted that the amount of Cs deposited on the concrete surfaces may far exceed what one would expect to encounter following an actual RDD event. We designed a device capable of delivering these higher concentrations to ensure that sufficient dispersal of Cs would be achieved to allow the characterisation of samples with available analytical instruments.

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