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RESULTS OF MOBILE GAMMA SCANNING ACTIVITIES IN ST. LOUIS, MISSOURI

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ABSTRACT

From 1942 through approximately 1966, the Mallinckrodt Chemical Works operated four plants in St. Louis, Missouri, for the Manhattan Engineer District (MED) and the Atomic Energy Commission (AEC). A variety of production processes using uranium- and radium-bearing ore materials were performed at the plants. It is the policy of the U. S. Department of Energy (DOE) to verify that radiological conditions at such sites or facilities comply with current DOE guidelines. Guidelines for release and use of such sites have become more stringent as research has provided more information since previous cleanups. The Formerly Utilized Sites Remedial Action Program (FUSRAP) was established as part of that effort to confirm the closeout status of facilities under contract to agencies preceding DOE during early nuclear energy development.

Under the FUSRAP program, the Mallinckrodt properties have been previously investigated to determine the extent of on-site radiological contamination. At the request of DOE, Oak Ridge National Laboratory (ORNL) conducted a survey in May, 1990, of public roadways and suspected haul routes between the Mallinckrodt plant and storage sites in St. Louis to ensure that no residual radioactive materials were conveyed off-site. A mobile gamma scanning van with an on–board computer system was used to identify possible anomalies. Suspect areas are those displaying measurements deviating from gamma exposure rates identified as typical for radiologically un-enhanced areas in the vicinity of the areas of interest.

The instrumentation highlighted three anomaly locations each of which measured less than 1 m^2 in size. None of the slightly elevated radiation levels originated from material associated with former AEC–related processing operations in the area. The anomalies resulted from elevated concentrations of radionuclides present in phosphate fertilizers, increased thorium in road–base gravel, and emanations from the radioactive storage site near the Latty Avenue airport.

RESULTS OF MOBILE GAMMA SCANNING ACTIVITIES IN ST. LOUIS, MISSOURI*

INTRODUCTION

From 1942 through approximately 1966, the Mallinckrodt Chemical Works operated four plants in St. Louis, Missouri, for the Manhattan Engineer District (MED) and the Atomic Energy Commission (AEC). In 1954 the contract was transferred to the AEC Oak Ridge Operations and remained there until 1966 at which time the contract was terminated.[1] The facilities were shut down for decontamination in 1957 after which some of the operations were resumed until the contract ended. A variety of production processes using uranium- and radium-bearing ore materials were performed at the plants. Operations included the production, conversion, extraction, concentration, purification, and reworking of uranium compounds; the machining, casting and recasting of uranium metal; and the recovery of scrap uranium metal. Thorium-230 was extracted and concentrated from pitchblende raffinate, and ^{226}Ra and its daughters were extracted from ore. Process wastes and residues resulting from the operations were transported from the plants for storage at a government site near the Lambert-St. Louis International Airport (SLAPS).

It is the policy of the U.S. Department of Energy (DOE) to verify that radiological conditions at such sites or facilities comply with current DOE guidelines. If they are found to significantly exceed those guidelines, remedial action may be implemented (where DOE has the authority to do so) to correct the unacceptable condition. Subsequent to original assessments and the release of these facilities, guidelines for release and use of such sites have become more stringent as research has provided more information since previous cleanups. Furthermore, documentation is sometimes limited or nonexistent, and release conditions at a specific site may be unknown. DOE established the Formerly Utilized Sites Remedial Action Program (FUSRAP) as part of the effort to confirm the closeout status of facilities under contract to agencies preceding DOE during early nuclear energy development.

The Mallinckrodt plants, vicinity properties, and some haul roads in St. Louis have been previously investigated under FUSRAP to determine the extent of on-site radiological contamination.[1-6] As a follow-up to earlier investigations and as a precaution to insure that no residual radioactive materials were conveyed to other off-site areas, DOE requested that ORNL conduct an additional survey in the vicinity of the SLAPS and the Mallinckrodt plant (Fig. 1). The survey was restricted to public roadways and to suspected haul routes used when wastes were transferred from the Hazelwood Interim Storage Site (HISS) at 9200 Latty Avenue to West Lake Landfill, and from the Mallinckrodt Plant to the SLAPS. Public roadways and accessible commercial parking areas surrounding the Mallinckrodt site within approximately one-half mile and railroad crossings on the appropriate roads were also scanned (Fig. 2). The purpose of the survey was to assess whether

*The survey was performed by the Measurement applications and Development Group of the Health and Safety Division of Oak Ridge National Laboratory under DOE contract DE-AC05-84OR21100.

any residual radioactive material could be detected along any of the routes. Although several haul routes near the airport storage sites were previously surveyed with the mobile gamma scanning van, selected intersections and/or roads near the airport radioactive storage site were also investigated during this survey. The survey was conducted May 15–17, 1990. Figure 3 shows the haul routes investigated by ORNL in 1990 and 1985, and diagrams the roads characterized by Bechtel National, Inc. (BNI) in 1990.

SURVEY METHODS

A brief description of the methods and instrumentation used for the mobile scanning of the described areas follows. Standard operating procedures for the mobile gamma scanning van were used to conduct the survey.[7] A more detailed description of the system and operation is provided in Myrick et al.[8] Anomalies were verified in the system's identification mode. The scoping approach used in this scanning method is described in *Procedures Manual for the ORNL Radiological Survey Activities (RASA) Program*, Oak Ridge National Laboratory, ORNL/TM-8600 (April 1987).[9]

MOBILE SCANNING METHOD

The initial step in searching for gamma radiation anomalies with the scanning van is to obtain the baseline (background) data with which to compare measurements taken in the relevant areas. Background exposure rates were measured along thoroughfares in similar, but radiologically unenhanced regions in the vicinity of the areas of interest. Scans were then performed of the suspect areas at a slow speed (<5 mph), minimizing the distance between the detectors and the properties. All accessible areas were scanned in both directions to maximize the number of views obtained for each surveyed property. Anomaly locations were highlighted by the computer system when the preset "hit" criteria, described as follows, were exceeded during the scan.

INSTRUMENTATION

The gamma radiation detection system employed in the ORNL scanning van is operator-controlled through keyboard instructions to an on-board computer on which data output is displayed. The data can be simultaneously printed and is stored on a dual floppy disk drive. The system consists of three 4 × 4 × 16-in. NaI(Tl) log crystals housed in a lead-shielded steel frame that is mounted on the right side of the van to provide two detector surface areas for acceptance of gamma radiation. A 12 × 16-in. surface measures radiation coming from sources on the right side of the van. The second surface is 4 × 12-in. in size and detects radiation from directly beneath the van. The detector and shield height can be varied with a hydraulic lift mechanism to optimize the detector field-of-view. The detector output is transferred to a computer-controlled eight-channel discriminator and interface that provides for continuous analysis of data inputs for correlation of system location with count rate information. Separate energy regions-of-interest are analyzed and a radionuclide-specific algorithm is employed to identify locations containing residual radium materials. Multichannel analysis capabilities are included in the system for additional qualitative radionuclide identification.

The algorithm used for data analysis compares the observed count rates arising from certain naturally occurring radionuclides to those produced by residual radioactive materials. Three regions of interest for ^{226}Ra (609 keV, 1120 keV, and 1764 keV energy peaks from ^{214}Bi) and one for ^{232}Th (2614 keV from ^{208}Tl) are analyzed. A Ra/Th ratio is computed from the observed background (bkg) observations and this ratio is used for comparison with scan data obtained from the suspect areas. In identifying locations containing residual ^{226}Ra-bearing material, the system utilizes criteria ("hit" criteria) based on the observed background Ra and Th count rates and the computed Ra/Th ratio. The "hit" criteria used are the following:

(1) a minimum count rate that results in a change in the background Ra/Th ratio;

(2) an observed Ra/Th ratio that is greater than the background Ra/Th ratio

$(Ra/Th_{bkg} + \sigma_{Ra/Th_{bkg}})$; and

(3) the positive difference between the observed Ra count and the background Ra count

{i.e., the difference = observed Ra - [observed Th $(Ra/Th_{bkg} + \sigma_{Ra/Th_{bkg}})$]}.

A hit is recorded when the first criterion and either the second or third criterion are met.

DETECTOR SENSITIVITY

The sensitivity of the detectors was determined by comparing the detector system response to its position at various distances from a point ^{226}Ra source and to different concentrations of ^{226}Ra in surface soil.[9] Exposure rates versus distance for these particular sources were well documented. The 1.0 mg ^{226}Ra point source, placed at 2 m above the ground surface (detector height), could be statistically detected at a distance of 65 m. This corresponds to an ~2% (0.25 μR/h) increase in normal background radiation levels. The sensitivity of the system was also found sufficient to detect 35 pCi/gm of ^{226}Ra in a 23 kg soil sample at ground level at 3 m distance from the detectors. Obviously, ease of detection increases with increase in total area and volume of ^{226}Ra-contaminated soil. However, the instrumentation cannot provide information regarding mass or quantity of contamination. Thus, a "hit" from a large area of elevated radiation levels at some distance from the detector will appear to be the same as a very small spot of highly radioactive material directly beneath the detector. It should be noted that no attempt has been made to establish a calibration factor for the scanning system since, during actual application, several factors are site-dependent. These include the source-to-detector distance, the counting geometry, and the potential for significant shielding caused by structures or buried material between the contamination and the detectors. An additional limitation is the fact that ^{230}Th, an occasional contaminant in wastes resulting from activities such as those under discussion, does not emit gamma radiation in amounts detectable by the type of instrumentation used for scanning. Elevated concentrations of ^{230}Th to a maximum of 3500 pCi/g were found along with ^{226}Ra and ^{238}U in soil samples taken from anomalies discovered during the 1985 ORNL van scan of haul roads.[5]

SURVEY RESULTS

Analysis of the mobile scan data indicated the presence of anomalies at three locations. One of these was found to be due to increased thorium in road–base gravel. All anomalies detected at a fertilizer plant were attributable to the naturally elevated radium and potassium concentrations known to be present in phosphate fertilizers. Other "hits" were associated with the radioactive storage site on Latty Avenue (HISS) as would be expected from earlier surveys. Radioactive contamination along the fenceline surrounding the former storage site, on several private vicinity properties, along a ~4000 ft Latty Avenue construction corridor, and along some haul routes has been previously verified and the results of the investigations published.[2-6] Radiation levels and concentrations of radionuclides in soil samples from anomalous regions are detailed in References 1–6.

SIGNIFICANCE OF FINDINGS

The results of these scanning activities along suspected haul routes in the vicinity of the Mallinckrodt plant revealed no evidence for the presence of radioactive residuals related to past AEC operations. The anomalies detected at three different locations were attributable to ^{232}Th-enhanced road–base gravel, phosphate fertilizers, and emanations from the nearby SLAPS.

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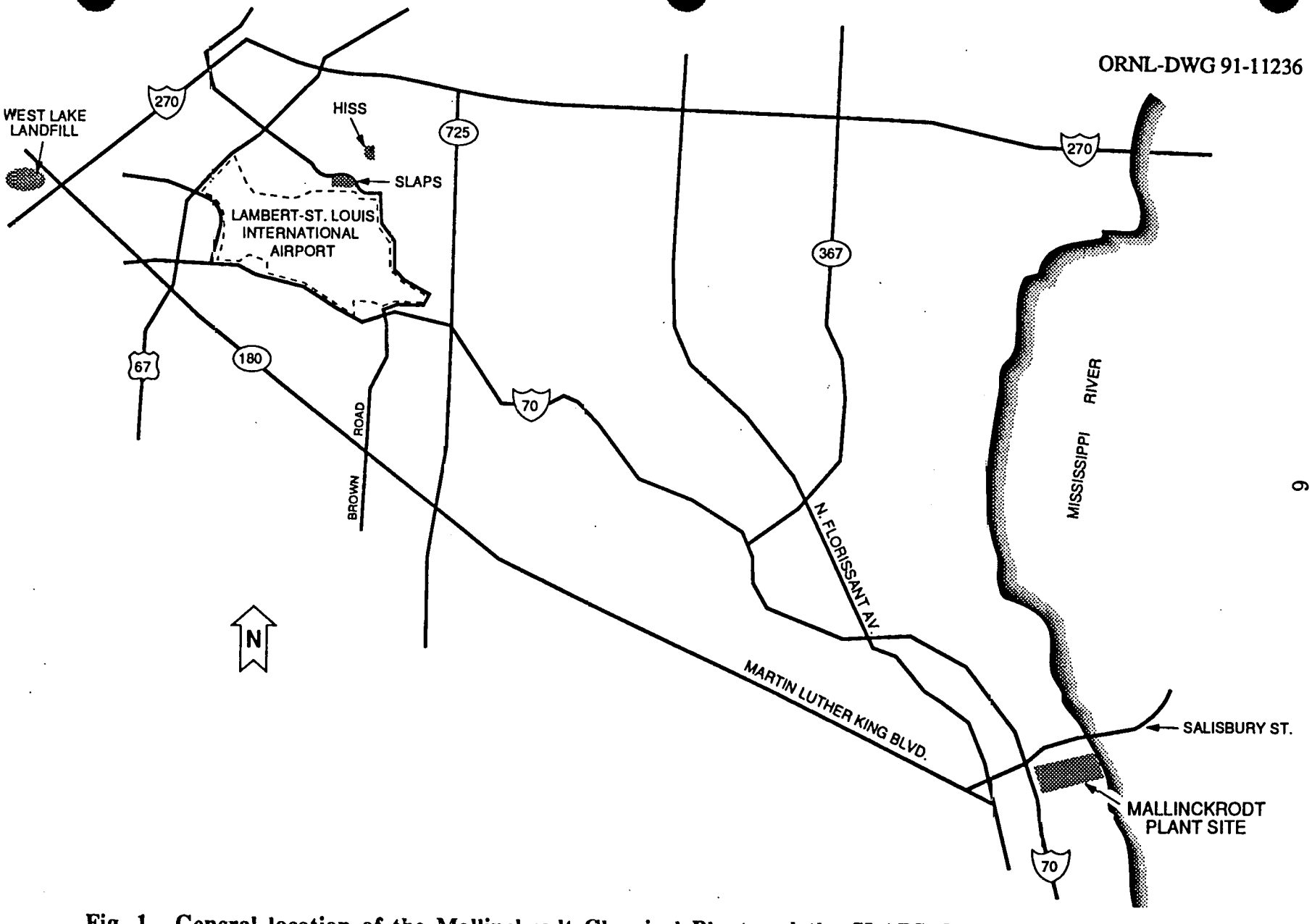

Fig. 1. General location of the Mallinckrodt Chemical Plant and the SLAPS, HISS, and West Lake Landfill storage sites in St. Louis, Missouri.

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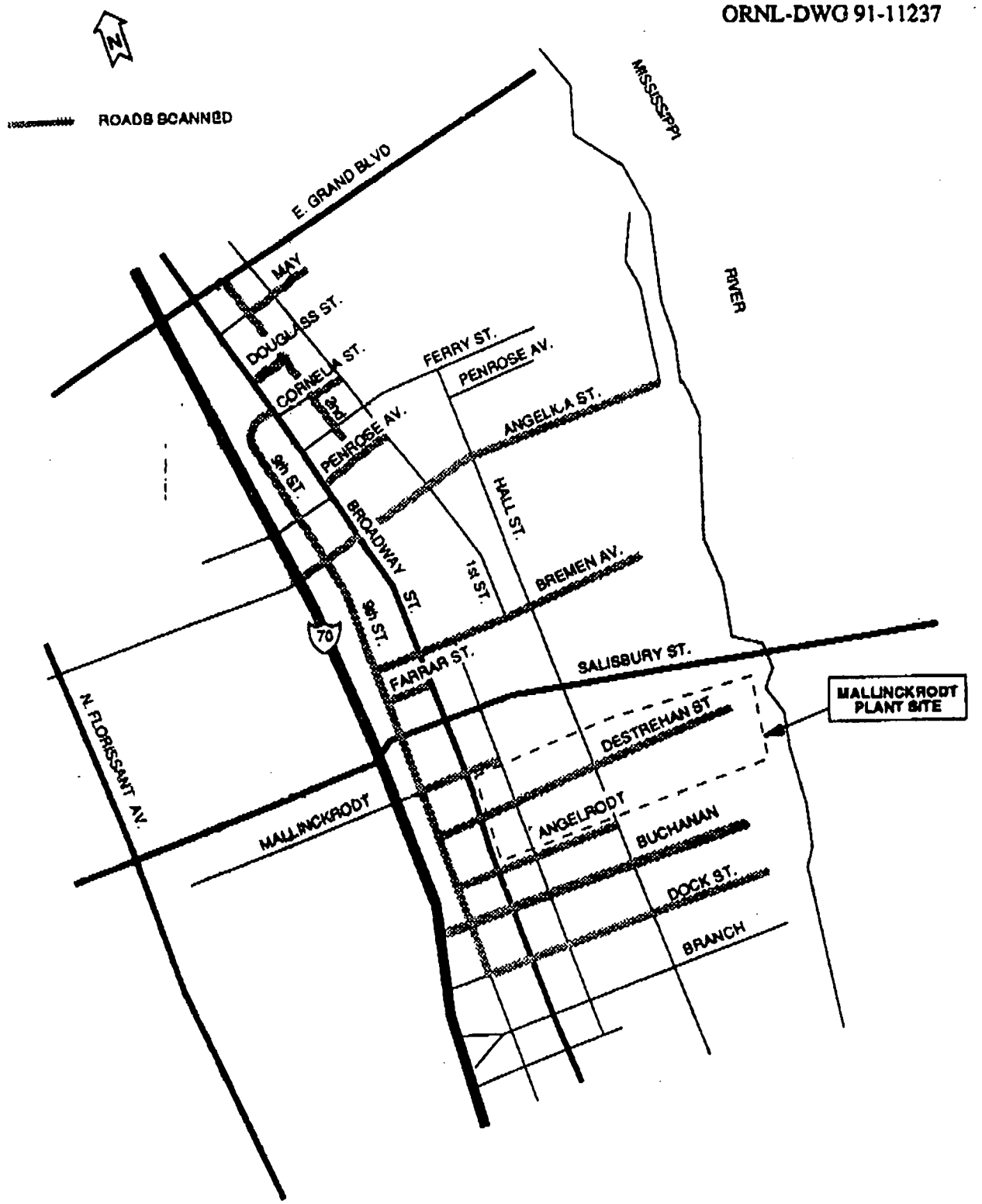

Fig. 2. Diagram of route scanned in the vicinity of the Mallinckrodt Chemical Plant site, St. Louis, Missouri.

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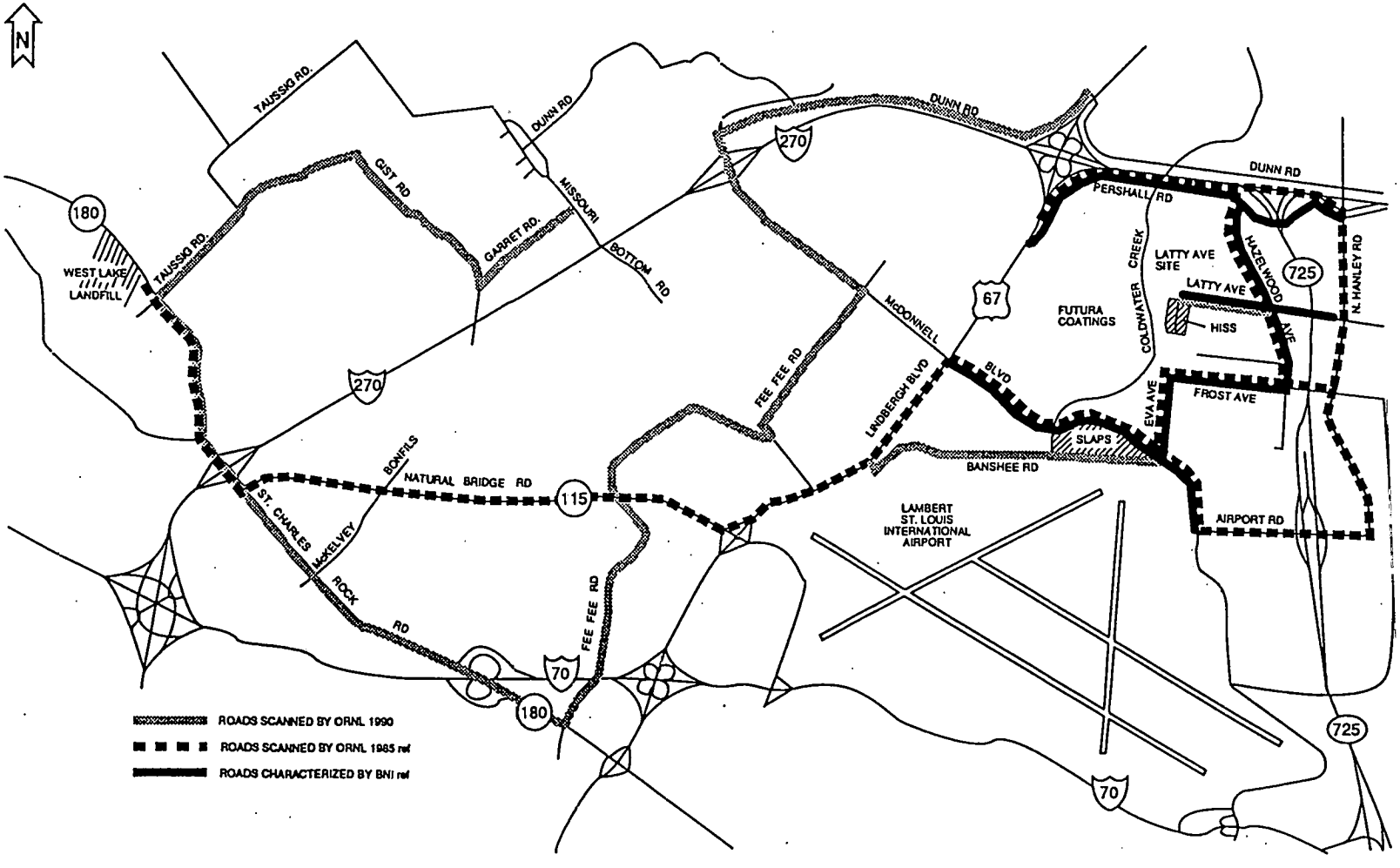

Fig. 3. Diagram of routes scanned by ORNL and routes characterized by BNI in the vicinity of the Lambert-St. Louis International Airport, St. Louis, Missouri.

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INTRODUCTION

The St. Louis Downtown Site (SLDS) is an active chemical plant owned by Mallinckrodt Specialty Chemical Company (MSCC). From 1942 to 1957, the facility manufactured various uranium compounds under contract to the Manhattan Engineer District (MED) and Atomic Energy Commission (AEC), predecessor organizations to the U.S. Department of Energy (DOE). The activities carried out for the MED and AEC resulted in the contamination of site soils and structures and the site was therefore included in the Formerly Utilized Sites Remedial Action Program (FUSRAP). The major radioactive contaminants identified were uranium-238, radium-226, thorium-230, and lesser quantities of thorium-232.

The site is located in the downtown area of St. Louis adjacent to the Mississippi River (Figure 1). There are ten (10) designated operating plants on the 18.2 ha site (Figure 2). Mallinckrodt intended to construct a bermed concrete pad within the Plant 7S portion of the MSCC. The plant 7S area contains areas where radionuclide soil concentration levels exceed the allowable DOE residual contamination limits; therefore, Bechtel National Inc. (BNI) the Project Management Contractor for FUSRAP conducted a response action in support of Mallinckrodt. The response action was confined to the pad construction area and included sampling and analyzing soils; excavating, packaging, and storage of contaminated soil; direct radiation scans and measurements; and post-remedial action sampling and analysis.

It is the policy of DOE to perform independent verifications of remedial actions conducted within FUSRAP. ESSAP has been selected as the organization responsible for this task at SLDS. Remedial actions were not completed during the September 17, 1992 site visit; therefore, ESSAP performed a Type A survey of the area.

DOCUMENT REVIEW

ESSAP reviewed both the RCRA Pad work plan and the post-remedial action letter report prepared by BNI.[1,2]

Document reviews indicated that the area had been adequately characterized, remedial actions were accurately described, and post-remedial action measurements and sampling were compared to the appropriate DOE guidelines.

CONFIRMATORY ANALYSES

ESSAP performed independent analyses of the post-remedial action soil samples collected, and analyzed by BNI's radiological contractor Thermo Analytical Inc./Eberline (TMA/E). Independent analyses of the three composite post-remedial action soil samples were performed at ESSAP's Oak Ridge Laboratory. Soil samples were analyzed by gamma spectrometry, for U-238, Ra-226, and any other identifiable photopeaks, and analyzed by alpha spectrometry for isotopic thorium. The comparative analytical results for the post-remedial action samples are

provided in Table 1 and indicate acceptable agreement between ESSAP and TMA/E analyses for each radionuclide. The maximum concentration levels determined by ESSAP analyses, for each radionuclide of concern were as follows: U-238, 8.8 pCi/g; Ra-226, 3.1 pCi/g; Th-232, 1.05 pCi/g; Th-230, 3.25 pCi/g.

SUMMARY

The analytical results were compared with the guidelines for residual radionuclides in soil.[3] The generic guidelines for Ra-226, Ra-228, Th-232, and Th-230 are as follows:

5 pCi/g	Averaged over the first 15 cm of soil below the surface.
15 pCi/g	Averaged over 15 cm thick layers of soil more than 15 below the surface.

Final site status following backfill and pad construction will result in the impacted soils being at a depth greater than 15 cm below the surface. Therefore the 15 pCi/g residual contamination levels for radium-226 and thorium would be applicable.

Guidelines for uranium are developed on a site specific basis. The uranium guideline developed for SLDS is 50 pCi/g of U-238.

It is ESSAP's opinion that the guidelines have been met and the area satisfies the requirements for release without radiological restrictions. This opinion is based on ESSAP's review of pertinent documents and data, and independent sample analyses.

TABLE 1
RADIONUCLIDE CONCENTRATIONS IN SOIL
COMPARISON OF POST-REMEDIAL ACTION SAMPLE RESULTS
ST. LOUIS DOWNTOWN SITE
ST. LOUIS, MISSOURI

Sample ID. No.	Radionuclide Concentration pCi/g							
	TMA/E				ESSAP			
	U-238	Ra-226	Th-232	Th-230	U-238[a]	Ra-226[a]	Th-232[b]	Th-230[b]
116V9201	<5.0	3.1 ± 0.2	1.2 ± 0.1	2.0 ± 0.7	4.8 ± 1.0	3.1 ± 0.5	1.02 ± 0.14	3.00 ± 0.24
116V9202	6.9 ± 3.0	3.2 ± 0.2	1.0 ± 0.1	2.1 ± 0.7	8.8 ± 2.8	2.4 ± 0.4	1.05 ± 0.14	3.22 ± 0.25
116V9203	6.6 ± 2.9	3.6 ± 0.1	1.2 ± 0.2	1.9 ± 0.7	7.1 ± 2.5	2.9 ± 0.5	1.01 ± 0.16	3.25 ± 0.28

[a]Analyzed by Gamma Spectrometry
[b]Analyzed by Alpha Spectrometry
[c]Uncertainties represent the 95% confidence level based only on counting statistics.

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