

## **Sensor Systems for Precise Location of Depleted Uranium in Soil and for Enhancing the Recovery of Both Zero Valence and Uranium Oxides**

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### **ABSTRACT**

Depleted uranium (DU) has been the primary material used for the past two decades by the US military in armor piercing rounds. Domestic firing ranges that have been used for DU training purposes are located around the country and vary with regard to soil type, depth of vadose zone, and extent of contamination with other types of projectiles. A project is underway to develop a set of sensor systems to locate expended DU rounds and to process soil and debris to recover the material. Reactivity of zero valence DU material, even in dry sandy soils, results in rapid oxidation and diffusion of uranium minerals within the soil column. Detection techniques must be robust for both metallic and uranyl species. Radiological sensor techniques including both gamma spectroscopy and prompt gamma neutron analysis are being used in conjunction with electromagnetic imaging to locate the DU for excavation. Detection limits for both zero valence DU (ZVDU) and oxidized material will be discussed. Applicability of active and passive optical methods, such as spectral imaging and fluorescence spectroscopy, will be discussed as aids for achieving clean soil margins while excavating DU materials. Instrumentation selection for controlling processing equipment used to separate ZVDU and uranyl species from contaminated soil and debris will also be discussed. Preliminary findings for use of sodium iodide detectors and multichannel analyzer software are discussed for locating 25 and 105 mm DU penetrators. Optimum detector height of 15 cm (six inches) and detection depths up to 15 cm are discussed. A comparison of detector response of the Geonics EM61 MKII electromagnetic induction unit for DU and ferrous materials is reported. Difficulty of locating small DU penetrators using the one meter detection coil and differences in detector response for target orientation relative to the detection coil are reported.

### **INTRODUCTION**

The US military has employed a variety of dense metal alloys as armor piercing projectiles since the Second World War. These have included tungsten carbide, an improved tungsten alloy, a four-component alloy containing depleted uranium, and a depleted uranium-titanium alloy (DU). Use of DU as an armor piercing round has become the standard since the 1980s and is currently the superior material for that application.

At least fourteen different locations in the US have been used by the military for testing and training purposes with DU ammunition. The sites vary widely in amount of DU munitions that have been expended. White Sands Missile Range, where a limited number of missiles were fired

in 1976, has no more than a few hundred kg of DU in its ranges. This is compared to over 100,000 kg being expended at both Los Alamos National Laboratory and Eglin Air Force Base.

The contaminated areas at each of these test sites vary in size, based on how targets were arranged. Catch boxes with a footprint of approximately one acre have been constructed for certain applications. There are also open firing ranges that have been used for both artillery and aircraft that can be as large as thousands of acres.

The ability to defeat armor using kinetic energy penetrator rounds constructed using depleted uranium represents a tactical advantage to the U.S. military on the battlefield. It is critical to maintain the capability to train with currently developed DU munitions and to test DU rounds undergoing improvement or development. The location, removal, and/or containment of DU residues generated during test and training operations is an integral part of range management.

### **DEPLETED URANIUM SENSING AND TREATMENT FOR RECOVERY**

The U.S. Department of Defense (DoD) has committed to recovery of expended DU materials from testing and training ranges. This recovery process will be somewhat complicated by the fact that metallic uranium is fairly reactive, rapidly oxidizing once it has been deposited in soil. Current intentions are to recover the majority of material, either metallic DU or corrosion products of the projectiles, as opposed to conducting a more complete decontamination of the impacted areas. This recovery process will employ a variety of measurement systems for precisely locating the DU material, facilitating recovery of the DU with a minimal quantity of contaminated soil, and controlling process equipment used to separate the DU from soil and associated debris.

As a part of the Depleted Uranium Sensing and Treatment for Removal (DUSTR) initiative, the Institute for Clean Energy Technology (ICET) at Mississippi State University (MSU) has inaugurated its efforts by reviewing the available literature before selecting techniques for development and deployment. The DUSTR initiative was established for the purpose of developing technologies for the sensing of depleted uranium munitions residues in soils and water and for the physical separation of depleted uranium from soils/water at test and training facilities. The ability to locate, remove, and/or contain DU following use on training ranges without generating large quantities of waste is critical to the sustainable operation of testing and training facilities.

DoD will benefit from DUSTR by the tools developed under this program for maintaining sustainable test and training facilities for munitions that contain depleted uranium. Developers, testers, and military personnel will benefit from the development of sensing, removal, and/or containment technologies that will provide a critical evaluation of munitions design, lethality, and use requirements. In addition, this program will reduce human and environmental risks by developing and applying scientifically defensible low cost range maintenance practices.

The UN reports on efforts of scientists to evaluate the potential for exposure to DU material both in Kosovo [1] and in Montenegro and Serbia [2] are similar with respect to the field methods employed by the teams of professionals and their findings. Both field studies utilized a variety of handheld beta and gamma measurement instruments to locate DU penetrators, jackets, and areas

of soil contamination. A magnetometer was also included in the suite of field instruments to screen for unexploded ordinance (UXO) and, possibly, DU.

While these reports do not detail a great deal of success using handheld meters, they do contain a great deal of information of significance to the DUSTR efforts. Both teams were able to identify impact locations for DU projectiles by discoloration of the surface of soil and structures. These impact areas were typically small and variable in size with dimensions ranging up to 20 x 20 cm or even one meter in diameter.

The Kosovo [1] and Montenegro/Serbia [2] field studies also included collection of soil samples to verify DU concentrations in soil via laboratory methods. These findings indicated that the DU concentrations in soil ranged from 0.01 to 0.1 g DU/kg soil. Soil contamination by DU penetrators was detected to depths of 20 cm.

Conclusions reached by the UNEP teams included the general failure of handheld beta and gamma survey instruments to locate widespread DU contamination in soil. Contamination that is detectable by such instruments is limited to the upper 20 cm of soil. While it is unclear how many penetrators were missed by the surveys, it is clear that only a very small fraction of the fired penetrators were found and/or recovered.

The DUSTR objective is the “recovery of DU materials from firing ranges and catch boxes so as to sustain activities at these sites.” Measurement technologies needed to achieve this objective fall into three categories. These are (1) sensors used for site screening to locate expended DU penetrators or their oxides, (2) real-time measurement devices to facilitate retrieval of DU penetrators and oxides with a minimum of uncontaminated soil, and (3) instrumentation that can effectively control process equipment to optimize the separation of DU metal and oxides from contaminated debris.

The DUSTR effort is employing a comprehensive characterization effort, based upon the reality that no single characterization technique is able by itself to provide precision location capability in all circumstances and that by combining results from different techniques, the amount of false

**Table 1.** Listing of measurement technologies by categories that have been considered for employment by the DUSTR effort.

Measurement Type	Specific Technology
<b>Optical Methods</b>	Spectral Imaging
	Long Wavelength Infrared
	Laser-induced Fluorescence
	Induced Fluorescence
	X-Ray Fluorescence
<b>Radiological Methods</b>	Scintillation Detectors
	Semiconductor Detectors
	Xenon Detectors
<b>Geophysical Methods</b>	Soil Conductivity
	Electromagnetic Imaging
	Ground Penetrating Radar
	Seismic Methods

positives and the amount of uncontaminated soil process can be minimized. Measurement technologies that have been considered can be generalized into three categories: (1) optical and imaging techniques, (2) radiological techniques, and (3) geophysical techniques. Table 1 lists the individual techniques within each category that have been considered.

A review of the scientific literature did not produce a wealth of data produced by application of the numerous methodologies to location of DU in the three areas of measurement applications. However, we did identify a few reports that discussed use of a few of the techniques in locating DU material. Our findings are capable of classifying a measurement technique into one of three categories with respect to a specific application. For instance, we can conclude that spectral imaging has more potential for rapid screening of a large area than does gamma ray spectrometry. With this example in mind, we will now describe what we believe is the optimum approach for detecting and monitoring the processing of spent DU munitions.

**Site Screening to Locate DU Materials** will be the most technically challenging set of measurements to successfully make. This will likely be accomplished by an ensemble of activities that can be sub-divided into rapid site screening, methodical site screening, and precision locating of DU material. Each of these types of activities will likely involve different measurement technologies.

**Rapid site screening** will best be accomplished using optical techniques, such as spectral imaging or fluorescence imaging. The Kosovo report [1] specifically identifies surface staining (yellow uranyl salts on the ground surface resulting from skips or penetrations) as a major facilitator to locating DU materials. Imaging techniques can be deployed using

helicopters and used to rapidly screen large amounts of surface area in a day's time to identify hot spots for more detailed investigation. No other techniques appear capable of providing equivalent rapid characterization of an area.

**Methodical site investigation** will involve a significantly slower rate of screening activity, possibly only an acre or two per day. This more rigorous investigation needs to provide more than a surface characterization of the site. The most likely candidates for accomplishing this task will be radiological measurements (either passive gamma spectrometry or prompt gamma neutron activation analysis) and a complementary geophysical technique (electromagnetic imaging with magnetometry). These methods will allow discrimination between DU and non-DU materials in the shallow sub-surface (20 cm) and identify metallic materials to significantly greater depths (1+ meters). The combination of these techniques will also be useful in discriminating between zero valence DU (ZVDU) and oxidized DU.

**Precision locating of DU materials** may or may not be necessary prior to initiating excavation of the material. However, it is conceivable that a more precise locating of the DU penetrators may lead to a significant reduction in the amount of contaminated soil excavated. The most likely techniques for accomplishing this are radiological measurements using push technologies and/or a geophysical technique like ground penetrating radar to detect the track left by the penetrator.

**Recovery of DU materials**, particularly uranyl ( $\text{UO}_2^{+2}$ ) minerals, can be enhanced using optical techniques to minimize the amount of uncontaminated soil removed during excavation. Minimizing the volume of soil exhumed while ensuring that the bulk of DU oxides has been reclaimed will greatly facilitate the recovery process. This does not represent as great a technical challenge as initial locating of the DU material and can likely be accomplished using spectral imaging or induced fluorescence. Either of these measurement techniques can be easily and rapidly employed in the recovery process to "visualize" the contaminated soil and DU mineral plume.

**Table 2.** Ideal performance characteristics of instrumentation for detection and recovery of DU penetrators or their oxides that are present on U.S. military testing and firing ranges.

Parameter	Rapid Type of Site Screening	Site Screening Set	High Resolution Screening	Excavation Enhancement	Reclamation Process Control
Detection Depth/size for a Standard Size Target	(Optical) 1000 cm <sup>2</sup>	Surface to 7.5cm for 25 mm penetrator	Surface to 7.5cm for 25 mm penetrator	Surface to 7.5cm for 25 mm penetrator	
Scan Width/ Detection Area				Evaluate 20,000 cm <sup>2</sup> w/in 2 min	Scalable from 1 cm to 1 m
Scan Speed	> 2500 ha/day	> 420 ha/day	> 21 ha/day	n/a	
Sensitivity to Soil Matrix Effects					
Sensitivity to Variability of Process Stream Density (Absolute vs Differential Measurement)	n/a	n/a	n/a	n/a	
Scalable					
Time over Sample					1 sec
Minimum Detection Limit					500 ppm ZVDU
Spatial Resolution	< 1000 cm <sup>2</sup>	<1000 cm <sup>2</sup>	< 1000 cm <sup>2</sup>	< 1000 cm <sup>2</sup>	
GPS Locating Accuracy	2 meters				
Calibration Frequency	< daily	< daily	< daily	< daily	< daily
Maintenance Frequency	< weekly	< weekly	< weekly	< weekly	< weekly
Terrain Traversing/ Surveying Capability	All terrains				
System Mass	< 200 kg	< 200 kg	< 200 kg	< 5 kg	< 200 kg
Power Requirements	Battery	Battery	115V 30A	Mobile power supply	< 20 kw
Special Requirements Weather Proof					
Vibration Resistance					
Data Interpretation Time					
Daily Turnaround					

Finally, developing an instrumentation system to provide **control parameters for the reclamation process** will not be as straightforward as instrumentation for enhancing the recovery process. However, it will not be as technically difficult as developing a system to rapidly and precisely locate DU materials randomly dispersed over a large area. Based on current knowledge from a review of the literature and likely methodology employed for soil processing, radiological measurements or spectral imaging are the most likely candidates for control instrumentation.

Based upon the above considerations, a list of ideal performance criteria for each of the measurement categories is presented in Table 2.

## RESULTS AND DISCUSSION

### Performance Evaluation Test Bed

In order to evaluate the performance of selected instrumentation under simulated conditions in preparation for ultimate deployment at U.S. military firing ranges, we have constructed a test bed (see Fig. 1). The test bed has an eight by eight feet footprint and is four feet deep (2.44 m x 2.44 m x 1.22 m). Since both the geophysical and prompt gamma neutron activation analysis (PGNAA) methods are sensitive to the presence of metals, the test bed has been constructed of plastic. Metal objects such as desks, cabinets, etc. are easily removed from the laboratory when measurements are made in which they may be problematic. Commercial-grade concrete sand is used to simulate a desert soil.



**Figure 1.** Test bed for evaluating performance of DU sensing technologies.

### Fluorescence Spectroscopy

Uranium metal is pyrophoric and hence DU penetrators fired into the ground tend to oxidize after the protective coating is removed. Most of the resulting oxidized uranium compounds contain the  $\text{UO}_2^{+2}$  moiety. Uranyl ( $\text{UO}_2^{+2}$ ) compounds have long been known to emit characteristic fluorescence in the 450-600 nm spectral region when excited in the ultraviolet or short-wavelength visible region [3]. Hence uranyl fluorescence provides a means of locating DU penetrator residues during site screening, excavation, and during soil processing. The

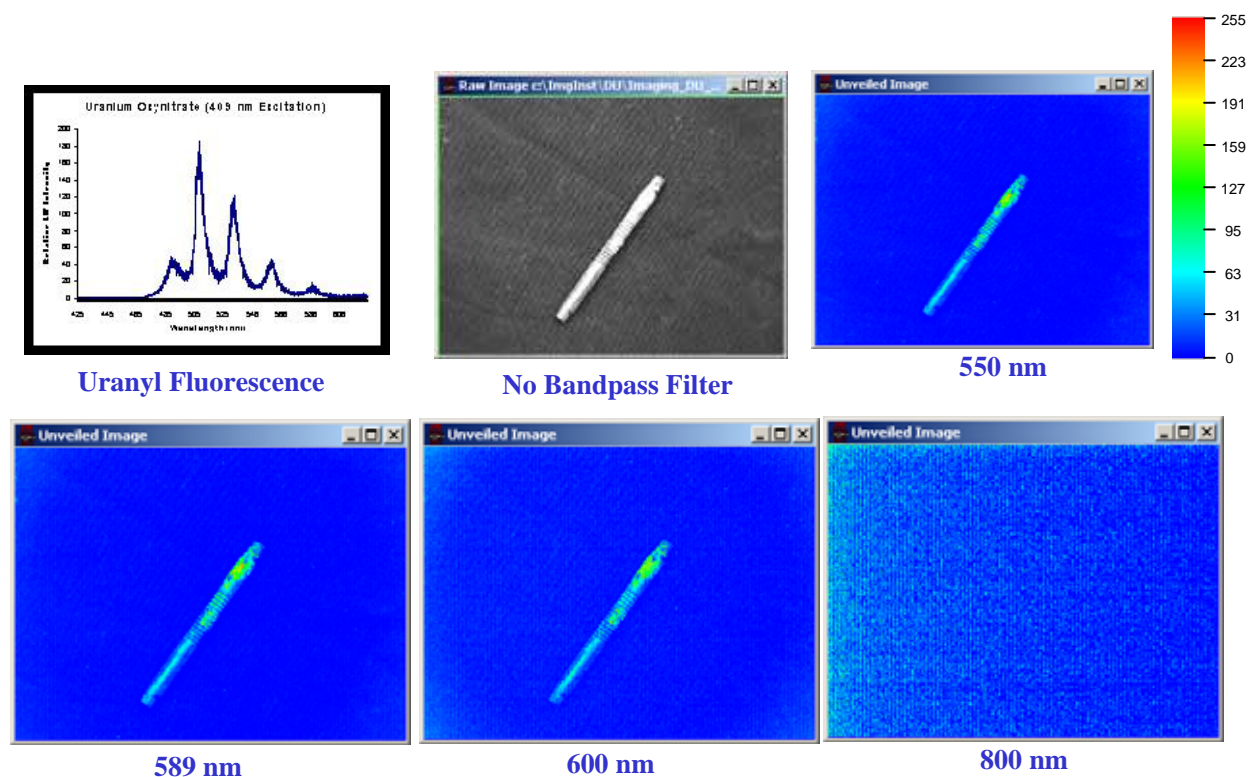
fluorescence can be excited either with a laser or with a high-intensity spectral lamp. Lasers have the advantage of higher intensities and hence the ability to detect lower concentrations; high-intensity spectral lamps (such as mercury lamps) have the advantage of lower acquisition cost and ease of on-the-spot repairs. When fluorescence is combined with spectral imaging, then spatial distribution/location can be readily determined.

A preliminary series of laser-induced fluorescence experiments have been done to investigate the effect of excitation wavelength upon the uranyl fluorescence signal intensity. Three different excitation wavelengths were used: 532 nm, 409 nm, and 355 nm. The 532-nm and 355-nm radiation was produced using a pulsed Nd:YAG laser while the 409-nm radiation was produced by a continuous wave diode laser. The resulting fluorescence pattern (shown in Fig. 2) is similar for both 355 and 409 nm excitation, while for 532-nm excitation, only the 548-nm and longer wavelength emission bands are observed. Emission at 510 nm is the most intense for most (but not all) of the uranyl compounds investigated. The emission intensity with 355-nm excitation is significantly larger than that with 409-nm excitation, but the 409-nm diode laser is much smaller and less expensive than the Nd:YAG laser, and operates using standard 115 VAC electrical power. Experiments have begun using 254-nm radiation from a mercury lamp in order to develop a non-laser uranyl fluorescence detection system.

Figure 2 also contains images of a fired 25 mm DU penetrator. The black image labeled No Bandpass Filter is equivalent to a black and white picture of the penetrator. The other images reveal diffuse spectral patterns in the 550 nm, 589 nm, and 600 nm ranges when the penetrator is exposed to ambient light. The 800 nm image represents a background for image processing within the experimental system. False colors for the penetrator are representative of relative levels of spectral reflectance; the legend at the right provides the relative scale. Images in Figure 2 are representative of data collected for other fired penetrators with nominal visible surface corrosion.

Pseudo-color images are result of image processing. The experimental system included a monochrome Pulnix 745E CCD camera (Pulnix Inc.) and filter wheel system (CVI Laser Corp.) mounted in front of the camera through a customized adapter. The filter wheel system can hold up to five narrow-band interference filters; bandwidths of the filters used in this experiment are 10nm. The CCD camera is controlled by a frame grabber inside a personal computer. Working distance for data collection is approximately 50 cm from the filter wheel housing to the penetrator.





**Figure 2.** Laser-induced fluorescence spectrum of uranyl nitrate excited at 409 nm.

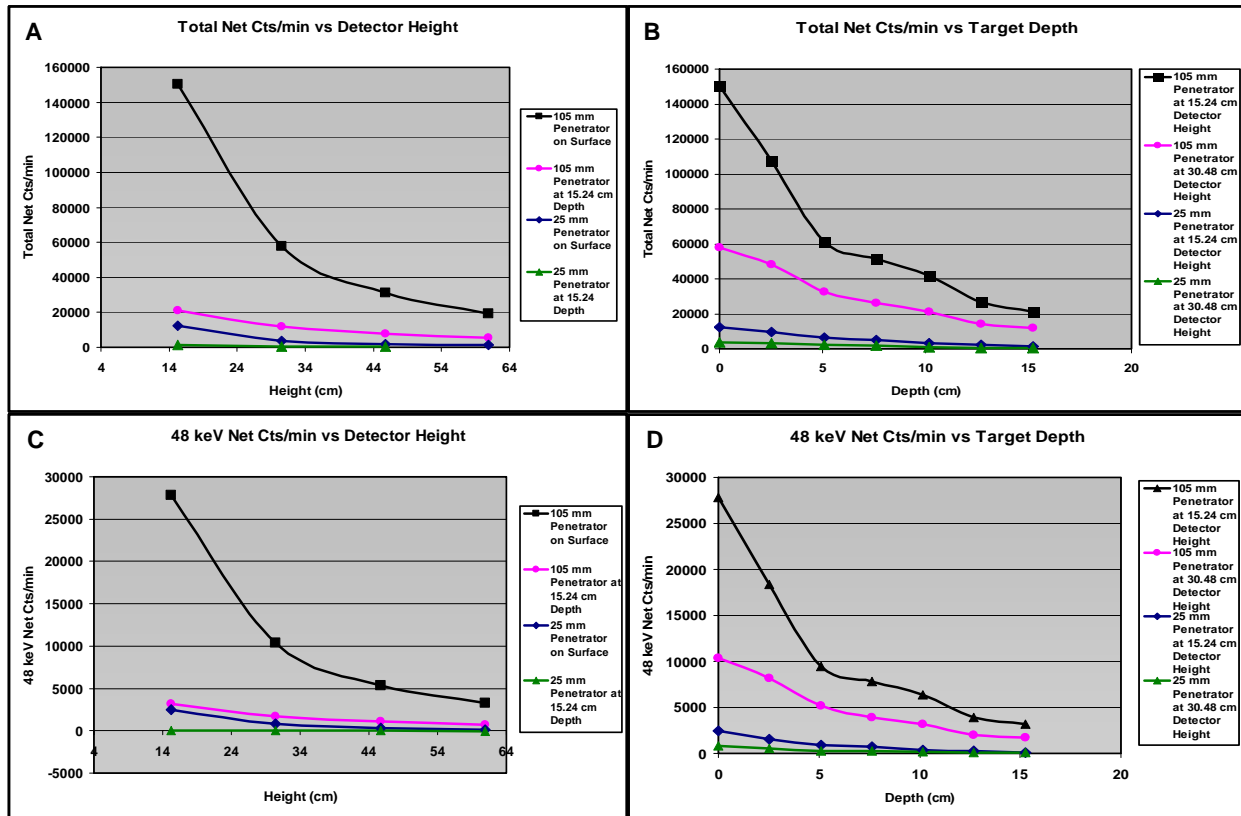
## Radiological Techniques

A preliminary set of data have been collected to establish baseline data and determine the relative effectiveness of sodium iodide detectors for locating different size DU penetrators. Equipment used in this series of testing included Bicron three by three inch (7.6 x 7.6 cm) sodium iodide (NaI) detectors, Ortec Digibase, and Ortec multi-spectral analysis (MCA) software (Maestro and ScintiVision 32).

Data were collected using single 25 mm and 105 mm unfired DU penetrators as targets. Gamma radiation spectra were collected using a single NaI detector at varying detector heights 15, 30, 46, and 61 cm (6, 12, 18, and 24 inches) above the surface of the test bed soil. Data were also collected with the targets buried at varying depths below the soil surface 0, 2.5, 5.1, 7.6, 10.1, 12.7, and 15.2 cm (0, 1, 2, 3, 4, 5, and 6 inches).

Plots included in Figure 3 demonstrate detection trends in counts per minute for 25 and 105 mm penetrators. Figure 3(A) provides the corrected total count rate (actual minus background) for penetrators on the surface of the test bed and at 15.2 cm depth for both 25 and 105 mm penetrators. Detection of the large penetrators at the 15.2 cm depth is very similar to the count rate for 25 mm penetrators on the surface. It can be seen from the plot that a detector height of 15 cm (six inches) is desirable. Figure 3(C) provides equivalent count rates for 48 keV photons.

Figure 3(B) is a plot of total net counts per minute for 25 and 105 penetrators as a function of depth within the test bed. Detection count rates are provided for penetrators on the soil surface and 2.5 cm increments to a depth of 15.2 cm. Count rate curves are provided for each depth with the detector at heights of 15.2 and 30.4 cm (six and 12 inches). Figure 3(D) provides equivalent data of count rates for 48 keV photons. From these curves it can be seen that small penetrators are unlikely to be detected at depths of 15 cm or greater using three by three NaI detectors.



**Figure 3.** Plots of net count rates for 25 mm and 105 mm DU penetrators at various depths using three by three NaI detector at heights above ground surface ranging from 15 to 61 cm (six to 24 inches).

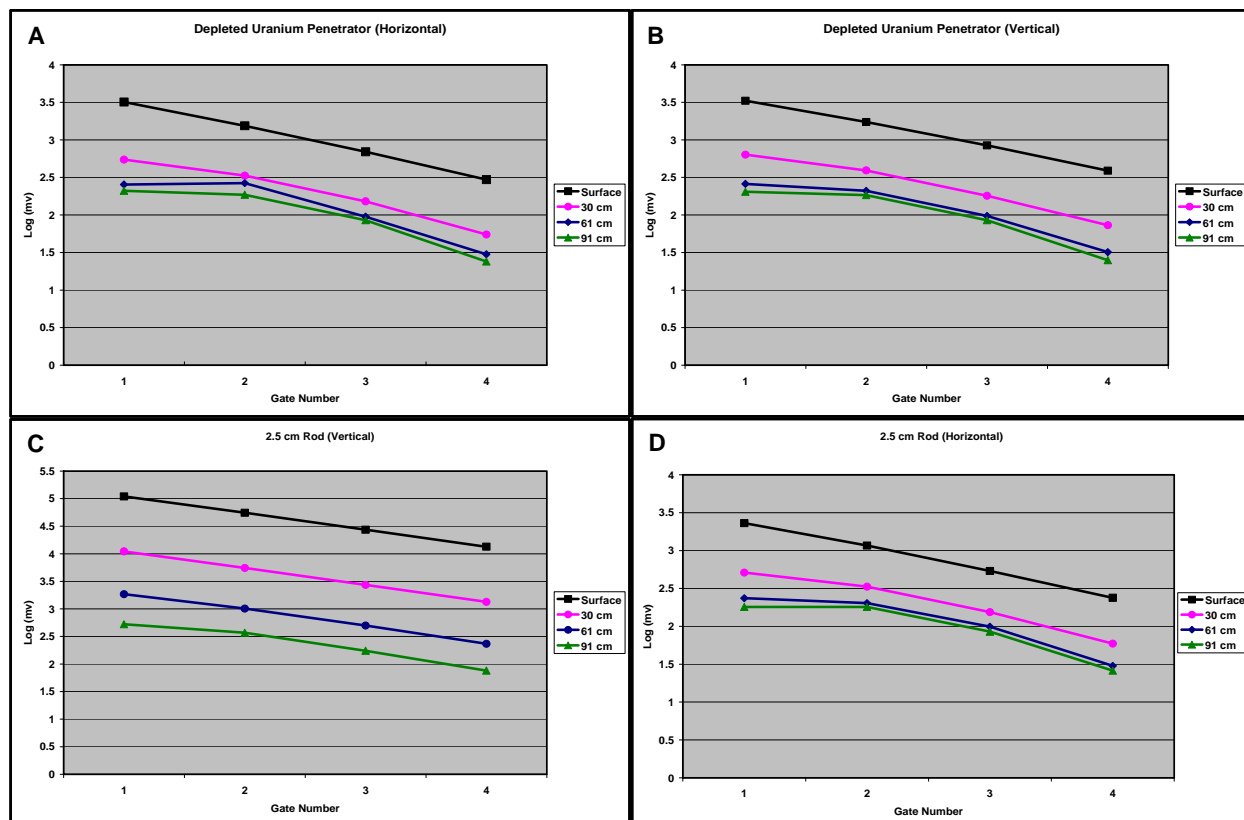
## Geophysical Techniques

The Geonics EM61 MKII is a time domain electromagnetic induction metal detector capable of detecting both ferrous and non ferrous electrically conductive metals. Time domain instruments send an electromagnetic pulse into the ground thereby inducing eddy currents in conductive subsurface metals. As the transmitted field collapses, the instrument monitors the field emanating from conductive subsurface targets as their eddy currents die out. The received signal is a pulse that is a sum of exponential functions which depend upon the material properties and the physical shape and dimensions of the target. The properties of the returned pulse can be used to help determine target depth and are also used to help discriminate between unexploded ordnance and metal “clutter”. The EM61 data is collected and stored during four discrete time gates after the fall of the transmitted pulse. The times at which these gates are positioned are pre-selected by the manufacturer and can not be modified by the user. We are using a High Power (HP) version of the standard EM61 MKII that has been one of the more successful instruments used for UXO

detection. The HP version provides twice the power to the transmitting coil as the standard EM61 MKII. It provides significantly greater depth of detection but also requires a heavier battery pack.

Preliminary testing was done with Depleted Uranium (DU) penetrators to determine their detectability versus depth. All preliminary tests were done with the targets suspended in the air beneath the EM61 transmit coil rather than actually burying the targets in the soil. This is not an unusual way to test time domain instruments. The EM61 starts reading the first time gate at 0.261 milliseconds after the fall of the transmit pulse. The time constant of soil is normally much shorter and its contribution to the received signal has long since died away before any gate is sampled. Although some highly conductive soils may have a small effect on the received signal it is considered insignificant in most cases.

Plots A and B in Figure 4 are the results measured using a 105mm DU penetrator in the horizontal and vertical positions respectively. (Horizontal being parallel to the plane of the transmit coil of the EM 61.) Plots C and D in Figure 4 are the results using a 2.5 cm steel rod 61 cm (two feet) in length. Although these data is preliminary, it appears that the detectability of DU is not as good as for iron. This is expected because the conductivity of uranium is less than that of iron. Also the effect of target orientation versus received signal can be compared for iron and DU. The iron bolt shows a much larger signal when the steel rod is in the vertical position. This is the expected result because in the vertical position the steel rod is perpendicular to the EM field and crosses more field lines than when parallel to the field. The DU penetrator does not seem to respond in the same way.



**Figure 4.** Detector responses of the Geonics EM61 MKII electromagnetic induction unit for DU and ferrous targets at various depths, 30, 61, and 91 cm (1, 2, and 3 ft) and in different orientations relative to the induction coil.

The noise floor for these tests is indicated on Plots A, B, C and D. This high noise level indicates more interference from metals and/or power lines in the area than was desirable. The high level of noise is likely the reason we were unable to detect the smaller DU penetrators from 25mm rounds. The smaller penetrators may be detectable in a less noisy environment.

## CONCLUSIONS

Initial series of tests conducted for this project show promise for application of each of the detection techniques for applications associated with locating fired DU penetrators. Optical methods including both fluorescence and spectral reflectance have potential for rapid screening of soil surfaces. Non-laser excitation methods for fluorescence detection of either DU penetrators or DU oxides on surfaces can offer a rapid screening method for large land masses or contaminated surfaces. Additionally, diffuse reflectance also has potential for this application. Experimentation is currently underway to determine minimum detection limits and optimum survey rates for each of these technologies.

United Nations teams have reported success using gamma detection devices to locate small (25 mm) penetrators and penetrator jackets. An initial series of studies conducted using three by three NaI detectors with MCA software indicates small (25 mm) penetrators can be detected to

depths less than 15 cm. It has also been shown that the desirable detector height is approximately 15 cm above the ground surface for flat terrain. Additional studies are under way to evaluate advanced signal processing techniques to enhance the speed and accuracy at which sites can be screened for DU metal and oxides. Additionally, larger NaI detectors – 10 x 10 x 41 cm (four by four by 16 inch) detectors and thin crystal (Fidler) units sensitive to 13 keV x-rays will also be evaluated. Studies will also include determination of the temperature dependence for each detector type and GIS software will be developed to map radiological measurements to geophysical data.

Little data have been identified in the literature relating the effectiveness of using EMI techniques for locating DU penetrators or discriminating between DU and UXO materials. A first series of tests with the Geonics EM61 MKII reveals a lesser sensitivity of DU penetrator orientation than for ferrous materials. Detectability of small targets is low for small penetrators when using a one meter coil and a smaller (20 cm) coil has been ordered. Current project plans include determination of detection limits for penetrator size as a function of target depth, target orientation, and soil conditions, as well as evaluation of the performance of smaller detection coils.

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