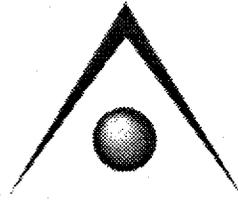


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AMES LABORATORY

Final Report
Ames Mobile Laboratory Project:

**The Development and
Operation of Instrumentation in a Mobile Laboratory
for *In Situ*,
Real-time Screening and Characterization of Soils
Using The
Laser Ablation Sampling Technique**

January 27, 1995

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Technology Integration Program

**AMES LABORATORY (USDOE)
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ABSTRACT

The main focus of the Ames Laboratory's Technology Integration Program, TIP, from May 1991 through December 1994 was the development, fabrication, and demonstration of a mobile instrumentation laboratory incorporating rapid *in situ* sampling systems for safe, rapid, and cost effective soil screening/characterization. The *Mobile Demonstration Laboratory for Environmental Screening Technologies*, MDLEST, containing the analysis instrumentation, along with surface and subsurface sampling probe prototypes employing the laser ablation sampling technique were chosen to satisfy the particular surface and subsurface soil characterization needs of the various Department of Energy facilities for determining the extent of heavy metal and radionuclide contamination.

The MDLEST, a 44 foot long 5th wheel trailer, is easily configured for the analysis instrumentation and sampling system required for the particular site work. This mobile laboratory contains all of the utilities needed to satisfy the operating requirements of the various instrumentation installed. These utilities include, an electric generator, a chilled water system, process gasses, a heating/air conditioning system, and computer monitoring and automatic operating systems. Once the MDLEST arrives at the job site, the instrumentation is aligned and calibration is completed, sampling and analysis operations begin. The sample is acquired, analyzed and the results reported in as little as 10 minutes. The surface sampling probe is used in two modes to acquire samples for analysis. It is either set directly on the ground over the site to be sampled, *in situ* sampling, or in a special fixture used for calibrating the sampling analysis system with standard soil samples, having the samples brought to the MDLEST. The surface sampling probe was used to *in situ* sample a flat concrete surface (nondestructively) with the ablated sample being analyzed by the instrumentation in the MDLEST.

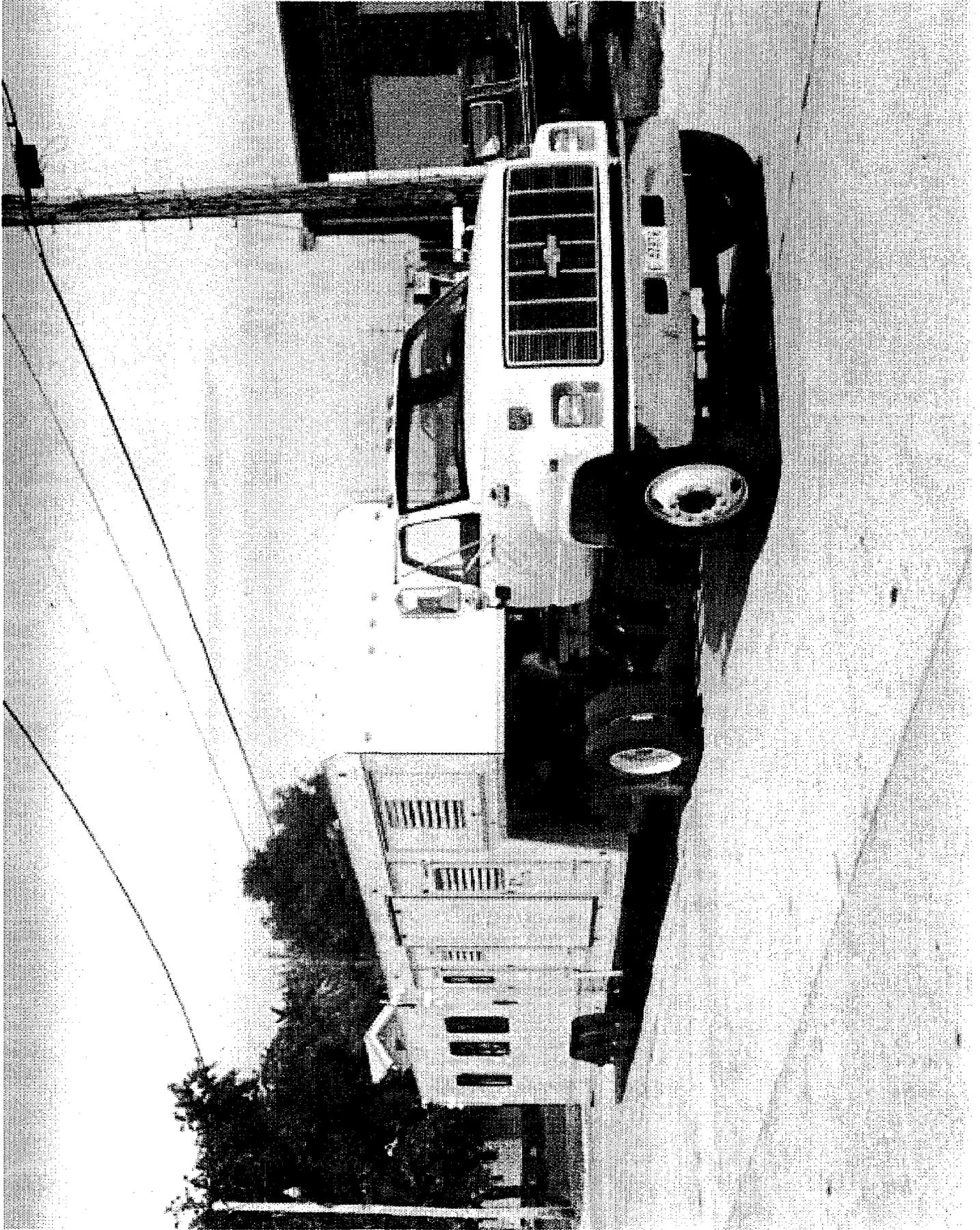
The subsurface probe was developed under a CRADA agreement between TIP and Layne Environmental Services Inc, LESI. This probe is lowered into a drive casing that has been set in the ground and mates with a special tool developed by LESI. The LESI tool has a operable window that allows direct access to the soil for the subsurface probe to directly sample the soil *in situ*. LESI provided the percussion hammer drill rig and operating crew to place the tool in the ground. The subsurface probe was developed by TIP and interfaces to the MDLEST for sample analysis. This probe was designed to operate and obtain samples down to 90 feet.

The MDLEST and laser ablation sampling systems have participated in several field demonstrations and a technology fair. The demonstrations were at various DOE sites showing the capabilities of an on-site rapid sampling and analysis system for satisfying the soil screening/characterization needs prior to the remediation operation. This program has shown that the acquisition, analysis, and reporting of soil sampling results can be safer, faster, and cheaper than is currently being practiced by the environmental remediation projects. While the MDLEST and sampling systems described in this report are operational, all systems have not been optimized, but the foundation for future sampling protocols and innovative sampling concepts have been successfully demonstrated. Termination of funding has brought this development program to a successful conclusion.

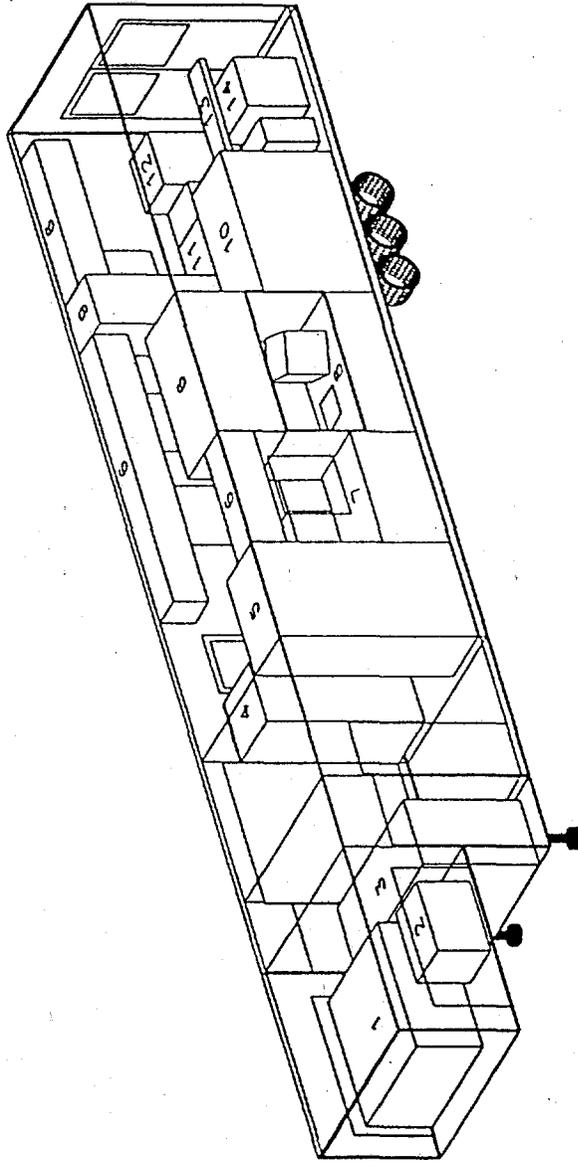
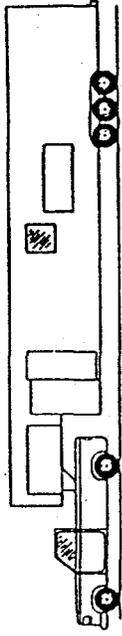
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Mobile Demonstration Laboratory
for
Environmental Screening Technologies



1. Air Handling System
2. Diesel Fuel Storage Tank
3. Diesel Generator
4. Cryogenic Gas Storage
5. Lavatory
6. Overhead Storage
7. Workspace
8. Sample Preparation Area
9. Electronics Equipment Rack
10. Spectrometer
11. System Control Center
12. Neslab Chiller
13. Laser Mount
14. Laser Power Supply

Ames Laboratory
Technology Integration Program (TIP)

Problem Description:

The need exists at various Department of Energy, Department of Defense, and private sector manufacturing facilities to determine the extent of surface and subsurface contamination from heavy metals, e.g. chromium, nickel, beryllium, cadmium, lead, etc. and radioactive elements, e.g. uranium, thorium, etc. used in manufacturing processes. Some of these metals were used directly in the production of weapons while others were used to facilitate the process. Accidental spills on soils or concrete floors and pads, leaking waste storage drums or tanks, effluent from smoke stacks, etc. may have been significant enough to not only contaminate the surface, but also penetrate into the subsurface, creating a plume. Soil screening/characterization is used to identify the location and contamination level, both surface and subsurface, and determine the type and amount of remediation needed, if any, for planning an efficient and affordable clean up operation.

During the field remediation material removal process, sample screening is needed to ensure that sufficient material is removed to satisfy the regulatory requirements driving the remediation process. Later, this removed material will undergo a treatment process to remove the contaminants. It will be necessary to screen/characterize the treated soil to determine whether or not it now satisfies the regulatory requirements, and can be spread back on the field. Post remediation monitoring ensures that the area remains in compliance.

Technology Description:

*Laser Ablation-Inductively Coupled Plasma-Atomic Emission Spectrometry/Mass Spectrometry, LA-ICP-AES/MS, techniques are used to screen/characterize soils and concrete (non-destructively) in situ to determine the extent of the contamination. Sampling probes have been developed and prototyped that will screen/characterize surface soils, concrete floors or pads, and subsurface soils. The sampling probes, both surface and subsurface, contain the laser, associated optics, and control circuitry to raster the laser (ablation) energy across one square inch of sample surface. Either sampling probe is connected by an umbilical, currently 20 m long, to the *Mobile Demonstration Laboratory for Screening Technologies, MDLEST*, a completely self-contained mobile laboratory containing the instrumentation to immediately analyze the samples generated by the laser ablation sampling system. The sample results are immediately available (~10 minutes) for field remediation decisions.*

Technology Need(s) Addressed:

The analysis instrumentation (ICP-AES/MS) in the MDLEST does not depend on radioactive decay (disintegrations per second) for detection but looks directly at the atomic make up of the samples of interest. A large number of metals including the longer half-life radioactive elements can be detected and quantified. The spectrometer is setup using either hardware, software, or both to simultaneously detect all elements of interest in each sample.

Surface soils are screened/characterized and areas having elevated contamination levels are

identified. The next step is to determine the extent of contamination penetration into the subsurface. Near surface samples, depths less than 5', are obtained using a manual core sampler and brought to the MDLEST for analysis using the laser ablation manual sampling mode. If these near surface samples indicate that the contamination has penetrated deeper, then the subsurface *in situ* sampling probe is employed. The subsurface probe prototype is designed to operate at depths between 5' and 90'. The combined use of surface and subsurface sampling will easily identify the extent of the contamination and the level of remediation needed.

Sample characterization, determining the contaminant and level present, will help select the remediation treatment process to be used. This important piece of information will provide information about the risk involved using an *in situ* remediation process, or whether the contaminated soil must be removed before treatment resulting in a more expensive process.

After collecting the results for a number of samples, a 3-D site map showing the areas and levels of contamination is generated. This information is immediately available to help remediation managers and contractors to make field decisions concerning the area and amount of material that needs to be remediated.

The MDLEST can be set up on-site to monitor soil treatment processes. This valuable function enables the remediation manager to monitor, in real-time, the treatment processes removing the contaminants and ensure that satisfactory agreement with both regulatory agency and QC/QA requirements is attained.

Technology Development:

Project Background:

FY1991 Activities

May 1991, The Ames Laboratory Technology Integration Program, TIP, group was formed with the primary mission: to actively interact with DOE sites to identify their needs, assess available technology solutions, and to demonstrate and deliver operable solutions. The main focus was mobile analytical instrumentation systems for safe, efficient, and cost effective site characterization, remediation process monitoring, and waste minimization.

- Staff was acquired to participate in the identification of technology and development needs
- ICP-AES analysis instrumentation using laser ablation (LA) sampling was identified as a FY1992 demonstration technology for the *uranium in soil integrated demonstration*, USID, at Fernald, OH. This technology will be used to screen surface soils for uranium and other heavy metals
- Initial development plan, design, and budget required to fabricate a self-contained mobile

laboratory prototype with all the utilities necessary to support the LA-ICP-AES instrumentation in the field

- Plan the development of a *robotic sampling accessory*, RSA, to complement the manual sampling capabilities of the *laser ablation*, LA, surface sampling probe

FY1992 Activities

The FY1992 activities involved scheduling and completing the tasks needed to produce a prototype laboratory, the *mobile demonstration laboratory for environmental screening technologies*, MDLEST, for demonstration of surface soil sampling using LA-ICP-AES technology by participating in the USID at Fernald, OH during September 1992. Additionally, a CRADA was signed with Layne Environmental Services Inc. as a partner in the development of a prototype subsurface *in situ* sampling probe using the laser ablation sampling technique.

- Schedule the projects and tasks that need to be accomplished in a timely manner for the prototype mobile laboratory to be demonstrated during the USID at Fernald, OH during September 1992
- LA-ICP-AES identified as a possible demonstration technology at Hanford and Oak Ridge during FY1993
- Acquire additional staff for the implementation of the technology development plan
- Execute the development plan; complete final design, write specifications and purchase the support utilities, instrumentation, trailer to house the laboratory, and the truck needed to move the laboratory
- Develop the LA surface sampling probe: design, fabricate, assemble, and test
- Design, fabricate, and integrate the equipment on the RSA with the MDLEST systems
- Design and fabricate the equipment and control systems needed for laboratory operation that are not commercially available
- Integrate all of the support utilities, instrumentation, and custom designed equipment and control systems into the mobile laboratory
- Develop the concept of an *in situ* subsurface sampling probe compatible with the mobile laboratory, and sign a CRADA with Layne Environmental Services Inc. as a development partner
- Issue a subcontract to Lockheed Missiles and Space Company for preliminary design of the laser raster system for the subsurface probe

- The first tool design by Layne for mating the casing to the subsurface probe was completed. The design was rejected after evaluation. Layne will rethink the tool design
- Obtain necessary environmental and safety training for staff that will operate the mobile laboratory in the field
- Receive delivery of the trailer that will house the laboratory, and install and integrate all utility systems, control and monitoring systems, and instrumentation
- Install site mapping software on MDLEST computer
- Perform extensive shop testing on the mobile laboratory and LA-ICP-AES instrumentation
- Complete NEPA documentation on the mobile laboratory and LA-ICP-AES instrumentation
- Locally field test the mobile laboratory systems and instrumentation using diesel generator power
- September 1992, two week participation of the mobile laboratory in the USID at Fernald, OH
- Poster presentations were made at two meetings
- A paper was presented at Spectrum '92 conference at Boise, ID

FY1993 Activities

The FY1993 activities involved continuing the development and optimization of the LA sampling system for the surface sampling probe, continue development of the subsurface probe working toward a field demonstration during FY1994, and locate and borrow, if possible, an ICP-MS. Modification of the MDLEST for the housing and operation of the ICP-MS for a field demonstration to detect ²³⁰Th at a *uranium mill tailing remedial action*, UMTRA, site during November 1994

- Issued final report of field activity during USID
- Optimize both hardware and software to stabilize the LA-ICP-AES operation
- Remove the RSA from duty and install components needed for LA-ICP-AES operation in the MDLEST
- Execute a loan agreement with the Perkin Elmer Corporation to borrow an ELAN 5000 ICP-MS

- Write specifications for a compact laser and issue a purchase requisition. This was eventually canceled due to vendor non-performance
- Complete the final design, fabricate the components, and shop test the subsurface sampling probe
- Write specifications for a *global positioning system*, GPS, and issue a purchase requisition
- Integrate the GPS into the surface sampling probe and MDLEST systems
- Modify the MDLEST and systems for installation and operation of the ICP-MS
- Consult with Allied Signal, Kansas City, MO, to learn the technique of launching high energy laser light into a fiber optic without destroying it
- Optimize the operation of the surface sampling probe rastering system
- Evaluate preliminary optics design for the subsurface probe submitted by Lockheed and request alternate design
- Final design, fabrication and successful shop testing of the subsurface probe ablation cell and subsystems
- *In situ* fit and gas seal testing of subsurface probe with Layne tool in Phoenix
- Presentation of MDLEST capabilities; at EMSL, Las Vegas, NV, for ER Engineering Group at Westinghouse Hanford Co, at the Region VII EPA state directors meeting Des Moines, IA, at DOE vitrification project West Valley, NY, at Thermo Jarrell Ash Instrument Co. Franklin, MA, to the Electro-Optics group at Allied Signal(DOE), Kansas City, MO
- Poster presentation at Houston, TX, TIE

FY1994 Activities

The FY1994 activities involved participation in a demonstration at the Gunnison, CO, UMTRA site; participation in the surface sampling USID demonstration at Fernald, OH, during June; a successful subsurface probe field demonstration with Layne Environmental Services Inc. during August in the Denver, CO, area; a surface sampling demonstration at the Saint Louis Airport, SLAPS, site during the first part of September; and finally, participation in the USID Technology Fair in Cincinnati, OH, during the last week of September 1994.

- Install, integrate, and test the ICP-MS in the MDLEST
- Demonstrate the MDLEST capabilities to detect ^{230}Th using the ICP-MS at the UMTRA site

in Gunnison, CO

- Design, fabricate, and test a lazy-susan to expedite analysis of containerized samples
- Ablation cell gas flow and particle entrainment optimization using ICP-MS
- Direct *in situ* ablation of concrete is successful
- Design, fabricate, install, and test new optic design in the surface sampling probe to accommodate the compact laser direct beam delivery
- Integrate the control of the compact laser into the MDLEST computer control system
- Formal report of the UMTRA demonstration
- Participate in the USID at Fernald, OH, June 12-23, 1994. Compact laser installed in the surface sampling probe
- Participate in the SLAPS demonstration Sept 6-13, 1994
- Exhibit the MDLEST in the Technology Fair hosted by FERMCO, Cincinnati, OH, Sept 27, 1994
- Joint participation with CRADA partner Layne Environmental Services Inc. in demonstrating the feasibility and capabilities of the subsurface *in situ* sampling probe at Commerce City, CO, August 14-19, 1994
- Briefing and tour of the MDLEST to five visiting Russian scientists
- Two patents from the development work of the MDLEST and LA have been filed and are pending

This completes the list of activities during the lifetime of the MDLEST and LA-ICP-AES/MS prototype development and demonstration project. The following will now discuss in more detail the total system that resulted in an operational prototype for field operation and evaluation.

The MDLEST:

The *Mobile Demonstration Laboratory for Environmental Screening Technologies*, MDLEST is a completely self-contained mobile laboratory. The generic mobile laboratory concept incorporated into the MDLEST design was to provide a platform with utilities, control, and monitoring systems to support whatever screening/characterization instrumentation might be chosen for demonstration. With this in mind, the following design considerations and utilities were

incorporated in the MDLEST.

The trailer

The MDLEST is a 44 ft. long by 8 ft. wide and 8 ft. high 5th wheel trailer towed with a medium duty truck. The interior of the MDLEST has aircraft type tie down tracking installed in the ceiling, walls, and floor. This allows quick removal and replacement of the desks, cabinetry, and instrumentation. An additional feature is the computer room type floor system. This floor system allows all utilities and control wiring to be run under the floor and can be rapidly changed for different instrumentation installations. The trailer running gear has three axles with six wide tread tires to allow the trailer to be pulled into undeveloped areas for laboratory operation. The following section lists the systems that are installed in the MDLEST for operation and instrumentation support.

Utilities:

Air conditioning

A four ton heating/cooling, HVAC, system is used to condition the air in the laboratory space for proper instrumentation operation. This system provides the MDLEST with a positive internal pressure which assured a net air flow from the trailer, preventing infiltration of dust and contamination from the outside. The HVAC outside make-up air is filtered through a high efficiency particulate air, HEPA, filter to remove any air borne environmental contamination. This unit is equipped with hot-gas-by-pass to prevent a surge with resulting spike in the electric circuits when the air conditioning unit turned on. This spike is capable of shutting down computers and other sensitive instrumentation.

Chilled water

Neslab Instruments Inc., Model HX-150A closed loop chilled water recirculator, is used to provide chilled water to the instruments for cooling.

Communication

The MDLEST is equipped with a radio communication system. Each member of the operating crew working outside the MDLEST carries a portable radio for coordination and monitoring of the LA sampling and calibration activities. In addition the radio system maintains a link with other site activities for safety purposes.

The MDLEST also is equipped with a cellular telephone to maintain contact with the home laboratory, instrument, and equipment manufacturers for operating or troubleshooting procedures and ordering replacement parts.

Electricity

Electricity for the operation of the mobile lab and the instrumentation is provided by a 50 KW, 3 phase, 208 VAC diesel driven Onan generator. Alternately, a local power source, 3 phase, 208 VAC, and 100 Amps per leg, can be used to power the MDLEST if available.

Global positioning system

An Ashtech model M-XII *Global Positioning System*, GPS, has been integrated into the MDLEST and the surface sampling probe to supply real-time coordinate information for mapping off-grid *in situ* sampling. The antenna for this instrument is mounted on the surface sampling probe and is connected to the receiver in the MDLEST using a cable in the umbilical. This system is capable of identifying the coordinates of a sampling site that are accurate to a few centimeters.

Instrumentation gases

An external gas cabinet provides cylinder storage and regulated gas distribution manifolds with automatic valves and pressure monitoring. The LA-ICP-AES/MS instrumentation uses large amounts of argon gas that is obtained from the boil off of a dewar containing liquid argon. This dewar provides a pure gas source (~19 cylinders) that lasts for more than two weeks under normal operation.

Safety and health monitoring

A Geiger counter (Ludlum model 3 Survey meter with a model 44-9 detector) is used for personnel and sample radiation monitoring and surveying purposes. This instrument is capable of detecting alpha, beta and gamma radiation.

A Gastech, model GX-820CO, gas monitor, which detects combustible gasses and the levels of oxygen and carbon-monoxide is used to monitor the air quality in the MDLEST.

Umbilical

Electrical power and instrumentation control wiring, fiber optic cable, argon gas supply tube, and ablated particle return tube are contained in the 20 or 40 meter flexible umbilical connecting the surface sampling probe to the MDLEST. The umbilical connecting the subsurface *in situ* sampling probe to the MDLEST uses the same connections. With the installation of the compact laser in the surface sampling probe, an auxiliary umbilical was constructed to carry the control signals, chilled water, and electrical power required for the laser operation. The umbilicals are enclosed in a flexible hard plastic outer shell that has been pulled through a thin plastic sleeve. This plastic sleeve can be easily stripped off and disposed of if it becomes contaminated.

Wet chemistry preparation area

A minimal wet chemistry sample preparation area was available in the external side bay for the Fernald, OH, *Uranium in Soils Integrated Demonstration*, USID, during September 1992. This area was equipped with a sink, splash shield, acid storage, hood vent fan, and a microwave sample preparation system, CEM model MDS-2100, to assist in the preparation of samples using wet chemistry techniques. Field samples were acquired, prepared using wet chemistry techniques and microwave digestion, and then nebulized in to the ICP-AES to obtain results for comparison with the LA-ICP-AES technique.

Computers for Monitoring and Control:

The current prototype of the MDLEST contains two PC computers. One is used to monitor and control the operation of the utility subsystems, while the other one collects and analyzes the data from the instrumentation, and reports the results. During the initial fast-track development stage, to get the MDLEST in the field by September 1992, four computers were required to handle monitoring and control all of the tasks that were automated. Over time the number of tasks that need to be handled at one time have been combined and reduced.

The computer system monitors and controls devices, log mechanical and electrical (not analytical) audit-trail type data, generate reports, plot analytical data, display information to the operator(s) in real time, and automatically sequence the various operations that are required during a sample run. The raw data and instrumentation parameters is written onto an optical disk (WORM) to preserve an electronic chain of custody. A part of the computer control system is a new high level computer language, Prolix, developed specifically for the MDLEST project. Prolix specialized in control of the complex inner workings of the analytical laboratory, handling common error conditions automatically, and performing all other rote functions of the laboratory control system.

The computers, A/D convertors and process control devices were housed in a single rack. The computers were vibration dampened using shock mounts. The electricity to the computers was conditioned and backed-up with uninterruptable power supplies.

Sampling and Analysis System:

Analysis Instrumentation:

Two production model analytical instruments, an *atomic emission spectrometer*, AES, and a *mass spectrometer*, MS, have been installed in the MDLEST and used at the various field demonstration sites, either singularly or simultaneously, to analyze samples. The instruments are mounted on vibration dampers to isolate the instrument from vibrations caused by the MDLEST's operating utilities, e.g. vibration from the diesel generator. The spectrometers are used to resolve the excitation from the ICP. Each instrument analyzes a different product from the ICP excitation, the

AES resolves the high intensity light, where as the MS injects and mass resolves the ion stream, to determine the elemental makeup and quantity of each in the sample. Using the LA sampling technique, the AES detection limit is ppm, while the MS has a detection limit of sub-ppb. The following sections discuss the way that the spectrometers were used during field operations, and the modifications that were made to optimize and stabilize each instrument..

Ion Coupled Plasma torch

The ion coupled plasma, ICP, torch, which produces the sample particle excitation, is common to both instruments and mounted in front of the AES and MS spectrometers. Each spectrometer analyzes a different product from the ICP excitation. In general, the ICP uses a radio frequency, RF, generator to excite an argon gas flow and produces a stable plasma at ~8000 K. For the LA process, sample particles are injected directly into the plasma for dissociation using an argon gas flow of 0.9 lpm, which was optimum for emission from the ICP. The gas flow optimization was accomplished by watching the results as different variables were adjusted.

The ICP process gasses are expelled through negative pressure hoods and HEPA filters before being released to the environment. The filters are designed to trap potentially hazardous sample residue, e.g. radioactive contaminants, that may pass through the ICP unprocessed and prevent its release to the environment.

Atomic Emission Spectrometry

The atomic emission spectrometer, a standard production model Thermo Jarrell Ash, ICAP 61E Plasma Emission Spectrometer, was installed in the MDLEST for the analysis of samples introduced either by LA or solution nebulization. This instrument is equipped with 20 channels for the simultaneous analysis of a number of elements including uranium and thorium. The sample, is introduced into the ICP torch where it is vaporized, atomized, and ionized by the high temperature plasma. Relaxation of the electronic excitation produces intense multi-wavelength light that is introduced into the optical cavity of the spectrometer where it is resolved into individual wavelengths and intensities. The wavelength identifies the elements present in the sample (qualitative), while the wavelength intensity is proportional to the concentration (quantitative). The ICP-AES is capable of detecting and quantitating elements in real-time at the parts-per-million level (ppm= $\mu\text{g/g}$) for direct sample injection (LA sampling) and ~10 parts-per-billion level (ppb= ng/g) for samples prepared for solution nebulization.

The elemental spectral lines chosen for analysis were selected to avoid (minimize) interferences and overlap. These interferences are the result of the inhomogeneity of the soil composition which contains the element(s) of interest (contaminants), and other elements. This also is an important consideration when selecting the calibration standards.

This instrument was prepared for mounting in the MDLEST by moving a number of electrical panels from the back of the instrument, that is against the wall when installed in the MDLEST, to the front for access. The heat from the RF matching network was vented to the outside to help

stabilize and ease control of the operating temperature in the MDLEST. Finally, the instrument is installed in a vibration damping mount in the MDLEST.

Prior to highway travel the focal curve in the AES is tied down with shipping bolts in accordance with manufacturers suggestions. Also, the instrument is securely tied down, defeating the vibration damping, to prevent road damage during transport. Once on site, the MDLEST is set-up, all travel restraints are removed from the AES, the channels (optics) are aligned, and the system is calibrated. The spectrometer optics are not aligned until the internal temperature of the AES has fully stabilized. These steps for travel preparation and site set-up have been incorporated into the AES operational procedure.

The sensitivity of the AES to temperature fluctuations in the laboratory was not appreciated until the results from the Fernald '92 demonstration were evaluated. Large variations in the results for the standard samples, between different days and during the same day, were attributed to temperature fluctuations in the MDLEST's ambient temperature. This problem was solved by mounting a temperature controlled heater in the optical cavity of the instrument and setting the controller to $\sim 32^{\circ}\text{C}$, approximately 17°F above the laboratory ambient operating temperature. The addition of this auxiliary heater has stabilized the operation of the AES. From a cold start, it takes approximately 3 hours for the temperature of the AES to stabilize. To minimize warmup time during the start up process, the AES controller/heater is tied into a separate 110 VAC circuit that can be plugged into site power when available. Keeping the instrument warm allows data to be acquired in approximately 30 minutes from a cold start.

The AES is a simultaneous spectrometer, however, instrument background corrections are acquired sequentially after the elemental data are taken. This time delay and sequential technique for acquiring background corrections produces an error in the determination of the elemental concentrations in the sample. While this is relatively unimportant when using the solution nebulization sample introduction, it was found that this correction protocol is capable of causing large errors when using the LA sampling technique. The sampling protocol was designed to minimize this problem.

Mass Spectrometry

A mass spectrometer, ELAN 5000 ICP-MS, was obtained on-loan from the Perkin-Elmer Corporation. This spectrometer, a standard production model, was installed in the MDLEST for the UMTRA demonstration at Gunnison, CO, during November 1993 and is capable of scanning from 3 to 250 atomic mass units (amu). The mass scan may be over the whole range of amu available or only a portion of it. Also, "peak hopping" is available where only the masses of interest are sampled while the others are skipped. The instrument detection limits are ~ 0.5 ppb (ng/g) for the direct sample injection (LA sampling), and ~ 0.03 parts-per-trillion (ppt=pg/g) for solution nebulization.

Within the instrument the ions are separated and counted based on their mass/charge ratio. The detection of a mass determines the presence (qualitative) of an element in the sample and the count

intensity is proportional to the concentration (quantitative). This instrument was chosen for its ability to detect and quantify elemental isotopes, whereas the AES is only capable of total elemental analysis. In particular, the MS can detect the ^{230}Th levels in the soil below the 15 pCi/g (0.8 ppb) which was needed to satisfy the Nuclear Regulatory Commission, NRC, remediation agreement for the Gunnison CO. site.

For the UMTRA demonstration, the MS was used in the "peak hopping" mode analyzing selected masses. During field operations the MS is shut down each evening and required approximately one hour to warmup and stabilize the next morning.

The ICP-MS was mounted, unmodified, in a vibration damping mount in the front bay of the MDLEST and integrated into the laboratory utility system. Prior to highway travel shipping blocks and constraints were installed to prevent damage to the instrument. Once on-site, with all shipping restraints removed, the MS was started, warmed up, aligned, and calibrated, and the instrument would be ready to acquire data within ~4 hours.

During data acquisition two MS problems appeared that affected the final results. First, variations in the quantity of sample reaching the ICP, caused by the rastering of the laser radiation producing a time varying quantity of material from the ablation cell, resulted in a variation in the intensity of the ion stream being analyzed, creating large, nonreproducible errors in quantitation. This error resulted from the spectrometer analyzing each mass unit individually and sequentially. Second, the ions pass from the ICP into the mass spectrometer through skimmer cones. These cones became coated with large particles of sample residue and, over time, the insulating dielectric coating build up would throttle the ions entering the MS for analysis. Both of these problems resulted in a decreased sensitive and the latter caused selectivity dependent on mass. It was found by using the 532 nm laser energy for sample ablation reduced the sample particle size minimizing the cone build up and plugging.

Laser Ablation Sample Introduction System:

The laser ablation, LA, technique is used for the direct sampling of soil and is capable of sampling other solids including concrete. Laser radiation, either from a compact laser mounted in the sampling probe or delivered to the probe via a fiber optic cable is focused and rastered over the sample. The focused radiation on the surface of the sample produces micron (μm) size sample particles that are entrained in an argon gas flow and swept out of the ablation cell, transported through a PVC tube, and directly injected into the ICP. The required laser energy intensity at the sample surface for directly sampling soil is greater than 10^8 W cm^{-2} to prevent preferential sampling. This technique eliminates the lengthy, approximately eight-hour, sample dissolution and concentration processes needed to prepare a sample for injection into the ICP using nebulization techniques.

Lasers

Two different lasers have been used to provide the energy needed to ablate the soil samples. A

Continuum laser (model NY81-30), mounted in the MDLEST, with its energy transported to the sampling site via a fiber optic cable, and a Big Sky Compact Folded Resonator, model CFR 225A, mounted in the surface sampling probe for direct beam delivery to the sampling site. Both lasers are Nd:YAG, Q-switched with a 30 Hz repetition rate, and a 1064 nm fundamental wavelength that is frequency doubled to obtain the 532 nm wavelength used for this work.

The stability of the laser energy is important in obtaining a representative sample from the area being sampled. At low or unstable laser energies sample ablation becomes selective causing interferences that affect the quantitation. Several factors were found that contribute to the instability problem: insufficient laser and power supply warmup, flash lamp aging, and algae growth in the laser flash chamber. Both lasers experienced some or all of these problems, however, the compact laser was more prone to the algae growth problem since it is mounted in the surface sampling probe where its operating environment is the outside ambient temperature and the laser head cooling water is transported and returned through a 100 ft. umbilical. This problem was solved by disassembling and cleaning the flash chamber components and sterilizing and treating the water used to refill the system. By closely monitoring the laser operation and settings, performing routine maintenance, and regularly analyzing calibration standards, these laser stability problems will be nonexistent or have minimal effect on the results.

The compact laser is an off-the-shelf model, and was not specifically designed or manufactured for field operation under ambient conditions as we have used it. During the USID at Fernald, OH, June 1994, the laser energy was observed to be degrading and becoming unstable. The flash lamp was changed to restore the laser energy, but the laser would not lase after this routine operation. The manufacture was contacted and they requested that the laser be returned to the factory for repair. The problem was found to be the adhesive used to secure the internal optical components had degraded resulting in the mirrors detaching from their mountings making it impossible to align the beam. The repaired laser was used in the field during the SLAPS demonstration, and performed within specifications without any of the optical problems encountered during the Fernald demonstration being repeated.

Fiber optic delivery

During the initial development of the laser ablation sampling technique a small Nd:YAG laser capable of supplying the needed laser energy for sample ablation was not commercially available, therefore, the large Continuum laser described above was purchased and mounted in the MDLEST. To deliver the laser energy to the sampling site from the Continuum laser required the laser energy be launched into a fiber optic cable for delivery to the sampling site. The fiber-optic cable, a 25 m long, 600 μm diameter core silica clad silica, (Polymicro Technologies model FVP-600660690) was chosen to carry the 532 nm laser energy through the umbilical to the surface sampling probe.

The disadvantages of the fiber optic delivery system are: 532 nm light with laser energy greater than 5 mJ ($\sim 2.6 \times 10^8 \text{ Wcm}^{-2}$) damages the launch end of the fiber optic cable requiring repolishing and realignment; a fiber optic cable longer than 30 m attenuates the beam energy sufficiently that the

energy density at the sampling site is less than that required for non-preferential sampling; the laser light emitted from the end of a fiber optic must be refocused onto the ablation site within ± 1.5 mm to obtain sufficient energy for non-preferential ablation; and the sample surface must be uniformly flat i.e. varying not more than 1.5 mm.

Even with these apparent disadvantages, the fiber optic cable is a compact way to deliver a large amount of pulsed laser energy over distance. The shortcomings pointed out will be resolved by innovative optical design, new fiber optic cable technology, use of fiber optic bundles and possibly larger core diameter fiber optic cables. The current development time frame and budget did not allow these possible solutions to be explored.

Direct beam delivery (compact laser)

A compact laser, mounted in the sampling probe, delivers ablation energy directly to the sample site using a minimal number of optical components. This laser, head size and weight ($3\frac{1}{2}$ " x $3\frac{1}{2}$ " x 14" long, 15 lbs.), has only been commercially available since mid-1993, and provides a stable energy source (~ 5 mJ) for the direct beam sample ablation system.

The advantages of the direct beam over the fiber optic delivery system are: the laser beam is focused to a smaller spot size on the sample producing higher ablation energy densities ($\sim 10^9$ Wcm⁻²), which insures non-preferential sampling; the focus on the sample is less critical (within 15 mm), thus allowing non-preferential ablation of rough sample surfaces; the laser head can be operated up to 30 m from the power supply with no degradation in beam energy, with the 30 m distance being limited only by the weight and length of the auxiliary umbilical needed to deliver utilities (flash lamp power, trigger signals, and cooling water) to the laser head.

Sampling Probes:

Two probes were developed for *in situ* sampling, one for surface and the other for a subsurface sampling. Both probes function, operationally, the same in acquiring samples. They contain: the electronics and hardware to focus and raster the laser beam across the sample site; to monitor the system performance; and an ablation cell to collect and entrain the sample in an argon gas flow for transport to the MDLEST for analysis. Either probe, interchangeably, is connected to MDLEST via an umbilical that carries the utilities, particle transport tube, and control and monitoring signals needed for operation. Initially, a fiber optic cable was used to deliver the sample ablation energy to the ablation cell in each probe. The current surface sample probe configuration incorporates a compact laser to supply the sample ablation energy. The following gives a description and discussion of each probe type, and the subsystem components.

Surface sampling probe

The first surface sampling probe design incorporates an ablation cell (2" ID x $2\frac{3}{4}$ " tall), located on the bottom of the probe housing which is attached to a flat plate (~ 10 " dia) fitted with a knife edge (~ 3 " dia x ~ 2 " long) that sits directly on the soil at the sampling site. The knife edge protrudes into

the soil to help make a mechanical gas tight seal with the soil. The surface sampling probe's free standing design allows the weight of the probe to help maintain the seal with the soil during the sampling operation. This gas seal is critical in maintaining a constant flow of argon through the cell to entrain the ablated particles and transport them back to the MDLEST for analysis. Alternatively, this cell system is set in a specially designed fixture that holds a 1 oz. polypropylene bottle containing ~10 g of soil sample. The soil samples are either standard samples used to calibrate the ablation sampling and analysis system, or field samples that have been collected and brought to the MDLEST for analysis.

The vertical ablation cell design used in the surface sampling probe is used for sampling horizontal surfaces. This vertical cell design means that the laser energy is directed through a window in the top of the cell normal to the surface being ablated. The argon gas flow enters the cell at the bottom near the sample surface, entrains the sample particles generated by the laser ablation process, and exits through two ports, 180 degrees apart, near the top of the cell, carrying the entrained particles into the transport tube. The important cell design parameters, affecting the intensity and stability of the spectrometer signal were found to be: the cell volume, the argon gas entry level, velocity, and flow pattern within the cell, and the laser beam raster pattern.

The laser energy for the first probe design was delivered to the probe via a fiber optic cable. This cable was terminated on an optical breadboard that is mounted on a z-axis optical table (all optical tables were purchased off-the-shelf from New England Affiliated Technologies, NEAT). The beam focusing lens, beam splitters, and photodiodes, used to monitor and focus the laser beam on the soil surface, also were mounted on this optical breadboard. The z-axis optical table was mounted vertically and perpendicular to the x- and y-axis optical tables. The x- and y-tables provided the motion to raster the laser energy across the sampling site while the z-axis table motion focuses the laser energy on the sampling site. To focus the laser beam on the sampling site, the beam energy is reduced below the ablation energy, the x- and y-axis tables are driven to the "home" position (the center of the sampling pattern), and the z-axis is adjusted to maximize the image of the 600 μm dia. optic fiber on the soil surface. This focus position gives the maximum achievable ablation energy on the sample site. The z-axis table is locked and the x- and y-axis tables are driven to the raster pattern starting position. During ablation sampling the laser energy is rastered, in a spiral pattern starting at the outside, over a 6.5 cm^2 area.

The ablation cell optimization was limited to modifying various components affecting the argon gas flow in the cell: varying the gas entry height above the sampling surface; increasing or decreasing the entering gas velocity; and adjusting the amount of turbulence in the cell. A standard soil sample was ablated after each change and the resulting MS signal recorded. The maximum MS signal, (~150% increase above that of the original design, and interpreted as the maximum sample entrained), was obtained with a high velocity gas flow entering the cell at the sample surface creating maximum turbulence. These enhancements were permanently incorporated into the ablation cell. The optimized cell was used for the USID, Fernald, OH, June 1994 and all subsequent demonstrations using the surface sampling probe.

Changing the cell volume and raster pattern parameters were not explored, however, these are

clearly important factors affecting the reproducibility of the analyzed results. The surface sampling ablation cell has two sample particle exit ports (180° apart) near the top of the cell. These are joined together outside of the cell using a "Y" to obtain a single particle stream for transport through the 20 m umbilical and injection into the ICP. Since the raster pattern is a spiral, the ablation site moves to and then away from the output ports. This allows the particles to be delivered to one port or the other preferentially causing oscillations in the intensity of the signal from the spectrometer. This oscillation problem was minimized by setting the spectrometer data collection period long enough to integrate over these oscillations. The curve generated from the spectrometer signal intensity versus time for the ablation cell easily fits the model of the constantly stirred tank reactor, CSTR. The CSTR models gas flow in, gas flow out, cell volume, and internal particle generation. Although an attempt was not made to optimize the ablation cell volume, the CSTR model suggests that maximum particle entrainment in the gas flow will not occur until all variables have been optimized as an operating system.

Early in 1994 a compact laser was available to test the direct beam delivery concept. The surface sampling probe was modified by adding a module to the side to house the compact laser head. The power supply for the compact laser was housed in the MDLEST, and the utilities and control signals between the laser head and power supply were routed through an auxiliary umbilical. A shutter and mirrors were installed to control and direct the laser beam onto the x-, y-, and z-axis focus and rastering system in the surface probe. This modified surface sampling probe with its *in situ* ablation energy source was used for the USID Fernald, OH, June 1994 and all subsequent demonstrations using the surface sampling probe.

During ablation sampling "dust" particles, nearly invisible to the unaided eye, are picked up from the gas flow and coat the window. This dust coating causes two problems, the incoming laser light is scattered reducing the ablation energy density at the sampling surface, and is believed to be the source of the cross contamination between sampling runs. The first problem can lead to preferential ablation if the energy is reduced enough. The cross contamination problem arises from the dust that has settled on the window, it would be ejected into the sample stream for analysis during the next time that the laser radiation is rastered over it, i.e. during the ablation of the next sample. Treating the window with an anti-static agent minimized the collection of large particles. Cleaning the window between ablation sampling runs appeared to eliminate this problem. The level of cross contamination between various ablation sampling runs was ~1% or less.

Particle transport tube

Polyvinyl tubing, 3/16" I.D. x 30 m long, is used to transport the sample particles from the ablation cell to the ICP torch. This tubing was selected because of its use in the pharmaceutical industry to transfer powders. The loss of sample particles from the gas flow during transport through the tubing is the result of settling due to gravity or by attraction from static charge buildup on the inside wall of the tube. Several different tubing lengths were used to determine the dependence of the spectrometer signal intensity on the loss of sample particles. This loss of sample was not sufficient to affect the quantitation of the sample for the range of concentrations encountered. An additional concern is the possible sample cross contamination caused by transient particle dropout reentering

the gas stream at a later time (during the ablation of a different sample). However, since the transport gas in the tube is under laminar flow conditions, (Reynold's number, $Re=359$), once a particle drops out of the gas flow it will not reenter unless an external disturbance is encountered. As a test for cross contamination, a blank soil samples, (no contamination), were ablated after contaminated soil samples. The analyzed results show that no cross contamination was detected. The sampling protocol requires data on blank soil samples to be acquired regularly during field sampling to insure that cross contamination is not occurring.

Subsurface sampling probe

The subsurface *in situ* sampling probe was developed and field tested under a *Cooperative Research and Development Agreement*, CRADA, partnership between the Technology Integration Program (TIP) of the Ames Laboratory and Layne Environmental Services Inc. (LESI) of Phoenix, Arizona. Under the CRADA, that was signed during August 1992, LESI was responsible for designing and fabricating the windowed drive tool that attaches to their casing and interfaces with the subsurface soil to be sampled, and to provide the percussion hammer rig, casing, and operating crew needed to drive the tool into the ground. The TIP team was responsible for designing and fabricating the probe that slides into the casing mating to the LESI tool and acquires the soil sample *in situ*, and to provide the MDLEST to operate the subsurface probe and analyzes the samples.

The subsurface probe was designed to fit into a 6" ID x 9" OD standard drive casing. The subsystems in the probe include the rastering and focusing systems, ablation cell, control and communication electronics, and the outer protective shell of the probe. This prototype subsurface probe and tool design incorporated off-the-shelf components, where possible, to minimize the cost and shorten the development time. This probe was successfully field tested at Commerce City, CO, during August 1994 (additional information under demonstrations below).

All of the design parameters discussed above for the surface sampling probe are also important in the subsurface probe. In addition, consideration must be given to factors involving the compact laser if direct beam sampling is used. The water cooling system in the laser must be capable of handling the added water pressure head and the water circulating pump must be capable of pumping against the added resistance. These compact laser considerations were expressed in specifications when the request for quotations were sent to laser manufacturers. The company that received the bid to produce a compact laser meeting the specifications was unable to deliver a working laser. After being late by ~3 months the order was canceled and the design of the subsurface probe was completed. Space in the probe was allocated for the eventual purchase and installation of a compact laser that would meet required specifications. The probe design was modified to use the fiber optic delivery system to supply the ablation sampling energy. Thus, making the decision to use the fiber optic energy delivery system allowed the probe design and fabrication to be completed so that the subsurface concept could be tested during FY94.

The horizontal ablation cell, 2" diameter x 1" long, is designed to sample a self-supporting vertical soil surface exposed through a 1" x 1" window in the side wall of the subsurface tool, entrain the ablated particles in an argon gas flow and transport them to the surface for analysis. A mechanical

gas tight seal between the wall of the subsurface tool and the surrounding soil is formed by compaction during the percussion driving of the tool. The space between the ablation cell and the inside wall of the tool is sealed by inflating a donut shaped bladder. The argon transport gas enters the bottom of the cell through a narrow slit and is directed upward across the face of the sampling surface, entraining the ablated particles and sweeping them out through the exit port at the top. The gas entry slit increases the velocity and turbulence of the gas as it enters the ablation cell which increases the sample entraining capability. The laser energy enters the cell through a window in the end opposite the sampling surface, and rasters horizontal lines starting at the top of the window and moving down. The stability and intensity of the sample particle flow from this cell, as seen from the stability and intensity of the signal from the spectrometer, was found to be superior to that of the vertical cell used in the surface sampling probe. This is believed to result from higher particle entrainment efficiency in the horizontal cell.

The raster and focus system is located on two separate optical tables mounted linearly. The initial optical system design was completed under a subcontract with Lockheed Missiles and Space Company, Austin, TX, division. The final optical design chosen for use in the subsurface probe was a modification and refinement of the original Lockheed design. The fiber optic cable termination and focus lens are mounted on the upper table along with the beam splitter and beam monitoring and focus diodes. The rotary mirror that oscillates the beam back and forth in the horizontal direction during the raster is mounted on the lower stage. During focusing, the lower stage is fixed and the upper stage is driven into focus by observing the intensity of the reflected light through a pinhole on the focus photo diode. Once focus is attained, the stages are locked and they are moved in the same direction together to raster the focused beam horizontally across the sample surface and stepwise down in the vertical direction after each horizontal sweep.

While this subsurface probe was successfully tested only once in the field, and functioned as designed, a study is needed to determine the problems that will be encountered when quantitating subsurface samples since the soil matrix is expected to be widely variable. For example, if silicon is to continue to be used as an internal standard, then a determination of the variability in the silicon content with depth will be needed to quantitate the other elements in the sample with minimal error.

Data Analysis Protocol:

The laser energy, 532 nm wavelength and 30 Hz pulse rate, is rastered, (surface probe in a spiral pattern, subsurface probe in a horizontal linear back and forth and a vertical stepwise down pattern), over the sampling area, 6.5 cm², at a constant linear velocity of 7.5 mm/sec with each successive pass offset by 0.21 mm.

Using the fiber optic cable to deliver the laser energy, the diameter of the fiber was imaged on the sample resulting in the ablation of material from a site ~0.600 mm diameter x ~0.1 mm deep. The velocity and offset of the raster pattern gives a sample ablation site an overlap of ~42%. Originally the sample protocol raster pattern did not allow ablation site overlap, and while an in-depth study of the non-overlapping vs overlapping was not done, there were no disadvantages observed when allowing overlap of the sample ablation sites. This overlap allowed for more analysis time, i.e.

more sample particle generation for analysis. From start to finish, approximately 9 minutes is required for the raster pattern to sample the 6.5 cm² area and, although the sample starts reaching the ICP ~20 seconds after the ablation sampling run starts, because of transport delay, data acquisition for analysis is delayed for 120 seconds to allow the sample flow from the ablation cell to stabilize.

With the modified surface sampling probe using the compact laser to directly deliver the ablation energy to the sample site, the energy is focused resulting in ablation from a site ~0.10 mm diameter x 0.1 mm deep, with no ablation pattern overlap during rastering.

Data for each sample site is taken during a single complete raster pattern. The spectrometer is set up to divide this raster pattern into five replicate samples allowing calculations of the average counts and %RSD for the total sample. The resulting large %RSDs seen for the LA-ICP process are not surprising since each replicate sample is taken from a different location within the raster pattern showing that these samples are very inhomogeneous.

The laser ablation sampling system and ICP-spectrometer were calibrated using one or several of the following: synthetic standards (site made standards), *Standard Reference Materials* (SRM), or *Nuclear Reference Materials* (NRM); standard soils of known composition containing the elements of interest. After the instrumentation is setup and aligned, and the initial calibration is completed then the system is ready to analyze field samples. An instrumentation quality check is repeated after every tenth sample. If the quality check fails then the system is recalibrated and the field samples repeated.

The analysis results for each replicate sample are calculated and stored by the spectrometer computer, and a hard copy is printed. These results together with the instrument settings and system parameters are transferred to a file on a *write once read many*, WORM, optical disk, creating an *electronic chain of custody* for the sampling and analysis acquisition system.

The factory installed data analysis and handling software for the spectrometers was not adequate for the type of field production analysis that the MDLEST was designed to handle. These software packages must be designed with the operation and efficiencies required for rapid sampling and analysis in mind. While this project did not address the design of software needs directly, field operations identified data analysis, file handling and retrieval problems. The solution to these problems will enhance and speed the data analysis and reporting process.

Field Demonstrations:

During the life of the AMLP, the MDLEST participated in five field demonstrations and one Technology Fair. A different objective and problem, was the focus for each demonstration. The five field demonstrations and objectives were: 1992, Fernald, OH site detection and quantitation of U in soil, the initial field trial of the new MDLEST, and the first field use of the laser ablation sampling technique for *in situ* surface sampling; November 1993, Gunnison, CO, Uranium Mill Tailing Remediation Action project, UMTRA, to detect ²³⁰Th levels in the soil after the mill tailing

pile had been removed; June 1994, return to Fernald, OH, to participate with other state-of-the-art field sampling technologies to test a direct probability sampling plan; August 1994, Denver, CO, area, to test the subsurface *in situ* sampling probe; September 1994, St. Louis, MO, to participate in the Expedited Site Characterization (ESC) methodology at the Saint Louis Airport Site (SLAPS), to evaluate the field capabilities of a broad range of characterization/monitoring technologies; and finally, September 27, 1994, Cincinnati, OH, to participate in the Uranium in Soils Technology Exposition, hosted by FERMCO and DOE, EM-50, displaying both field screening/characterization technologies and soil remediation treatment processes. In addition, a local feasibility test using laser ablation *in situ* surface sampling to non-destructively sample concrete to determine the contaminant level.

Fernald '92

The detection and quantitation of U levels in the soil surrounding the sewage treatment/incinerator facility was the focus for the September 1992 Uranium in Soils Integrated Demonstration, USID, hosted by *Fernald Environmental Restoration Management Corporation*, FERMCO, Fernald, OH. This was the first operational field test of the totally integrated MDLEST and LA-ICP-AES surface sampling system. The AES, a multi-channel analytical instrument capable of detecting a number of elements, (20), simultaneously, and quantitating at the parts per million (ppm) level, was installed in the MDLEST and used during this demonstration. While U was the element of primary interest during this demonstration, Th was also quantitated, and other elements including Al, Ba, Ca, Cr, Cu, Fe, K, Mg, Mn, Na, Pb, Si, Sr, Ti, V, and Zr were detected. Synthetic soil standards, made by putting U and Th solution standards into non-contaminated Fernald site soil, were used to calibrate the LA-ICP-AES instrumentation. The time required for sampling, analysis, and reporting averaged approximately 15 minutes per sample. In addition, a number of field samples were chosen and prepared using wet chemistry techniques. The wet chemistry sample preparation process is time consuming: the samples were dried overnight, ground, and sieved, then microwave digested and analyzed; after drying the rest of the process takes approximately 2 hours. These wet chemistry prepared samples were introduced into the ICP-AES using solution nebulization. The results from these solution nebulized samples were compared with the results obtained using the LA-ICP-AES technique.

While the MDLEST was developed over a very short time period prior to this demonstration, the system worked extremely well during field operations. From the lessons learned, a number of improvements were made that increased the precision of the analytical results, enhanced the field mobility of the system, increased the operational efficiency, and reduced the size of the operating team. Additional information concerning this demonstration can be obtained by requesting a copy of the formal report listed in the reference section.

Gunnison '93

The detection and quantitation of ^{230}Th and possibly ^{226}Ra levels in the soil after the uranium mill tailing pile has been removed was the focus of the demonstration during November 1993 at the Gunnison, CO, *Uranium Mill Tailing Remediation Action* project site, UMTRA. The ICP-MS is

capable of detecting and quantitating elements at the sub-parts per billion (ppb) level that fall within the 300 amu (atomic mass unit) range of this instrument. The levels of ^{230}Th and ^{226}Ra in the soil at the Gunnison site are expected to range from 0.05 - 1000 ppb (ng/g of soil), while the levels needed to satisfy the Nuclear Regulatory Commission (NRC) remediation agreement are ~0.80 and ~0.05 ppb (activity level of 15 pCi/g), respectively.

The ICP-MS was vibration damping mounted in the MDLEST and transported to the site. The LA sampling system in the surface sampling probe was used to ablate samples that were then injected into the ICP. The fiber optic cable delivery system was used to deliver the laser energy to the sampling probe. Due to poor weather conditions, snow and ice, and the close proximity of the automatic landing system, ALS, at the airport, the MDLEST was prevented from demonstrating the *in situ* sampling capability, instead samples were brought to the MDLEST in polypropylene bottles for analysis. Three *Nuclear Reference Materials*, NRM, samples with ^{230}Th activities of 13.5, 25.5, and 49.7 pCi/g were used to calibrate the LA-ICP-MS instrumentation. Analysis of the NRM samples showed the MS detection limit to be ~0.45 ppb. While this detection limit is satisfactory for ^{230}Th quantitation it is definitely above the detection limit for ^{226}Ra (0.05 ppb) as stated in the NRC remediation agreement, thus, ^{230}Th was the element of primary interest. Samples required approximately 20 minutes each to ablate, analyze, and report the results.

This demonstration showed that a standard production-model ICP-MS is sufficiently rugged to be installed in a completely self-contained mobile laboratory, be transported across the country, and be operationally ready to acquire data, on-site, in a few hours. The system operated in the field for nine days, under winter conditions (as low as -4°F), without a catastrophic failure.

A *global positioning system* (GPS), recently integrated into the MDLEST, was used to demonstrate the acquisition and use of satellite information, in real-time, for the determination of site map coordinates for off-grid sample points. Additional information concerning this demonstration can be obtained by requesting a copy of the formal report listed in the reference section.

Fernald '94

The focus of this June 1994 USID field demonstration hosted by FERMCO, Fernald, OH, was two fold: the evaluation of high resolution field screening tools acquiring, in a cost-time effective manner, uranium contamination distribution information in surface soils; and the testing of a cost/risk decision model to guide the real-time characterization of the site. The LA-ICP-AES sampling and analysis instrumentation was used to screen the soil samples. LA *in situ* sampling did not work since the ground was too dry, it had not rained at the site for more than three weeks, and it was impossible to make a gas tight seal with the powdery cracked soil, therefore, samples were collected in polypropylene bottles and brought to the MDLEST for sampling and analysis. The surface sampling probe was modified for this demonstration incorporating a compact laser, allowing ablation sampling using the direct beam delivery system. The compact laser performed marginally.

The ambient temperature, 95° F, outside of the MDLEST in the sun where the laser was operating affected the internal optical and beam adjusting components. After the demonstration was completed the laser was returned to the manufacturer for repair.

Three soil standards used for LA-ICP-AES calibration were prepared, on-site, by FERMCO personnel, by blending contaminated soil with site blank soil. Unfortunately, the simple blending was not as successful as the site personnel had expected so the activity levels of the mixed standards were 50-250% above the values expected. All of the instruments participating in the field screening demonstration were required to use these soil standards for calibration. The site field samples were analyzed and the results were printed, recorded on a floppy disk, and transmitted to Sandia National Laboratory for entry into the cost/risk decision model. This model determined the location of additional sampling sites to more fully define the extent of the uranium contamination. A formal report of this activity has not been written by the TIP team, however, a formal report on the demonstration results is expected to be issued by USID and FERMCO in the near future.

Denver '94

The objective of this demonstration was to determine the feasibility of obtaining data on subsurface soils *in situ* (directly), to field test the subsurface sampling probe and Layne tool, and to evaluate the current design. The demonstration was hosted by Layne Environmental Services Inc., LESI, at the Becker Drills Yard, Commerce City, CO, during August 1994. This field test was observed by personnel from the Rocky Flats site, local DOE Field Office, and Layne Inc. and LESI employees. A site containing only background levels of radioactivity was chosen for the field testing of the subsurface probe system to gain operating experience, without incurring the complications that can result when acquiring and analyzing radioactive samples. The subsurface *in situ* ablated soil samples were entrained in an argon gas flow, transported to the MDLEST, and immediately analyzed for heavy metals using the ICP-AES.

The initial field test of the prototype subsurface sampling system was very successful, with all systems performing as designed. Over the two day demonstration a number of samples were ablated and analyzed from two different bore holes. No cuttings were generated from either hole drilled (punched). The water table at this site (below 40 feet) was not encountered during this test. The LESI tool passed the critical test by consistently producing a gas tight seal (100%) with the surrounding soil at each level that a sample was acquired. Failure to produce an adequate seal would have inhibited sample transport to the MDLEST resulting in no sample to analyze. The operation of the percussion hammer driving the tool into the ground did not affect the instrumentation in the MDLEST. Standard soils, SRMs, were used to calibrate the LA-ICP-AES. A numerical method, using the macroelemental matrix of the standard soil sample, gave good results in determining the subsurface sample matrix. The results from the subsurface samples suggest a change in the strata at 16 feet from a clay type fill (sediment) to a sandstone that continued down to 25.5 feet where the bit was rejected (further drill advance ceased) due to a shale layer. This

result was confirmed by extracting a core sample of the strata. Personnel from the Rocky Flats site and the DOE Field Office observed the field test of this probe and were extremely interested in the possibility that this system could be available for use at their site.

This demonstration is the final milestone in the development and testing of the prototype subsurface probe and tool. This prototype was developed using off-the-shelf components to minimize the development cost. While this probe is functional, it has not been optimized in either size or function. This successful field test demonstrates the viability of the subsurface *in situ* concept, and the current prototype probe can be put to limited use. However, real time and cost efficiencies in subsurface characterization will be experienced only after further development makes the probe compatible for use with cone penetrometers and Geoprobes®.

SLAPS '94

The detection and quantitation of the isotopes of uranium, thorium, and radium, in particular; ^{238}U , ^{235}U , ^{232}Th , ^{230}Th , and ^{226}Ra , was the focus for the September 1994, *Saint Louis Airport Site*, SLAPS, demonstration hosted by the Ames Laboratory *Expedited Site Characterization*, ESC, program in Saint Louis, MO. A large group of participants (~105) representing federal, state, and local regulatory officials, stakeholders, managers and scientists from across the DOE complex, local educators, and private sector contractors and scientists attended the ESC program and visited the field site to observe the demonstrated technologies in operation.

The ICP-AES and the Perkin-Elmer ICP-MS were both installed in the MDLEST and integrated into the operating system so that both instruments could be operated simultaneously. While one instrument was analyzing samples acquired using LA sampling; the other instrument was analyzing solution nebulized samples prepared using wet chemistry techniques (see discussion above in Fernald '92). Mr. Daniel Zamzow from the Environmental Chemistry Group supported the field operation of the MDLEST at SLAPS by preparing the samples for solution nebulization and analyzing them using each of the spectrometer systems. SLAPS is the first direct comparison of results from LA sampling and solution nebulization on the same sample using the same analytical instrument. Using either the LA or solution nebulization sampling, total uranium and total thorium concentrations in the soil is given by the ICP-AES analysis, while the ICP-MS quantitated the uranium and thorium isotopic concentrations, but is not sensitive enough to quantitate radium at the remediation level (15 pCi/g).

Each sampling technique and ICP-spectrometer system was calibrated using three NRM standard soils with different activity levels and a NIST SRM soil blank. A layer of fill dirt had been placed on the site to cover the contamination and protect the public. Thus, to screen/characterize the contaminated soil a manual core sampler was used to acquire sample cores ~36" long. The fill dirt was removed from these cores with the remaining soil being split and compounded (mixed) to obtain relative homogenous samples. These samples were brought to the MDLEST in polypropylene

bottles for LA ablation sampling, while additional sample splits were prepared for analysis using solution nebulization.

The compounded SLAPS samples had higher radioactive contamination levels (radioactivity levels $\gg 100$ pCi/g) than those measured during demonstrations at other sites. Using the LA sampling system to sample high level radioactive contamination caused a cross-contamination problem, not previously observed, in the analyzed results. The problem was traced to ablation "dust" collecting on the window of the cell, and then being removed by the laser during the acquisition of the next sample, creating an uncertainty in the results. This was not a significant problem, once recognized, since the solution is to simply run a blank soil sample until the results return to normal.

The repaired compact laser, mounted in the surface sampling probe, performed well at SLAPS providing the ablation sampling energy. The problems encountered using the compact laser during the Fernald '94 demonstration did not reoccur during the SLAPS demonstration.

The TIP team has not submitted a formal report for this activity, however, the results will be reported in the SLAPS demonstration formal report that will be issued in the near future by the TIP ESC group.

Technology Fair '94

The Technology Fair was hosted by FERMCO in Cincinnati, OH, September 27, 1994. The purpose of this exposition was to display and demonstrate technologies that have, or will participate in the future, in the cleanup program at the Fernald, OH, site. These technologies included primary and secondary soil treatment remediation processes, real-time soil screening/characterization instrumentation, bench-scale soil treatment studies, risk analysis (environmental decision making), and technology transfer opportunities.

The MDLEST participated in the Fair displaying and demonstrating the capabilities of the mobile LA-ICP-AES/MS instrumentation, and the surface and subsurface sampling probes. The Fair was attended by approximately 175 individuals representing: DOE managers from headquarters, field offices, and site personnel; remediation contractors, EPA regulators, private sector companies, stakeholders, and the public. It was hoped that this technology exposure would lead to private sector interest in commercialization of several of these technologies.

Ablation Sampling of Concrete

The focus of this test was to determine the feasibility of using the surface sampling probe to *in situ* ablate a concrete surface and analyze the ablated sample to determine whether or not the concrete had been exposed to contamination and at what level. The concrete walk outside the TIP shop was used for this test. The fiber optic cable delivered the laser energy to the sampling probe and the ICP-

MS was used to analyze the samples. Two different wavelengths of laser light, 1064 nm and 532 nm, were used to ablate the concrete surface. The fiber optic cable easily handles, without damage, 25 mJ of 1064 nm laser light, however, this wavelength and energy level produced large ablated particles that quickly plugged the skimmer cone in the MS. Four millijoules of 532 nm laser light was sufficient to produce small sample particles that were easily analyzed by the MS with minimal skimmer cone plugging. The preliminary results from this test has shown that this technique is feasible for screening concrete floors and pads *in situ* to determine if they have been contaminated by accidental spills, leaking waste storage drums, or tanks.

Current screening techniques require the removal of concrete suspected of being contaminated, then samples are taken, prepared, and analyzed to determine the extent of the contamination. This unnecessary destruction, of possibly non-contaminated floors and pads, is time consuming, expensive, creates a dust hazard, adds to the amount of waste that needs to be handled and disposed. It also dilutes the level of contamination, thereby, interfering with the detection limits and quantitation of the contaminants. The laser ablation technique can sample the surface of the concrete *in situ*, identify the areas and levels of the contamination, without the physical removal of any concrete (a non-destructive screening). Remediation is started after the contaminated areas have been identified, resulting in a highly cost and time efficient operation.

Anticipated Savings/Benefit:

Current costs for conventional sampling and analysis is quite high, \$1200-2500 per sample, depending on the radioactivity level, the number of elements analyzed, the detection level desired, and the turn-around time required. Also included in this sampling cost is acquiring the sample at the site, packaging and transportation to the off-site laboratory for analysis, and disposal costs of the waste.

The MDLEST is parked on-site, the instrumentation is aligned and calibrated, and the system is ready to acquire and analyze samples in a few hours. Using the, currently installed, ICP-AES analysis system, each sample is analyzed simultaneously for 20 elements. The detection level for uranium for instance is 10 ppm (parts per million, approximately 1/5 the remediation level) with the results reported in 10 minutes. After the results for a number of samples from an area of the site have been acquired, a contour map is generated to help identify specific areas where additional sampling is needed to more precisely define the area and contamination level before any remediation takes place.

The MDLEST is operated by a crew of four, two technicians and two scientists. In the surface sampling configuration, 20 field samples can be completed a day at a cost of \$300-500 per sample. These costs reflect multielement determinations, amortization of the capital cost of the MDLEST and associated equipment, and covers all operating expenses. This is a 75-80% savings over current conventional sampling and analysis costs. More important than the sampling and analysis cost

savings is the time saved in obtaining the results needed for remediation decision making. While the sampling results from the MDLEST are available in approximately 10 minutes, results from off-site conventional laboratories can take from three days to several months. With timely results and immediate field remediation decision making, contractors can plan for the efficient continuous operation of expensive heavy equipment reducing both the clean up time and the overall cost to the customer.

The operation of the subsurface probe requires the operating crew for the MDLEST described above, in addition to a two man crew to operate the percussion hammer drilling rig used to insert the casing and tool into the ground. With the development of a sampling protocol two samples per hour with immediate analysis is achievable at a cost of \$625-875 per sample. This per sample cost covers the complete cost of the MDLEST and percussion hammer drilling rig and all associated personnel. The same analysis instrumentation described above for the surface sampling system is used allowing each sample to be simultaneously analyzed for 20 elements. For cost comparison; a commercial Gamma Spectrometry/Neutron bore hole logging system, sampling bore holes already in place, cost \$9450 per bore hole to identify a discrete set of radioactive elements in the soil surrounding the bore hole. The results from this survey are available two weeks after the logging activity is completed.

Project Cost/Schedule:

The project to develop a mobile laboratory, the MDLEST, started in late FY91. FY92 was the first fully funded year. The initial program objective was to develop the mobile laboratory with surface sampling capabilities using the *laser ablation* sampling technique to directly inject samples into the ICP-AES for immediate analysis. The MDLEST is a completely self-contained, fully instrumented and automated mobile laboratory. It soon became apparent that a subsurface sampling capability would be as important as the surface capability. To pursue the subsurface capability a CRADA was signed between Layne Environmental Services Inc., Phoenix, AZ, and the Ames Laboratory (August 1992) to jointly participate in the design, fabrication, and testing of a subsurface environmental characterization system (CRADA NO. AL-92-01). \$400,000 was spent developing and testing the prototype subsurface sampling probe and tool. The successful field test, August 17-18, 1994, of the subsurface *in situ* sampling probe and tool completes the plan of work set forth in the CRADA.

Commercialization:

Layne Inc., the commercial partner in the development of the subsurface sampling capability, does not possess the capability to commercialize the complete system. However, if a system becomes commercially available, they might be interested in becoming a contract operator.

Several companies have expressed an interest in the mobile laboratory concept and the surface sampling capability (the subsurface capability is just now being reported), however, they have taken a wait and see attitude until the size of the customer base is determined. It is extremely difficult to

interest a commercial concern, in a state-of-the-art product that requires capital investment and additional development, before the customer base and number of units needed has been discretely defined.

The Technology Integration Program of the Ames Laboratory is capable of assisting in the integration of a limited number of both surface and subsurface sampling systems described above. This would make these systems immediately available to the DOE complex to speed the remediation effort and provide cost savings. As soon as a customer base and number of units needed is defined, TIP will be happy to participate in the transfer this technology to an industrial partner, and work with them to insure that the final product meets the required specifications for field screening/characterization.

Ames has filed patent applications for a subsurface probe design that would be compatible with cone penetrometers and on an *in situ* calibration system.

Regulatory Acceptance:

The ICP-AES/MS analytical system has been accepted by the EPA as a *certified laboratory procedure*, CLP, method for environmental contamination quantification when using solution chemistry techniques with aerosol nebulization introduction of the analyte into the ICP torch. This acceptance is not dependant upon location of the instrumentation. Therefore, the MDLEST can be operated as a CLP lab in the field when using solution chemistry techniques. This acceptance has not yet been extended to the laser ablation sampling technique of sample introduction. However, non-CLP acceptance does not prevent the LA technique from being a very effective tool when used as a screening technology.

Recommendations and Conclusions:

The foregoing description of the development activities, instrumentation, and various field demonstrations of the MDLEST, the laser ablation sampling system, and the surface and subsurface sampling probes, both technical and applied, is the result of experience acquired during the evolution of the prototype systems described. Problems appeared during each of the field demonstrations, that were not perceived in the laboratory, but were successfully solved in the field and did not prevent the completion of the demonstration. In fact, these field engineering modifications have led to a more reliable system and will be incorporated into future models. These engineering modifications were not, nor could they have been anticipate, even with an infinite amount of shop testing. The field demonstrations, while performed under a variety of adverse weather and field conditions, will result in more ruggedized equipment and instrumentation with increased sensitivity for use in either a fixed or mobile laboratory environment in the future. Notwithstanding, sampling techniques, protocols, and instrument operating software have all been severely challenged during this development process. And finally, all of the personnel, engineers, scientists, technicians, and

students working as a team during this development process have learned to use utilities, instrumentation, and control systems in non-routine ways to successfully develop and field demonstrate the operational prototype systems described.

The MDLEST is a prototype instrumentation test bed with designed flexibility to allow rapid transitions between various types of instrumentation to satisfy site characterization needs. Because of this generalization of operation, the first model of the MDLEST is physically large. With the engineering advancements made during the development and operation of this field deployable laboratory, and with the improvements currently available in compact lasers and instrumentation with increased sensitivity, a smaller less costly version of the MDLEST could be assembled. Thus, making high precision quantitative instrumentation for rapid characterization readily available on-site to remediation managers.

The development of the laser ablation sampling system has identified areas of research and development that haven't been fully exploited. As mentioned above, the ablation cell has gone through a series of optimization experiments to improve the sample particle generation and entrainment that has already resulted in increased spectrometer signal intensity. Further improvements in signal intensity are possible if cell design parameters, particular cell volume and geometry are studied and optimized. In addition to cell design, the gas flow and transport tube parameters can be improved for efficient particle transport, and the ICP torch design needs to be optimized for the LA sampling technique (direct particle injection). Improvements in the raster pattern of the laser energy across the sample has the potential to increase both the ablation efficiency and stability of the sample particle stream generated. These improvements will result in improved detection limits and consistency of the analyzed results, and allow the ablation sampling system to satisfy the rigorous scrutiny of regulatory acceptance.

Sample acquisition time reduction with rapid analysis resulting in subsurface characterization cost efficiencies, will be experienced with further development of the *in situ* subsurface sampling concept, successfully demonstrated in the prototype, and adapting it for use with cone penetrometers and Geoprobos®. The demonstration of *in situ* non-destructive sampling of a concrete surface with immediate analysis, should be valuable to a number of DOE operations that have large areas of concrete that needs to be screened for contamination. Additional development of the prototype demonstrated would result in a specialized probe that could rapidly test large flat areas of concrete, resulting in cost and time efficiencies over the technique currently being used, coring the surface and sending the samples off-site for analysis. One of the important, non-trivial, design problems that has to be solved for this probe to operate rapidly is the design of a dynamic gas tight seal to seal the moving ablation cell to the concrete surface. With the creativity displayed above in the design and prototyping of the MDLEST and sampling systems this dynamic gas seal should not present an impossible problem to solve.

Finally, sample matrix variations, between prepared soil standards and surface soil samples, and subsurface soils (strata), presents a problem when quantitating contaminants. This issue has not

been fully addressed nor is a solution obvious, research into this area is sorely needed. We have used techniques currently used in the laboratory for solid samples, however, they are inadequate to give good consistent results on soils because of the extreme variability. The LA method of sample introduction is not a currently accepted CLP procedure, however, with additional research and incorporation of the improvements pointed out in this report it is possible that certification will become reality in the near future.

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