

A SAFETY ASSESSMENT OF SPENT FUEL  
TRANSPORTATION THROUGH URBAN REGIONS<sup>a</sup>

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INTRODUCTION

Public concern has been focused on the risks that the transportation of nuclear materials pose to human health and the environment. Various scenarios involving accidents and sabotage of spent fuel transports have been analyzed in an effort to determine the resulting health consequences. In 1979, the NRC sponsored a draft environmental assessment of the transport of radionuclides in urban environments. The so-called "Urban Study" evaluated the radiological hazards resulting from the transportation of radioactive material in urban areas for various types of environments including those caused by sabotage. A significant conclusion of the Urban Study was that approximately 0.07% of the heavy metal solid inventory (1.4 metric tonnes) of a truck transported cask could be released as respirable aerosol as a result of a postulated sabotage event. Since no experimental data were available for these analyses, certain bounding conditions were assumed and specific chemical and physical characteristics of the released material were anticipated. We report results from experiments on a full scale fuel cask containing a single surrogate spent fuel assembly (0.5 metric tonnes) subjected to an explosive attack. These results will be used to develop improved estimates of the radiological consequences resulting from the sabotage of spent fuel transports in urban regions.

EXPERIMENTAL PROCEDURE

Experiments subjecting 1/4-scale fuel casks containing depleted UO<sub>2</sub> fuel to explosive attacks were performed to provide data addressing the operational and analytical difficulties that could be encountered in a full scale test of a spent fuel transport subjected to an explosive attack. In addition, these tests provided useful experimental data characterizing the aerosol and debris released from a spent fuel cask subjected to an extreme

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malevolent environment. The details and results of these scaled experiments are reported in the literature.<sup>2</sup>

A full scale test subjecting a 25.45 metric tonne generic truck cask containing a section of a single surrogate PWR spent fuel assembly to a reference full scale high explosive device (HED) was performed to corroborate the results of the scaled experiments. The surrogate fuel assembly consisted of a 15 x 15 array of 1.2 m long zircalloy tubing filled with depleted  $UO_2$  fuel pellets. The dimension and mass of the  $UO_2$  pellets simulated those of fresh reactor fuel pellets. The stainless steel/lead shipping cask was placed inside a 3.1 m ID x 0.02 m wall x 6.1 m L cylindrical chamber for aerosol containment. The HED was mounted and detonated external to the chamber. An explosive isolation valve placed between the HED and a chamber port was designed to close milliseconds after detonation in order to prevent release of the source aerosol and fragments to the surrounding area. Eleven sampling ports penetrated the chamber at various locations. These 2.5 cm ID sampling ports were closed before and during detonation and were opened shortly after detonation in order to allow sampling of the aerosol.

A 61 cm cubical fiberglass filter was connected to the chamber via a 15 cm ID port to vent the high pressure gases from the chamber to the atmosphere. The filter was designed to handle the high pressure shock wave and a flow rate of 31  $m^3$  per minute. The filter had a filtration efficiency of 99.99% for a 0.3  $\mu m$  monodisperse aerosol. After the test, the filter was washed in a HCl acid solution and analyzed for uranium using uranium fluorometry.

A battery of instruments listed in Table I was selected to sample and characterize the released particles and/or aerosols. The sampling procedure was designed to provide a history of the parameters of the high-velocity particles as well as the lower-velocity aerosols in the chamber. From this history, calculations could be performed to determine the initial release parameters such as initial aerosol mass concentration and released fuel mass. Aerosol size parameters as a function of time were determined from cascade impactor samples obtained at incremental time intervals after HED detonation. Similarly, filter samples provided a time history of the change in elemental mass concentration. Changes in particle morphology and elemental composition as a function of time were determined using sequential electrostatic precipitator (ESP) samples. Scanning electron microscopic (SEM) and energy dispersive spectroscopic (EDS) analyses of the ESP samples provided information on particle size, morphology, and elemental composition. Electrical aerosol analyzers provided information on size distribution for particles smaller than 1.0  $\mu m$  aerodynamic diameter.

Time-resolving rotating plate aerosol samplers were used to collect the high-velocity particles for examination by SEM and EDS. These samplers consisted of a collector plate which mechanically rotated behind a cover plate containing two 3 mm x 4 mm rectangular sampling ports. Three millimeter diameter copper wire discs were placed 6 mm apart on an inside circular path and

Table I

## Sampling Instruments Used in Full Scale Cask Test

<u>Instrument and Applicable Size Range</u>	<u>Purpose of Samples</u>	<u>Analytical Method</u>
Cascade impactor 0.5 - 12 $\mu\text{m}$	Aerodynamic size, geometric standard deviation of total aerosol mass and $\text{UO}_2$ mass.	Gravimetric (Cahn microbalance) and fluorometry for determination of uranium.
Point-to-plane electrostatic precipitator 0.01 - 12 $\mu\text{m}$	Particle morphology, count distribution and elemental distribution.	Transmission and scanning electron microscopy and energy dispersive x-ray analysis.
Filter 37 mm, sequential, 0.01 - 12 $\mu\text{m}$	Provide a time history of total aerosol and uranium mass after HED detonation. Samples for surface area measurements.	Gravimetric for total mass and uranium by fluorometry, nitrogen absorption for surface area measurements.
Filter 37 mm, front surface reentrant filter (FSRP) 0.01 - 12 $\mu\text{m}$	Sequential filter samples from filters inside chamber to compare with filters obtained by extractive techniques to address aerosol line losses.	Gravimetric for total mass of aerosol and uranium by fluorometric techniques.
<sup>a</sup> Condensation nuclei counter (CNC) 0.001 - 12 $\mu\text{m}$	Total count of aerosol particles vs. time after detonation.	Optical light scattering instrument.
<sup>a</sup> Electrical aerosol analyzer (EAA) Dia < 1.0 $\mu\text{m}$	Size distribution parameters for particles between 0.01 and 1.0 $\mu\text{m}$ .	Electrical mobility.
Sieves (38 - 2000 $\mu\text{m}$ )	Provide size distribution data on larger particles of surrogate spent fuel.	Mechanical sieving followed by weighing for total mass and fluorometry for uranium determination.
Integrated time deposition aluminum planchets (0.01 $\mu\text{m}$ to 100 $\mu\text{m}$ )	Provide size, morphology, and elemental composition data of particles diffusively and explosively deposited on chamber surfaces.	Scanning electron microscopy (SEM) and energy dispersive x-ray (EDS) analyses ( $Z > 11$ ) of deposited particles.
Rotating Plate Samplers (RPS): discrete time (0.01 $\mu\text{m}$ to 100 $\mu\text{m}$ )	Provide size, morphology and elemental composition data on explosively deposited particles.	Scanning electron microscopy (SEM) and energy dispersive x-ray (EDS) analyses ( $Z > 11$ ) of deposited particles.

12 mm apart on an outside circular path of the rotating collector plate. The grid spacing allowed sampling at 200 ms intervals. The sampling grids were removed after the test for examination using SEM and EDS analyses. The problem of diffusively deposited particles covering and obscuring the early time deposits was solved by pressurizing the housing with air so that the gas flow exiting the sampling ports permitted entry of early-time high-velocity particles but prevented entry of the lower-velocity aerosols. In addition to these surface deposition samplers, time-integrated deposition aluminum planchets were placed at various locations in the test chamber to sample particles deposited over long periods of time. The aluminum planchets provided information on size, morphology and elemental composition of diffusively and explosively deposited particles.

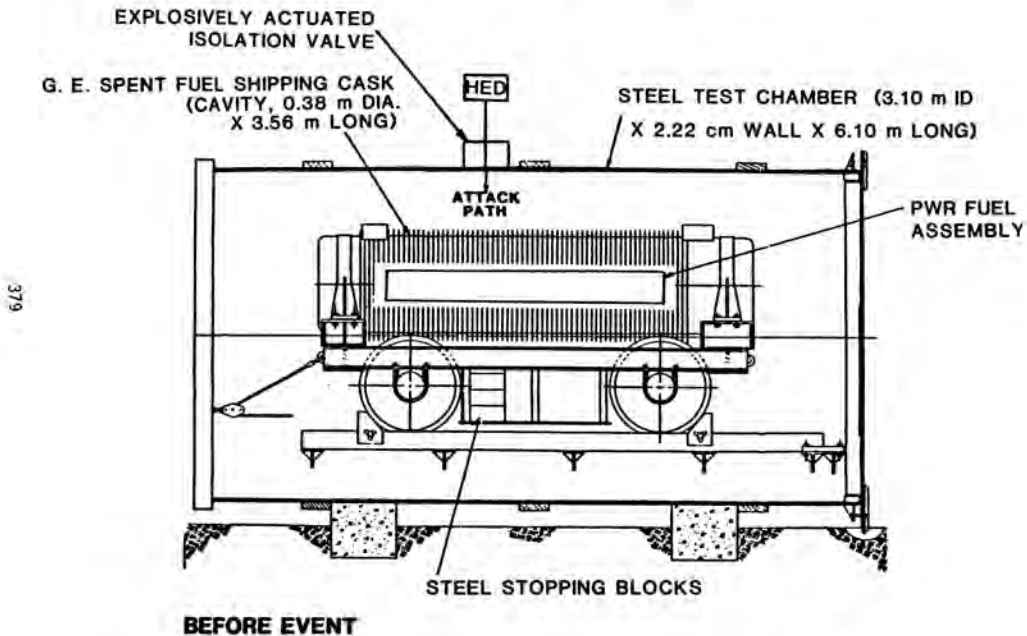
After the test, all debris were collected from surfaces inside the test chamber and separated into obvious non-uranium containing material and suspected uranium containing material. All suspected uranium debris was sieved, the mass determined, and uranium fluorometric analyses performed. The uranium dioxide fuel and zircalloy clad mass were measured before and after the test and the quantity of  $UO_2$  mass removed as a result of the high energy event determined.

## RESULTS AND DISCUSSION

A full scale stainless steel/lead cask containing a 15 x 15 array of 1.2-meter-long zircalloy clad  $UO_2$  fuel pins was subjected to a high explosive attack in a large cylindrical chamber. Figure 1 is a schematic of the set-up prior to detonation. A total  $UO_2$  fuel mass of 201.053 kg was measured before the event. Figure 2 is a reconstruction of the test configuration immediately after detonation. A total  $UO_2$  fuel mass of 195.593 kg remained in the cask after the event which indicated that approximately 5.460 kg of  $UO_2$  fuel was removed from the fuel assembly as a result of the event. Figure 3 shows a time history of the  $UO_2$  aerosol mass within the chamber based on sequential filter samples. A maximum  $UO_2$  aerosol mass concentration of 23.8  $\mu\text{g}/\text{liter}$  was detected at two minutes post detonation. Using the containment chamber volume of 42.29  $\text{m}^3$  and assuming a uniform spatial concentration, a total released  $UO_2$  aerosol mass of 1.008 g was calculated. Another 1.93 g of  $UO_2$  was collected by the pressure relief fiber glass filter. Assuming that one hundred percent of the measured  $UO_2$  aerosol mass is in the respirable size range, a total respirable  $UO_2$  mass of 2.94 g was released from the cask as a result of the event.

The swept and vacuumed debris deposited on the chamber surfaces were sieved and analyzed using uranium fluorometry to determine the size distribution of the larger non-airborne  $UO_2$  particles. Figure 4 shows the cumulative  $UO_2$  particle size distribution of the

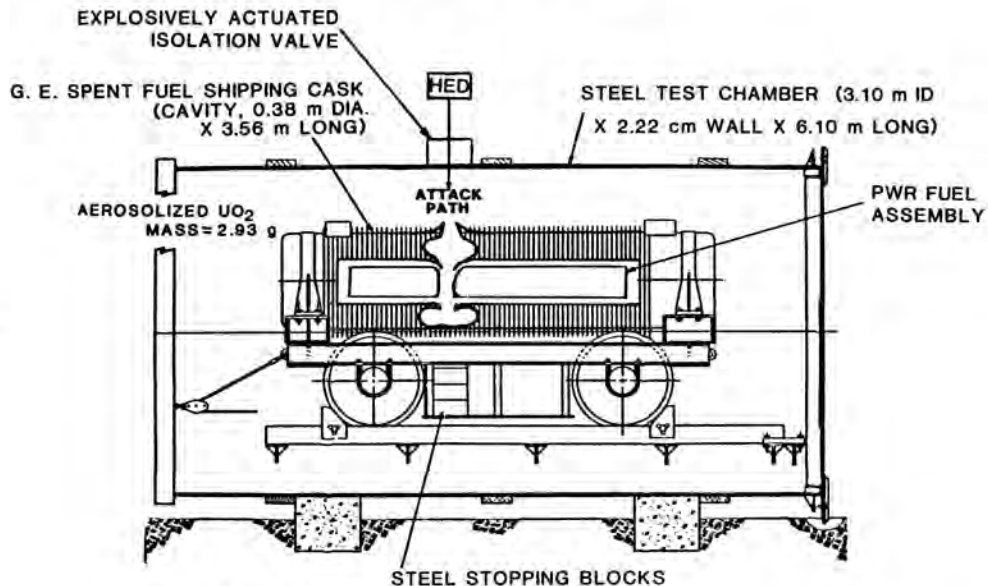
## FULL SCALE TEST



258.048 kg FUEL PINS: 201.053 kg  $\text{UO}_2$  + 56.995 kg Zr-4

Fig. 1. Schematic of full scale test set-up prior to detonation.

## FULL SCALE TEST



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### AFTER EVENT

REMOVED  $UO_2$  MASS : 5.460 kg

REMOVED Zr-4 MASS : 2.063 kg

Fig. 2. Schematic of test configuration immediately after detonation.

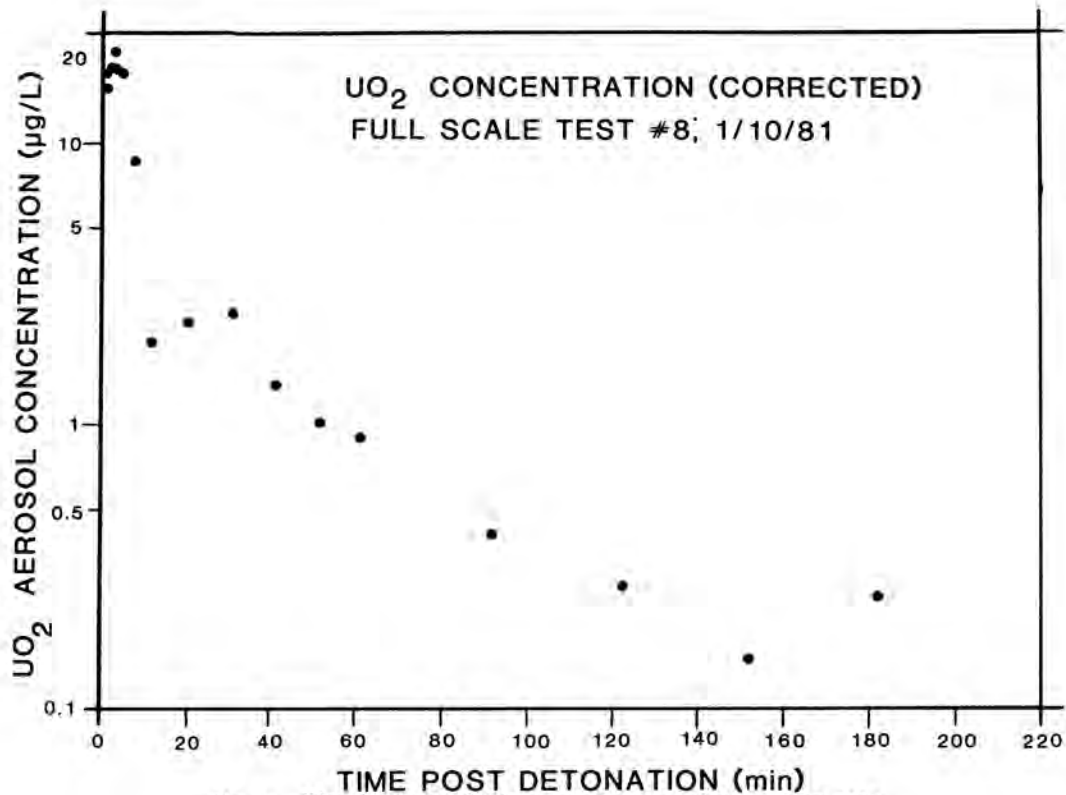


Fig. 3. Time history of UO<sub>2</sub> aerosol mass released from cask based on sequential filter samples.

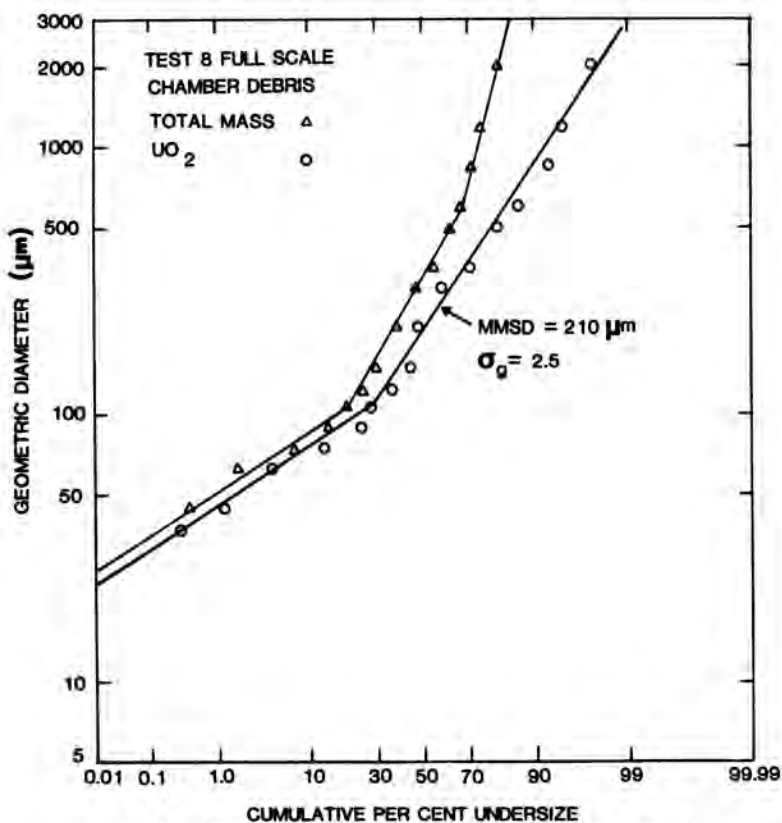


Fig. 4. Cumulative UO<sub>2</sub> particle size distribution of sieved debris collected from chamber surfaces.



sieved debris. Extrapolation of the size distribution to the respirable regime indicates that less than  $10^{-3}$  % of the surface deposited  $UO_2$  debris was smaller than  $10 \mu m$  aerodynamic diameter. The collected chamber surface debris contained approximately 540 g of  $UO_2$ . Thus less than 5 mg of  $UO_2$  particles deposited on the chamber surfaces were smaller than  $10 \mu m$  aerodynamic diameter.

Measurements were made to determine the quantity, particle size and morphology of the explosively deposited uranium particles on chamber surfaces. Time-resolving-rotating-plate samplers (RPS) were placed at various points on the chamber's surface to collect the high-velocity particles at early times (milliseconds after detonation) and time-integrated aluminum deposition planchets were placed at various locations in the chamber to collect the diffusively-driven as well as the high-velocity particles.

Figure 5 shows a SEM photograph of a typical RPS sample taken 400 ms after detonation. High-velocity particles were collected on this sample. No uranium particles and/or coating was detected on this sample or any of the other RPS samples analyzed. However, numerous spherical globules of stainless steel (Fe, Cr, Ni) ranging in size from submicron to tens of microns were found on the copper substrates. In addition, networks of submicron lead fibers and "whisks" of lead were detected on the surfaces of the stainless steel globules and the copper substrate surfaces.

Figure 6 shows a SEM photograph of a time-integrated aluminum planchet sample. This planchet collected both high and low-velocity material during and after detonation. Fractured chunks of uranium ranging in size from 132 to several hundred micrometers aerodynamic diameter were detected using EDS. The morphology of the uranium particles was irregular and was indicative of a mechanical fracturing mechanism. The aluminum substrate was covered with stainless steel (Fe, Cr, Ni) globules and both the stainless steel globules and uranium particles were covered with a network of submicron lead fibers and/or coating. The lead coating has the appearance of having been deposited in a vapor state. The stainless steel globules have the appearance of having been deposited in a molten state.

The analyses of the RPS and time-integrated aluminum planchet samples suggest the following: (1) a lead vaporization/condensation aerosol formation mechanism, (2) a stainless steel melting/solidification aerosol formation mechanism, and (3) a  $UO_2$  nonthermal mechanical fracture aerosol formation mechanism.

Using standard precision measure propagation techniques, the uncertainty of the measured respirable  $UO_2$  mass released from the cask was calculated. Uncertainties of the measured parameters such as mass concentration, chamber volume, and mass fraction of uranium determined by fluorometric spectroscopy were estimated and propagated to determine the uncertainty of the derived  $UO_2$  respirable released mass. The estimated error of the released  $UO_2$  aerosol mass is  $\pm 7$  %.

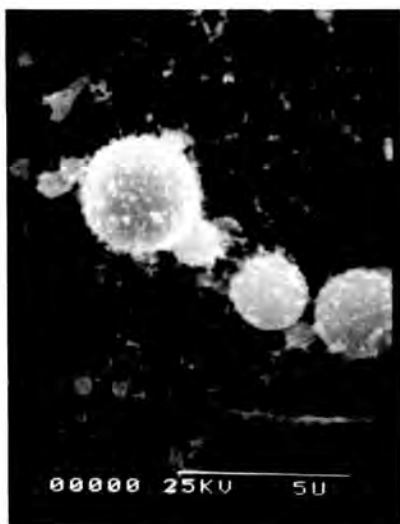


Fig. 5. SEM photograph of a typical Rotating Plate Sample taken 400 ms after detonation.



Fig. 6. A SEM photograph of a time-integrated aluminum planchet sample showing a 70  $\mu\text{m}$  geometric diameter  $\text{UO}_2$  particle.

## CONSEQUENCE ANALYSIS

Although the surrogate spent fuel assembly used in this experimental study was one-third the length of an actual PWR reactor fuel assembly, the damaged or affected length was localized to approximately 6.2% of the total length (1.2 m) of the tested fuel assembly. Indeed, for the reference HED used in this study, the removed fuel mass and/or damaged volume is independent of the fuel assembly length for assembly lengths greater than 0.3 m. The fraction of the solid inventory of a single PWR spent fuel assembly truck cask released as respirable radioactive material was calculated based upon the measured  $UO_2$  aerosol mass of 2.94 g and the equivalent heavy metal inventory of a single spent fuel assembly truck cask (0.5 metric tonnes). Based on these considerations, the release fraction was calculated to be  $6 \times 10^{-4}$  percent.

The expected health consequences were calculated using the measured release fraction of  $6 \times 10^{-4}$  percent for each of the radionuclides shown in Table II. The spent fuel radionuclide inventory used for this analysis has been generated using the full burnup code ORIGIN<sup>3</sup> assuming light water reactor fuel with 33,000 MWd/t of heavy metal burnup at 40 kW/kg power density and 150 days cooling. The truck-mounted cask is assumed to contain radionuclides equivalent to 0.5 t of heavy metal charged to the reactor. Consequence estimates have been generated using the measured source term characteristics (respirable released mass, particle size, etc.) as the primary input to the consequence reactor safety model called CRAC.<sup>4</sup> The detailed population distribution employed in this model is equivalent to the Manhattan borough of New York City (approximately 16,000 people per square kilometer). The total population distribution used in this model closely approximates the actual population within 800 kilometers of the assumed release point. One hundred sequences of New York City weather conditions representative of weather near the release point were used in these calculations. The estimated time of release was midafternoon and a street intersection was the assumed release point. A thermal source in CRAC was used to account for the effects of high explosives lofting the material (thus reducing the close ground level concentrations). All the consequence estimates have been made with the population in place. No attempt was made to model or account for evacuation to avoid early exposure.

Table II

Spent Fuel Cask Radionuclide Inventory  
0.5 MTHM Charged to Reactor (1 assembly)  
33,000 MWd/MTHM Burnup at 40 kW/kg  
150 days Cooling

Radionuclide	Curies
Co-58	$1.09 \times 10^3$
Co-60	$4.17 \times 10^2$
Kr-85	$5.25 \times 10^3$
Sr-89	$7.48 \times 10^4$

RadionuclideCuries

Sr-90	$4.01 \times 10^4$
Y-90	$4.01 \times 10^4$
Y-91	$1.14 \times 10^5$
Zr-95	$1.81 \times 10^5$
Nb-95	$3.37 \times 10^5$
Rn-103	$5.79 \times 10^4$
Ru-106	$1.94 \times 10^5$
Te-127	$2.94 \times 10^3$
Te-127m	$3.00 \times 10^3$
Te-129	$1.23 \times 10^3$
Te-129m	$1.93 \times 10^3$
Cs-134	$1.20 \times 10^5$
Cs-136	$1.10 \times 10^1$
Cs-137	$5.32 \times 10^4$
Ba-140	$2.79 \times 10^2$
La-140	$3.21 \times 10^2$
Ce-141	$3.71 \times 10^4$
Ce-144	$4.57 \times 10^5$
Pr-143	$4.23 \times 10^2$
Nd-147	$2.99 \times 10^1$
Np-239	$1.01 \times 10^1$
Pu-238	$1.48 \times 10^3$
Pu-239	$1.55 \times 10^2$
Pu-240	$2.29 \times 10^2$
Pu-241	$5.78 \times 10^4$
Am-241	$6.78 \times 10^1$
Cm-242	$8.73 \times 10^3$
Cm-244	$1.46 \times 10^3$

Table III shows the results of the CRAC calculation in total latent cancer fatalities, early morbidities and early fatalities. Because the source terms used never produced the threshold dosage for early fatalities and early morbidities, the number of early fatalities and morbidities predicted are zero. The latent cancer fatalities are a result of initial exposure, particle resuspension and long-term exposure to contaminated ground. The results of the study indicate that one peak latent cancer fatality and 0.19 mean latent cancer fatalities are possible from an explosive attack of a single PWR fuel assembly truck cask in downtown New York City.

Table III

CRAC Code Results for this Experimental Study <sup>a</sup>	
Early Fatalities	0
Early Morbidities	0
Total Latent Cancer Fatalities (mean/peak)	0.19/1

<sup>a</sup>Based on measured release of  $6 \times 10^{-4}$  percent.  
 Early fatalities: within 1 year after exposure  
 Early morbidities: illnesses within weeks after exposure.  
 Latent cancer fatalities: anytime after exposure as a result of initial exposure (also long term exposure to low levels of contamination).  
 Population to 800 kilometers radius.

Peak thyroid and bone marrow dose in rems was also calculated as a function of distance from the release point. At a distance of 30 m from the release point, the peak bone marrow dose was calculated to be 28 mrem and the peak thyroid dose was calculated to be 19 mrems. At 1.6 kilometers from the release point, the peak bone marrow dose was calculated to be 0.5 mrems and the peak thyroid dose was calculated to be 0.3 mrems. The Protective Action Guide (PAG) threshold for these distances is 1 rem. The peak bone marrow and thyroid dose for distances of 30 meters or more from the release point are significantly less than the PAG threshold.

#### SUMMARY AND CONCLUSIONS

A full scale test subjecting a 25.45 metric tonne stainless steel/lead cask containing surrogate spent fuel to a reference high explosive device was performed to provide data for developing improved estimates of the radiological consequences resulting from the sabotage of spent fuel transports in urban regions. A total  $UO_2$  fuel mass of 5.460 kg was removed from the 201.053 kg  $UO_2$  fuel assembly as a result of the test. A total respirable  $UO_2$  aerosol mass of 2.94 g was released from the cask as a result of the explosive attack. A total of 540 g of  $UO_2$  particles were deposited on the chamber surfaces. Less than 5 mg of the surface deposited  $UO_2$  particles were smaller than 10  $\mu m$  aerodynamic diameter. These results are in good agreement with the time-integrated aluminum planchet samples which showed trace amounts of mechanically fractured uranium particles deposited on the chamber surfaces. Analyses of the time-resolving RPS indicated that no uranium particles and/or vapor were explosively deposited on the chamber surfaces.

The results of this full-scale test indicate that approximately  $6 \times 10^{-4}$  % of the total solid heavy metal inventory (0.5 t) could be released as a respirable radioactive aerosol as a result of an explosive attack on a single-PWR fuel assembly truck cask. The measured respirable release fraction has been determined to within  $\pm 0.4 \times 10^{-4}$  % using standard error propagation techniques; however, release fractions for the volatile fission products will be larger than that for the actinides. Experimental studies are currently underway to provide data characterizing the release of the volatile fission products.

The expected health consequences were calculated using the derived release fraction of  $6 \times 10^{-4}$  percent as the primary input to the consequence reactor safety model called CRAC.<sup>4</sup> The release conditions such as population distribution and weather conditions were assumed to be equivalent to those of the Manhattan borough of New York City. The results of this consequence analysis indicate that one peak latent cancer fatality and no early fatalities or early morbidities could occur as a result of this postulated radioactive release from a single PWR spent fuel truck cask in downtown New York City.

#### REFERENCES

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