Recommended Sampling Strategies for Spatial Evaluation of Windblown Contamination Around Uranium Tailings Piles

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Operated by
Battelle Memorial Institute

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Commission

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ABSTRACT

Spatially-distributed ^{226}Ra concentrations in soil were statistically analyzed using Kriging to determine an optimum soil sampling strategy for detecting and delineating uranium mill tailings spread from storage piles by wind. Kriging is a geostatistical technique for analyzing data that contain information about the spatial location of sampling sites; it produces unbiased estimates of the inventory and spatial distribution of a material. A two-stage sampling plan augmented by Kriging is suggested. Its use would result in significant savings in sampling effort over that required to accomplish the same end with a single sampling or when attempting to locate areas requiring cleanup without the spatial distribution information provided by the Kriging process. Gamma-measurement techniques found in the literature that are currently used near uranium tailings piles are not appropriate for detecting and delineating windblown material that contains ^{226}Ra concentrations near background levels on properties within a few hundred feet of uranium tailings piles. Soil sampling with Kriging or possibly highly specialized in situ gamma measurement techniques (currently being developed) are more appropriate. Detection of hot spots requires that small grid spacings approaching the size of the hot spot of concern be used when sampling soil in order to attain a high (>75%) probability of detection. Differentiation between native materials containing uranium decay chain products and residual radioactive materials from uranium milling is best accomplished by using intrinsic germanium detectors onsite or in the laboratory to examine uranium (measured as ^{234}Th) to uranium decay chain daughter (^{226}Ra , ^{230}Th , ^{210}Pb) activity ratios.

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EXECUTIVE SUMMARY

Pacific Northwest Laboratory has reviewed sampling and radiological survey strategies in the literature that are applicable to the detection and delineation of windblown uranium mill tailings in the vicinity of uranium mill tailings piles. To support the requirements of the Uranium Mill Tailings Act of 1978, an optimum sampling strategy is proposed based on an analysis of published strategies reviewed, and a statistical analysis of an existing data base.

A portion of the ^{226}Ra concentration in soil data collected in the Cottonwood district of Edgemont, South Dakota, was chosen as the data base. Data suspected of being other than windblown contamination were not used. These data were collected as part of the Edgemont Remedial Action Program.

Linear regression analysis of delta measurements made in Edgemont near tailings piles with a collimated, lead-shielded gamma scintillometer (micro-R-meter) and ^{226}Ra concentrations measured in soil core samples gave low coefficients of determination (coefficient of correlation squared). These analyses indicated that collimated micro-R-meters may ascertain the presence or absence of contamination above-average background in the general vicinity of uranium mill tailings storage piles. However, the results also show that it would be more appropriate to use soil sampling or some other technique to make these same determinations for samples in the close vicinity of tailings piles.

Kriging, a geostatistical technique for estimating average concentrations at points or blocks over space, was applied to the Cottonwood data base. The spatial distribution and the total inventory of ^{226}Ra , of ^{226}Ra above background, and of ^{226}Ra above 5 pCi/g in the Cottonwood study area were estimated. It was also shown that for situations like that at Cottonwood, the relative error in concentration estimates for blocks made with Kriging increases very slowly with larger grid spacing. Hence, there is not much loss in efficiency due to using a large grid and few samples. However, a two-stage soil sampling strategy using Kriging would be required to be reasonably assured of differentiating residual radioactive material from 100-m² areas containing average ^{226}Ra concentrations greater than 5 pCi/g above background (the final EPA standard). A few samples on a wide grid spacing would identify areas where more intensive sampling should be performed. A second sampling on a smaller grid in areas selected from the first Kriging would accurately identify the 100-m² areas that exceed the 5 pCi/g standard. An illustrative example is shown that identifies areas in the Cottonwood tract which exceed the 5 pCi/g standard due to windblown tailings. Contour maps of ^{226}Ra in the study area, generated by the Kriging technique, are included. In the example, this two-stage sampling strategy would result in a 35% savings in sampling effort from that which would be required to accomplish the same end with a single sampling.

The detection of both hot spots and windblown tailings requires that a regular grid of sampling points be used and that the grid spacing be approximately equal to the longest dimension of the hot spot. This would find (with greater than 75% probability of detection) a circular or elliptical hot spot near or on the surface.

General procedures are described for differentiating between residual radioactive material, uranium byproduct material and native deposits of material naturally enhanced in uranium decay-chain products.

INTRODUCTION

The Uranium Mill Tailings Radiation Control Act of 1978 (Public Law 95-604, 42 USC 7901) was enacted by Congress to provide that "...every reasonable effort" be made to "provide for the stabilization, disposal, and control in a safe and environmentally sound manner of ... uranium mill tailings located at active and inactive mill operations (which) may pose a potential and significant radiation health hazard to the public..." (U.S. Congress 1978).

PL 95-604 requires that the Department of Energy (DOE) perform the necessary remedial actions at designated inactive uranium processing sites to achieve compliance with standards established by the EPA. These standards were published in the Federal Register (40 CFR 192) January 5, 1983 (U.S. EPA 1983). PL 95-604 also requires that the Nuclear Regulatory Commission (NRC) concur in selecting and performing remedial actions. One of the requirements of 40 CFR 192 is that "Remedial actions shall be conducted so as to provide reasonable assurance that as a result of residual radioactive materials from any designated processing site:

- (a) The concentration of radium-226 in land averaged over any area of 100 square meters shall not exceed the background level by more than -
 - (1) 5 pCi/g, averaged over the first 15 cm of soil below the surface, and
 - (2) 15 pCi/g, averaged over 15 cm thick layers of soil more than 15 cm below the surface" (U.S. EPA 1983).

Uranium mill tailings have historically been stored on or near the mill sites in large piles. Usually no stabilization procedures have been applied, making the tailings susceptible to being spread by the wind. The resultant contaminated areas may extend to several kilometers (Schwendiman et al. 1980). Estimates of contaminated areas (based on gamma-dose rate measurements, Douglas and Hans 1975) at 20 western mill sites indicate that tailings piles having areas of from a few acres (<10) to about 100 acres have been spread to additional areas of from a few acres (<10) to as much as a few hundred acres.

For remedial action to be completed at these tailings sites, radiological surveys must be performed to:

- (1) locate, identify and delineate the contamination requiring cleanup,
- (2) guide cleanup activities while underway, and
- (3) verify after remedial action has been performed that it has successfully satisfied the implementation guidelines.

Inherent in (1) above, is differentiation between displaced uranium mill tailings or ore (residual radioactive materials) and naturally occurring elevated concentrations of uranium chain products in geological formations (e.g. underlying clay layers and ore outcroppings). Guidance of cleanup activities must, by its nature, be flexible to accommodate unexpected, unplanned guidance needs. Communications between the cleanup (earth-moving) crew and the guidance (survey and monitoring) crew must be interactive, cooperative, and iterative to allow timely decisions based on evaluation and implementation of changing survey

and guidance needs. Verification of compliance with cleanup standards should be "...performed within the accuracy of presently available field and laboratory measurement capabilities and in conjunction with reasonable survey and sampling procedures designed to minimize the cost of verification." (U.S. EPA 1983).

The purpose of this report is to review selected existing sampling and survey strategies which have been suggested in the literature and/or used for radiological assessments for the purpose of detecting windblown uranium mill tailings in the vicinity of uranium mill tailings piles and to recommend a sampling strategy based on a statistical evaluation of an existing data base. A data base was selected which contained enough sample data points to be used to not only accurately define the ^{226}Ra concentration contours of the surface (upper 15 cm) soil, but also provide data to allow reasonable estimates of the background ^{226}Ra concentrations to be made. Based on the literature review and the selected data base, a recommended sampling strategy was developed and is presented in following sections to give reasonable assurance of detecting wind-blown residual radioactivity, differentiating between residual radioactivity (uranium mill tailings) and natural ore deposits, and estimating boundaries of areas requiring remedial action. General guidance is also given to assist in the differentiation of displaced uranium ore (also a residual radioactive material) from natural ore deposits or other natural mineral deposits having elevated concentrations of uranium-decay chain products. The recommended procedures apply to radiological surveys before and after remedial action for the dual purposes of determining the remedial action required and later determining compliance with the final EPA standards issued in 40 CFR 192.

It is always difficult to make plans and decisions based upon imprecise information about, or definition of, a problem. Judgment is required based on sound thinking and evaluation of existing facts. Surveying and sampling near uranium mill tailings piles to provide "reasonable assurance" of detecting, differentiating and delineating residual radioactive material and then verifying the decontamination of the affected areas while attempting to "minimize the cost" is an example of this type of situation. Because the standard requires only "reasonable assurance" of compliance with its specifications, then "The implementors (of remedial action) ultimately must make the judgement (SIC) whether or not a control system will meet the requirements." (U.S. EPA 1982).

LITERATURE REVIEW

The many literature sources reviewed preclude discussions of all sampling and survey protocols encountered. However, several pertinent studies will be briefly discussed. Other sources reviewed are included in a supplemental bibliography.

U.S. EPA

Douglas and Hans (1975) reported gamma radiation surveys conducted by the EPA at 20 inactive uranium mill sites to measure the extent of radioactive contamination spread into the environment from these sites by wind, water and milling activities. They concluded that... "hundreds of acres of land exclusive of the tailings piles have been contaminated to above-background levels." The following survey procedures were used.

Modified Baird-Atomic scintillometers (Model NE-148A) containing 1 X 1-1/2-in. NaI(Tl) detectors and equipped with removable bottom lead shields were used for the surveys. These instruments had previously been calibrated to ^{226}Ra on the ground surface. To decrease fluctuation, the time constant of the scintillometers was increased to ten seconds. In practice, two readings were made in contact with the ground: one with the lead shield in place between the crystal and the surface of the ground, and one with the shield removed. This technique enhanced the ability to detect that portion of the measured radiation originating beneath the instrument. The difference between the two readings was called the "delta" (Δ) reading, and was enhanced more by radiation from beneath the detector than by radiation originating from other directions. This radiation from the surface may be from tailings, displaced ore, natural deposits or outcroppings of ore, or from any other β - γ emitting radionuclide present on the surface. Unfortunately, it can also be due to Compton scattering in the soil surface of the incident gamma rays from a nearby radiation source (e.g. tailings pile) standing above the horizon. These degenerated γ -rays are detected by the scintillometer as having originated in the surface and therefore bias the delta reading high in spite of the theoretical design of the technique.

Delta measurements were made at the 20 inactive sites using a variety of locating techniques to establish those physical measurement points which fit defined criteria. Delta values which corresponded via cross-calibrations to predetermined post-cleanup exposure rates (at 3 ft above ground) were located (by search techniques) around 5 mill sites. Douglas and Hans (1975) correlated delta values with tailings surface contamination levels, assumed that the ^{226}Ra progeny were in equilibrium with the ^{226}Ra and consequently calculated the above-ground (3 ft) exposure rate resulting from a given contamination level. The predetermined post-cleanup exposure rates were background, 10 $\mu\text{R/hr}$ and 40 $\mu\text{R/hr}$. These were in keeping with then-current EPA/Office of Radiation Programs guidance for decontamination of inactive uranium mill sites. Iso-exposure rate points were connected to form iso-exposure rate contour lines around the mill site to be used to facilitate site decontamination decisions. At the other 15 sites, polar coordinate grids with radial lines were usually extended out from one or more control points located on the pile. In some cases, radial lines were extended outwards at right angles from base lines paced (or infrequently chained) around the pile perimeter. Delta measurements were normally made at 200-ft intervals along the radial lines until background readings were obtained. Iso-exposure points were then located on the radial lines and connected to form contour lines.

An increase in the background delta reading of one unit was interpreted as an indication of tailings contamination.

In residential and commercially developed areas, other techniques were used. The piles and mill sites themselves were not surveyed and no information of activity with depth was reported.

FORD, BACON AND DAVIS UTAH INC.

Ford, Bacon and Davis Utah Inc. (FBDU 1977-1981) reported data from existing radiological surveys done by the EPA, Oak Ridge National Laboratory (ORNL) and others including some new measurements (by FBDU) for over 20 inactive uranium mill sites in engineering assessment reports for the NRC and Department of Energy (DOE). Gamma measurements presented were made in several ways. The EPA post-cleanup iso-exposure lines (Douglas and Hans 1975) determined using a gamma scintillometer (unshielded except for a removable bottom lead shield) were reported as were uncorrected gamma dose rate measurements ($\mu\text{R/hr}$) made at 3-ft elevation with unshielded hand-held scintillometers. These dose rate measurements were usually reported for on-site locations, locations in the near vicinity, and background locations for the area. The locations of wind-blown tailings were estimated by making delta measurements (reported in units of c/m) usually at 1 in. from the surface using lead-shielded gamma scintillometers having one removable end shield to take the delta-type measurements. These delta measurements made with shielded instruments were used to estimate the 5 pCi/g ^{226}Ra soil concentration boundary (it had previously been determined that a Δ of 400 c/m was about equal to a ^{226}Ra soil concentration of 5 pCi/g). Measurements were usually made on 8 radial lines (range 5-17 radials) extending from the tailings piles or from base lines around the piles, with usually from 3 to 10 measurements made per line (range 2-10 measurements). These radial measurements were supplemented with rectangular grid measurements on selected portions of the site at a few mill locations.

For most of the mill tailings sites, soil samples were taken from the top 3 cm (surface samples) and from the 16 through 18-cm depth (6-in. samples) for the determination of ^{226}Ra concentrations onsite and in the near vicinity. An estimate of background concentration for each site was also made by taking offsite samples within a few miles of the site. Soil analysis results were reported in the FBDU series of engineering assessment reports. The usual number of surface samples taken onsite was about 18 (range 0 to 46), and the usual number of "6-in." samples taken was about 6 (range 0 to 12). The number of offsite samples and near-vicinity samples taken to estimate background varied from 3 to 22 but usually numbered about 11. Several samples were collected at other depths. The soil sampling spatial distribution strategies were not always obvious, and did not appear to be according to a set protocol. At some sites, the samples were collected in conjunction with the gamma exposure rate measurements on the radial sampling lines (e.g. Edgemont, South Dakota). At some sites the soil appeared to be randomly sampled around the perimeter of the site in the immediate vicinity (e.g. Salt Lake City, Utah), and at other sites the sampling appeared to have been done preferentially in the direction of expected contamination (e.g. Lowman, Idaho).

PACIFIC NORTHWEST LABORATORY

Schwendiman et al. (1980) measured ^{226}Ra concentrations in soil sampled around tailing piles in the Ambrosia Lake District. Samples were taken at the intersections of circular arcs (at 1/2, 1, 2, 3, 4, and 5 miles) with 16 radial lines (separated by $22\frac{1}{2}^\circ$) emanating from a tailings pile. At one tailings pile, 85 surface samples (0-5 cm) were taken at all accessible grid points, and 23 (0-24 cm) samples were also taken to determine downward migration of deposited materials. Isopleths of ^{226}Ra concentrations were drawn indicating that concentrations of ^{226}Ra (including natural ^{226}Ra) in excess of 5 pCi/g extended to about 5 km to the north and northeast, about 3 km to the southeast, and only about 1 to 2 km to the south and southwest. Analysis of depth profiles of ^{210}Pb concentrations at the grid points of one radial line (generally downwind) demonstrated the mixing and weathering of surface-deposited tailings to soil depths of 20 cm. Lead-210 concentrations decreased approximately exponentially with a half thickness of about 1.6 cm for the first 5 cm for up to 2 miles from the tailings pile. The decrease in activity slowed with depth until at 20 cm near-background levels of ^{210}Pb were reached for all distances from the tailings pile, indicating little, if any, mixing at that depth.

TECHNICAL MEASUREMENTS CENTER

In 1982, DOE established the Technical Measurements Center (TMC) at Grand Junction, Colorado, to meet the "...need for standardization, calibrations, comparability, verification of data, quality assurance, and cost effectiveness" for the Department of Energy Remedial Action Programs (TMC and White 1982). These remedial action programs include the Grand Junction Remedial Action Program (GJRAP), the Formerly Utilized Sites Remedial Action Program (FUSRAP), the Surplus Facilities Management Program (SFMP), and the Uranium Mill Tailings Remedial Action Program (UMTRAP). TMC and White (1982) reviewed, among other topics, soil sample collection and preparation procedures from soil sampling plan designs, and concluded that "The FY 1982 measurement plans contained very little information on these topics. Thus, a comprehensive review was not feasible. In general there is a lack of consensus on field sampling procedures for both water and soil. This includes both the number of samples and the quantity of each sample." Of documents received from six remedial action project sites for review, only two contained soil sample plan designs. A third site had been characterized by Battelle Memorial Institute (BMI), Columbus Laboratories, under contract to National Lead of Ohio (NLO), and their sampling protocol was also reviewed by TMC.

Battelle Memorial Institute

The Battelle (BMI) sampling at the Niagara Falls storage site (SFMP) was conducted on a square grid with grid nodes on 15-m centers (7.5-m centers where radiation was known to be high) over the 190-acre property. Surface soil (500 g) was taken at each grid node from the top 5 cm of soil.

National Lead of Ohio (Cannonsburg)

TMC and White (1982) reviewed a soil sample plan submitted by NLO as part of a draft Cannonsburg (UMTRAP) vicinity property radiological control plan. In this plan, the area to be sampled was subdivided into 10 x 10-m blocks and soil sampling was conducted at the center of each 5 X 5-m quadrant of the 10 X 10-m

area to a depth of 15 cm. The average ^{226}Ra concentration for each 100-m² area was compared to the radium criterion. In-situ gamma-ray measurements were also made in conjunction with the soil sampling. Guidelines for sampling "excavation areas" between 100 m² and 25 m² called for collection of one sample at the center of each 5 X 5-m survey block and a sample from 1-m offsets inside each of the four corners of the survey blocks. For smaller areas, 5 equally spaced samples were collected within the excavation boundary, with one being at the center of the excavation.

Bechtel National, Incorporated and National Lead of Ohio (Middlesex)

Procedures to be used at Middlesex (FUSRAP) submitted by Bechtel National, Incorporated (BNI) and NLO were evaluated. The NLO procedure was from earlier work at Middlesex, and the BNI procedure was generic in nature to be used at all other FUSRAP sites. The two procedures were similar and were apparently based on Holoway et al. (1981). The BNI plan for pre-remedial action surveys called for soil sampling and in-situ gamma measurements at the grid points of a square grid of from 5 m to 10 m on a side to yield a minimum of 30 sample points. In practice, BNI collects more than 30 samples per survey block. Remedial action excavation effectiveness was monitored primarily by gamma measurements verified by an occasional soil sample collection. Post-remedial action sampling called for a minimum of 30 soil samples. NLO used NaI(Tl) gamma detectors in survey blocks of 100 m² divided by a grid with 1 m intervals between sampling (measurement) points. If an area-average criterion was "met", the area was resurveyed on a grid with nodes at 2 m intervals using an EG&G IMP. (The word IMP and its variations as used in this report were derived from a trademark of the DeLorean Manufacturing Company.) The criterion to be met was not described in the review. The EG&G IMP is a high resolution gamma spectrometer utilizing a high purity germanium detector coupled to a 4096 channel analyzer and programmable calculator or microprocessor (Frieson 1982). Should the criterion be met again, soil samples were taken on the same 2-m grid for analysis.

TMC and White (1982) reported that the minimum statistical number of samples reportedly taken by BNI and NLO was derived from Equation 1.

$$n \geq 45 \left(\frac{S}{\bar{x}} \right)^2 \quad (\text{Equation 1})$$

where n is the number of samples, and $S/\bar{x}$ is the coefficient of variation, or the ratio of the standard deviation to the mean of the measurements taken. Equation 1 was derived by Holoway et al. (1981 p. 70).

TMC and White (1982) recommended that no modifications be made at that time to the overall design of the NLO and BNI soil sample plans due to the large number of measurements taken in each grid block and due to the practicality and flexibility of the procedures. However, they cautioned against placing too much confidence in Equation 1. They also gave some recommendations concerning techniques for sample collection and preparation prior to analysis.

Technical Measurements Center - Workshop

In April, 1983, TMC conducted a workshop at Niagara Falls to review a draft report "Surface Gamma-Ray Measurement Protocol for Small Parcels on Open Lands," (Marutzky, Steele and Key 1983). The protocol prescribed guidelines for gamma-

ray surveys of open land parcels of 10 acres or less that would enable a remedial action contractor to have reasonable assurance that a parcel of land had been cleaned up to meet the EPA radium in soil standard. Specifications, calibrations and uses of scintillation counters, scintillometer/micro-R meters, differential/delta counters, and portable spectrometers were included in the report. Soil sampling and analysis were advised against due to slowness of physical sampling, relative expense and its poor reliability in characterizing inhomogeneous soils. Assuming appropriate corrections are made to the data, in-situ radiometric assaying was recommended to improve the reported shortcomings of soil sampling. The corrections recommended for radium assay included natural background, a calibration factor for ^{226}Ra , and area-specific average correction factors for ^{40}K , ^{232}Th , gamma absorption due to soil moisture, and disequilibrium between radon daughters and ^{226}Ra in near-surface soil due to radon loss.

Measurement guidelines were included in the draft report which covered 1) determination of average background in the surrounding area, 2) the preparation for and conduct of a comprehensive radiological survey to detect contamination spread by man and wind, 3) survey activities required during contamination removal, and 4) a suggested survey plan for cleanup verification survey procedures.

The procedure recommended by TMC for spatial location of windblown contamination assumed this type of contamination to be "reasonably uniform" and as such would not require the intensive 100 percent continuous gross-gamma scan of each 10-m square-grid block recommended for detection of hot spots associated with contaminated material transported by man. TMC recommended gross-gamma measurements be made at each 10-m grid intersection to determine the approximate boundaries of the contamination. The estimated 5 pCi/g above background boundary could then be more closely located by using scanning techniques with gross-gamma instruments and "further quantitative measurements." The depths of the deposits were to be determined by making delta measurements at successive 15-cm depths to as deep as 60 cm or more. Borehole logging was suggested for deeper measurements.

A tentative post-remedial action verification survey procedure was recommended pending receipt of final verification requirements from the Remedial Action Program Office. The original survey grid was to be re-established to guide a gross-gamma scan of 100 percent of the excavated areas for detection of any residual contamination for immediate removal. Confirmatory measurements were described as consisting of ground and waist-level exposure rate and collimated gross-gamma or delta measurements at all intersections of a 5-m by 5-m grid superimposed on the 10-m by 10-m grid. Each excavated area was required to contain or be adjacent to a minimum of 4 grid point measurements. Verification that cleanup was satisfactory would require that the average of any four adjacent measurements be less than 5 pCi/g ^{226}Ra as estimated by measurement of its ^{214}Bi daughter. Representative soil samples would be taken and analyzed to verify correction factors used for ^{40}K , ^{232}Th and soil moisture.

OAK RIDGE NATIONAL LABORATORY

Holoway et al. (1981) of Oak Ridge National Laboratory have published recommendations for preliminary and termination survey designs and procedures for licensed nuclear fuel cycle and non-fuel cycle facilities. Specific recommendations and equations were presented to assist in subdividing the site into survey blocks, estimating the numbers and types of survey blocks needed, estimating the numbers of samples required to estimate the population mean of measurements and

deciding the types of measurements most beneficial to the objectives of the survey. The development of Equation 1 was reviewed and its use recommended. A generic application of the recommended monitoring program design and procedures to a uranium mill site was reported by Holoway et al. (1981) in an appendix. For windblown tailings, they recommend sampling on a 30-m grid, sampling at at least 30 grid points, and decreasing the grid spacing to 5 to 15 m in areas having gamma radiation levels twice background.

Black and Veatch

A procedures manual for the Remedial Action Survey and Certification Activities (RASCA) Program of Oak Ridge National Laboratory (ORNL) was prepared by the Southern Science Office of Black & Veatch, Dunedin, Florida, under sub-contract to ORNL. This manual includes specifications for layout and sizes of measurement/sampling grids, procedures for gamma exposure rate and radiation measurements and soil, sediment and water sample collection. However, at the time of this writing, the procedures manual had not been released for publication.

SUMMARY OF METHODS FOUND IN THE LITERATURE

In summary, the purpose for which a radiological survey is being performed and the known or assumed type(s) of distribution of contamination (based on prior knowledge, records, prior surveys or on expert judgment) determine to some extent the procedures recommended to be used. Other controlling factors are the time and funds available, the controlling regulatory or cleanup standards which must be met, and the degree of certainty desired that these standards be met.

Most of the surveys reviewed that were conducted in the vicinity of tailings piles for the detection of residual radioactivity employed gamma detection devices such as vehicle-mounted scintillation detectors, hand-held gamma dose-rate or count-rate instruments and/or pressurized ionization chambers. Problems were encountered with the interference of the radiation shine from nearby radiation sources such as the tailings pile or storage piles of ore. Gamma rays from the nearby relatively high level source(s) were detected as interference by the instrument being used to measure radiation from contamination on the ground near background levels. Techniques such as the delta-measurement procedure were employed to differentiate the gamma contribution originating beneath the detector from that coming from the tailings pile.

Although Marutzky, Steele and Key (1983) advised against soil sampling, it has been demonstrated in the field that up to 100 soil samples can be collected and analyzed for ^{210}Pb , ^{226}Ra , ^{230}Th and ^{238}U per day, and the results ready within 48 hours (Weimer, Kinnison and Reeves 1981). The samples were dried, pulverized and pressed into pellets for instrumental analysis in one of two planar intrinsic germanium detectors housed in a mobile laboratory. During 8 weeks in the period September 22, 1979, through December 13, 1979, PNL personnel analyzed 2400 soil samples in the PNL/NRC mobile laboratory van. The purpose of these measurements was to determine the effectiveness of the cleanup operations that were ongoing at Church Rock, New Mexico, and to evaluate required additional cleanup of uranium mill tailings.

Although some survey sites reported in the literature were revisited for further radiological surveys, the later surveys were usually for a different

purpose and so different measurements were made, or the later measurements were made with different instruments using different measurement techniques and following different protocols. Often subsequent surveys did not repeat measurements at past measurement locations, but rather added more measurements at new locations to augment the existing data. Therefore, very little data were found which could be used for temporal evaluation of sampling strategies.

GAMMA-RAY MEASUREMENTS NEAR TAILINGS PILES

Radiological survey protocols for locating fugitive radioactivity often call for gamma-ray measurements in the form of dose rate measurements, concentration measurements, count-rate measurements, or, as discussed earlier, delta measurements. These initial measurements are often followed by soil sampling, core sampling, bore-hole measurements and sometimes laboratory analysis of soil samples by wet chemical methods. The following discussion of an attempt to use gamma-ray measurements in the near vicinity of uranium mill tailings piles illustrates the lack of correlation generally observed between ^{226}Ra concentrations measured by in situ gamma-ray instruments and the concentrations determined by laboratory gamma-ray measurements of soil samples collected from the locations where the gamma measurements were made.

As mentioned in the literature review, in situ gamma measurements are complicated in the vicinity of high level sources of the gamma radiation being sought. An example of this is looking for residual radioactive material concentrations near background levels in the radiation "shine" from a nearby uranium tailings pile. These complications are demonstrated by the recent experience of PNL during the radiological surveys performed in support of the Edgemont Remedial Action Program (Edgemont, South Dakota).

During PNL radiological surveys of properties possibly contaminated by residual radioactive material from a uranium processing site in Edgemont, gamma dose-rate measurements were made using Ludlum micro-R-meters (Model 12S gamma scintillators) (Young, Jackson and Thomas 1983). Each property was treated as a separate survey block. Outdoor measurements on land with a permanent habitable building(s) were made at the grid points of a 7-ft square grid. For large lots, only that portion of the lot within 50 ft of a habitable building was surveyed using the 7-ft grid. The remainder of the larger properties and open land having no permanent habitable structures was divided by grids containing 4 survey blocks along the shorter dimension, and 5 survey blocks along the longer dimension of the property, unless additional survey blocks were required to keep the distance between measurements from exceeding 200 ft. Actually, the maximum spacing was normally about 100 ft. If the shape of the property was very irregular, 20 or more measurements were made on a grid pattern to give more uniform spacing within the area. Exposure rate measurements were made at an elevation of about 1 m at the center of each survey block. A serpentine walk was also made through each row of sampling locations making continuous waist-level gamma exposure rate readings to locate isolated small hot spots. The need for subsequent contact exposure rate measurements, soil sampling and/or more detailed engineering assessments (which included borehole logging and further soil analysis) was determined by comparing the results of the gamma dose rate measurements with the upper 95% limit of the EPA measured background dose rate (Thrall, Hans and Kallameyn 1980) to determine whether residual radioactive material was present. PNL measurements in Edgemont later verified the upper 95% limit of the background dose rates measured by the EPA (Perkins et al. 1981; Young, Jackson and Thomas 1983).

Exposure rate measurements taken in the Cottonwood area of Edgemont were affected by the radiation shine from the tailings pile, and therefore the decision levels based on the EPA background measurements which directed further sampling or engineering assessments were no longer valid. Exposure rates of over 1000 $\mu\text{R/hr}$ have been measured on properties near tailings piles. Delta measurements with unshielded micro-R-meters yield results which are differences between

large numbers and therefore are not very sensitive indicators of deposits of residual radioactivity near background levels. Young, Jackson and Thomas (1983) recommended the use of a modified micro-R-meter in which the detector was shielded on all sides by a lead collimator to reduce background by a factor of 100. A removable bottom shield would allow delta measurements to be made.

For the Edgemont Remedial Action Program, a gamma-ray collimator was constructed for use with the Ludlum Model 12S micro-R-meter. This modified instrument was evaluated for making waist-level delta measurements near the tailings piles in the Cottonwood district of Edgemont to help determine boundaries of deposits of residual radioactivity. Although it was recommended by Young, Jackson and Thomas (1983) that the collimator be constructed to encase just the detector and not the entire case of the scintillometer to reduce weight, this collimator was constructed to quickly slip over the micro-R-meter and give 2.54 cm of lead shielding to the front and two sides and a 2.54-cm removable bottom shield. The lead was beveled where possible to minimize the weight and allow the meter to be carried by its original handle. Figure 1 shows several views of the collimator and its geometric relationship to the internal scintillation detector.

The collimator reduced the background by about a factor of 2 or 3 depending on location. Readings were taken at most of the Cottonwood properties near the mill tailings sites with this shielded instrument following the normal grid survey protocols (Young, Jackson and Thomas 1983; Perkins et al. 1981) with the following modifications:

1. Since the collimated micro-R-meter readings could not be related to the free field exposure rate readings measured with a pressurized ionization chamber, no attempt was made to apply correction factors to the micro-R-meter readings.
2. Delta readings were taken at waist level (approximately 1-m height) with and without the bottom shield, but with the rest of the collimator in place.
3. At each location where a 5-cm thick soil sample was taken, a collimated micro-R-meter contact reading was taken in addition to the two readings at 1-m height.

The difference or delta (Δ) reading between the two 1-m readings taken at each grid point would be expected to emphasize photons originating beneath and a short distance to the sides of the detector. Local sources would also be expected to be largely responsible for the collimated meter readings made in contact with the ground.

Linear regression analysis was applied to 52 comparison measurements from the Cottonwood district data to test these hypotheses. Radium-226 concentrations in surface soil were the independent variables and delta readings at 1-m height were the dependent variables. The linear dependence of delta readings on ^{226}Ra concentrations gave a coefficient of determination, r^2 (i.e. linear correlation coefficient squared) of only 0.049, which is essentially the same as r^2 of 0.048 obtained for collimated measurements taken at 1 m with the bottom shield removed. Contact measurements with the shield removed showed slightly more dependence on ^{226}Ra concentrations in the top 5 cm with $r^2 = 0.14$.

FIGURE 1. Views of a Lead Collimator for Use with a Portable Micro-R-Meter

FIGURE 1: A series of 1659 collimated micro-R-meter readings were taken at 1-m height in the vicinity of the mill tailings storage areas. The readings were plotted against the local ^{226}Ra concentration data (Figure 2) and showed that about one-fourth of the data points had much higher delta measurements than predicted from the local ^{226}Ra content of surface soil, based on the remaining data points. Those higher delta readings were measured on properties within one city block of the mill tailings storage areas. Deletion of the 9 data points within 1/2 of a city block distance from the tailings areas (denoted by the symbol Δ in Figure 2) resulted in a considerably increased dependence of delta readings on ^{226}Ra concentrations ($r^2 = 0.42$). Eliminating, in addition, those points with a bottom shielded reading of over 10 $\mu\text{R/hr}$ (denoted by the symbol $\square$ in Figure 2 and indicating a possible contribution from the tailings pile shine) resulted in a correlation with $r^2 = 0.62$. However, the linear dependence of the readings taken at 1-m height at these same locations with the bottom shield removed on ^{226}Ra concentrations had almost the same coefficient of determination ($r^2 = 0.60$), indicating that in this case the added effort of taking delta readings was not justified.

These measurements taken in the vicinity of tailings storage piles indicated that collimated micro-R-meter readings may provide an evaluation of the presence or absence of contamination above the average background for all but those properties closest to the storage areas, but another technique would be more appropriate for the properties in the close vicinity of storage areas.

One alternate technique would be the collection of 30-cm deep soil samples in 15-cm segments to allow the detection of surface contamination and to verify that contamination has not mixed below the top 15 cm. Should average ^{226}Ra concentration higher than the standard be found in the lower 15-cm sample, further investigation would be required. Analysis of these soil samples in the field would allow determination of ^{226}Ra concentrations in depth intervals consistent with the EPA final standards for ^{226}Ra in soil. In addition, determination of other uranium daughters would allow differentiation between uranium tailings and natural deposits of material containing enhanced uranium concentrations or displaced ore. This sampling technique, using a 2-in. diameter split-spoon sampler, was used in the Cottonwood district, which is adjacent to tailings storage areas.

FIGURE 2. Delta Measurements at One Meter Height Versus ^{226}Ra Concentrations in Cottonwood District Surface Soil

THE SOIL SAMPLING DATA BASE FOR STATISTICAL ANALYSIS

A data base with the following characteristics was desired to develop a recommended soil sampling strategy for the detection and delineation of windblown uranium mill tailings in the vicinity of uranium mill tailings storage piles.

- 1) The area sampled was adjacent to uranium mill tailings piles.
- 2) The contamination in the soil sampled was uranium mill tailings.
- 3) The contamination was deposited primarily by the action of wind on tailings piles.
- 4) The area of probable contamination was relatively unpopulated (i.e. a minimum of large obstructions such as commercial buildings).
- 5) An extensive soil sampling program had been completed according to a rigid sampling protocol.
- 6) Radium-226 concentrations had been determined for surface samples and subsurface samples at a large number of the soil sampling locations.
- 7) The area had been resampled by the same protocol on several occasions over as long a period of time as possible to allow temporal analyses to be made.
- 8) As much other radiological and physical information was available as possible for each of the sampling locations in order to better interpret the results of the sampling.

The literature review had revealed that many radiological survey data sets possessed the first four characteristics, but not the remaining four. Fortunately, PNL was engaged in completion of a survey program at Edgemont, South Dakota, which not only satisfied the first four, but also the fifth, sixth and eighth needs. In addition, the persons performing the surveys, samplings and analyses were readily available for consultation on questions related to the data. A further feature of the data from the Cottonwood district of Edgemont was that since they were typical of windblown tailings situations, any data analysis conclusions obtained from these data should be applicable to the design of surveys for other problem areas.

The Cottonwood district lies just east of Edgemont proper (Figure 3). Several tailings piles and ponds from the Tennessee Valley Authority (TVA) mill are adjacent to the Cottonwood properties on the east, north and northwest (Figure 4). The edge of tailings pond 10 lies about 50 ft southeast of the southeast corner of the Cottonwood study area. Pond 7 lies along the entire east boundary of the study area, pond 4 is about 450 ft northeast by north, and the east sand tailings pile is about 800 ft north by northeast from Cottonwood. Sand tailings area B is about 400 ft north, the mill site is about 900 ft north by northeast and sand tailings pile area A is only 100 ft northwest of Cottonwood. Sand tailings pile area A is also upwind in the direction of prevailing winds (FBDU 1977-1981 b). The 9-block study area from which the data base was collected is also shown in Figure 4. This 9-block area is about 1200 ft (366 meters) on a side or about 33 acres (134,000 m²).

FIGURE 3. Diagram of Edgemont, South Dakota, and Vicinity

FIGURE 4. Diagram of Cottonwood Study Area and Immediate Vicinity

STATISTICAL ANALYSIS

The health-physics literature seems to contain as many methods of analyzing tailings contamination data as there are authors of papers. These methods tend to ignore the spatial information in the data and use statistics designed for analyzing independent replicates, or to statistically analyze the data for each property within an area to be evaluated separately and independently of all other properties. The statistics found in the geography and geology literature show more of a consensus on methodology. In these disciplines a general methodology of analyzing spatial data, called Kriging, has become accepted. A recent textbook (Clark 1979) gives a relatively simple explanation of Kriging.

METHODS

Kriging is a geostatistical technique for analyzing data which contain the spatial coordinates of sampling sites. Samples that are collected close together, spatially, intuitively should be correlated and samples collected far apart should be independent. Kriging makes use of estimates of such a correlation to obtain unbiased estimates of the inventory and spatial distribution of a material. The variation in the concentration with distance between measured points is used to calculate the concentrations at unmeasured locations. The results are typically presented as a contour map of location versus concentration. The concentration at all points in an area of interest can be integrated to yield inventory. A very useful feature of Kriging is that it gives estimates of the standard deviations of the estimated concentrations at all points in an area.

Kriging is a spatial interpolation and averaging methodology. Typically the specified interpolation points are all points on a regular grid pattern. In order to compute standard deviations it is necessary to use information about the spatial autocorrelation of data values and assume regularity in the true values of the concentrations. Regularity means that concentrations at points close together have similar values. Regularity is equivalent to stationary mean values. Regularity differs from autocorrelation in that regularity describes the true concentrations and autocorrelation measures the relationship of data values versus distance between data values. Autocorrelations are low for closely spaced data points if there is low regularity (non-stationary means) or if there are large measurement errors.

DATA ANALYSIS

The first step in the process of Kriging is to examine some typical data to determine how the correlations between data values vary with the distance between sampling sites (locations). The mathematical shape of this correlation versus distance curve is used to determine the optimum sampling pattern and interpolate between data points to estimate inventory and distribution. The interpolation algorithm, called Kriging, actually uses a curve called a variogram which is mathematically related to the correlation versus distance curve.

The data chosen for this study were ²²⁶Ra concentrations in soil of the Cottonwood district. These data had a number of characteristics that made them useful for evaluating scenarios for spatially distributed phenomena. A relatively large number of sampling points was collected for this project. The sampling points were randomly spread throughout the area and some sections of the area were more intensively sampled than other sections. Because the area is adjacent

to old mill tailings piles, it is expected that there was a spatial distribution of ^{226}Ra concentrations caused by windblown sand. Background data were available in the form of soil cores collected deep enough so that extensive disturbance by such human activities as cultivation of the soil was unlikely.

The Cottonwood tract data did not satisfy the regularity requirement of Kriging because of human activity. Activities such as backfill, cultivation, landscaping, and the use of tailings as construction material tend to cause both surface and deep soil samples to have high activity at some locations. Data from samples taken at such locations were deleted from the data since the purpose of this study is to consider surface contamination by windblown tailings. When those samples were deleted, the regularity requirement of Kriging was satisfied. It should be emphasized that the data and sampling schemes were originally intended to find contamination due to human activity in the presence of windblown contamination, thus the conclusions and recommendations reached in the following analyses are not applicable to the Edgemont Remedial Action Program engineering assessments, but are applicable to an environmental assessment.

The Cottonwood radium data were first subjected to exploratory data analysis procedures to determine their overall statistical characteristics. The surface data were found to have a lognormal statistical distribution. Thus, the remainder of the data analysis was done on the natural logarithms of the radium concentrations rather than the actual concentrations. In the log-scale, the overall mean was 1.994 (7.34 pCi/g) and the standard deviation was 0.735. There were 438 data values. The background data showed a more complicated statistical structure. It was described by a mixture of two lognormal distribution functions. Again in logarithms, 35% of the background data came from a population with a mean value of 1.136 (3.11 pCi/g) and a standard deviation of 0.380, and 65% came from a population with a mean of 0.910 (2.48 pCi/g) and a standard deviation of 0.176.

SAMPLING METHODS

Some of the concepts upon which Kriging is based can be used to examine sampling protocols in a general way (McBratney, Webster and Burgess 1981). The most efficient sampling plan is to sample on a grid of equilateral triangles. Using a square grid causes a loss of only a few percent in efficiency (i.e., the standard errors of ^{226}Ra concentration block means are a few percent larger). McBratney, Webster and Burgess (1981) gave a computer program for examining different grid spacings; this program was used with Cottonwood data to generate the coefficient of variation of the block means as a function of grid spacing (Table 1). The coefficient of variation of block means (relative error or S.D. of the logarithms/mean of the logarithms) is the appropriate measure of error when lognormal data are analyzed. The input to this program was information on spatial correlations in the Cottonwood tract in the form of a variogram.

Table 1
Cottonwood Tract Spatial Correlations

Square Grid Spacing		Number of Samples	Coefficient of Variation
Feet	Meters		
10	3.1	15,000	0.949
25	7.6	2,400	0.953
50	15.5	600	0.958
100	30.5	150	0.970
150	46	66	0.980
200	61	37	0.990
250	76	24	0.999
300	92	16	1.008

By using this program and the resulting table one can avoid the lengthy task of simulating data from some specified sampling plan a number of times, analyzing the data sets, and interpreting the results. This table shows that for situations exemplified by these data, the relative error does increase with larger grid spacing but it increases very slowly so there is not much increase in the relative error of block means due to using a large grid and fewer samples. The Cottonwood tract is about 1200 ft (366 m) on a side, and sampling every 10 ft (3.1 m) would require about 15,000 samples; sampling every 250 ft (76 m) would require 24 samples. The above table suggests that the 15,000 samples would only produce a 5% decrease in the relative error of block means.

The Holoway et al. (1981) protocol (30-m grid) would require 150 samples and gain only about 3% over a 100 meter grid spacing. They base most of their sample size and grid spacing recommendations on statistical considerations of the accuracy of estimated mean values (e.g. Equation 1). This is distinct from Kriging which is concerned with the spatial patterns within the data. (However, the McBratney, Webster and Burgess (1981) technique is also concerned with the accuracy (Kriging variance) of the Kriging mean, but they take correlation into account, whereas Holoway et al. (1981) do not, at least explicitly.)

An empirical evaluation of the detail produced by Kriging for planning clean-up activities was implemented by computing spatial patterns for different random subsets of the Edgemont data. The spatial patterns produced were then judged from the perspective of planning a cleanup program. Two kinds of situations were evaluated. First, a number of spatial patterns were produced using one-third of the data to see if the pattern changed significantly with the same number of samples but different samples with different data values. Second, a few spatial patterns were produced for samples of 1/2, 1/5, and 1/10 of the total data. These, along with the samples of 1/3 of the data, were compared to the spatial pattern estimated using all the data. The smaller sample sets did show less detail and had smoother concentration contours. However, from a perspective of planning a cleanup, there were no important differences in any of the spatial patterns produced. If the total 438 samples taken were collected on a square grid rather than randomly over the Cottonwood tract in Edgemont, a grid spacing of 60 ft (18 m) would have to be used. A tenth of the data, or 44 samples in the same area, would require a grid spacing of about 200 ft (60 m).

Examples of the calculated spatial patterns are given in Figures 5, 6 and 7. Figure 5 is a computer printer plot of Kriging of the Cottonwood tract using a 10-meter grid spacing to indicate the ease with which contour lines maybe identified on the computer output. Figure 6 is an artist's version of Figure 5 for clarity. Figure 7 was prepared using only 4% of the data (50-m grid) used for Figures 5 and 6, yet the overall trends in concentrations are seen to be similar, although the fine detail (differentiation of smaller areas) is lost. The application of this technique will be discussed in the recommended sampling protocol section.

To produce the plot depicted in Figures 5 and 6 the Cottonwood background data were first Kriged to estimate the average background concentration of radium for each 100-m² block (squares with 10 m on a side). The same Kriging was then done for the surface soil samples. The surface and background data were not all collected at the same sampling sites, but the average concentrations for each 100-m² block were estimated for the same locations. Then for each block the background was subtracted from the surface concentration, the results were tabulated and plotted. In Figure 5, 10 m horizontally is represented by 2.5 columns, and 10 m vertically is represented by 1.5 rows. A blank area has estimated concentrations less than 1.25 pCi/g above background averaged over 100-m² blocks. Ones represent areas with estimated concentrations between 1.25 and 2.5 pCi/g. Twos are areas between 2.5 and 5, 3s are areas between 5 and 10, 4s are areas between 10 and 20, and 5s represent areas between 20 and 40 pCi/g. To perform remedial action on all areas with an estimated concentration over 5 pCi/g above background, one would draw landmarks on Figures 5 or 6 (the boundaries in Figure 5 are centers of streets and in Figure 6 are property lines), to locate the areas >5 pCi/g.

The discussion and conclusions reached above are valid for the Cottonwood area, or any similar area, although a similar area would not necessarily have the same size and isotope concentrations as Edgemont. The size of the area can be scaled up or down, and as long as the relative size of a grid of sampling points is the same as for Edgemont, the conclusions reached above are valid. The concentration values are immaterial as long as the spatial correlation remains about the same; even if values averaging 2 pCi/g, as was the case for Edgemont, have the same spatial correlation structure as another site with values averaging 50 pCi/g then the conclusions above would in general hold.

The "sufficient detail" must also be scaled up or down with area size. In Edgemont, the evaluation of "sufficient detail" was made on the basis of determining which properties require remedial action. The properties varied from around 100 ft on a side up to 300 ft on a side. If the 1200 by 1250 ft of the Edgemont site were scaled up to a site 12 mi by 12 mi, then the criteria of equivalent "sufficient detail" would determine the need for remedial action on properties 1 mi on a side to 3 mi on a side.

The Cottonwood area was not sampled on a regular grid. Rather, a number of samples were taken on each property at places chosen by the field crews according to protocol. The sampling pattern appears to be random although it actually is not. An advantage of Kriging is that it can analyze data collected on any spatial pattern. When the data are not collected on a regular grid the only consequence is that some efficiency is lost. This is an important and useful attribute because it is usually difficult to accurately measure out a grid in the field and often a grid sampling point will contain an obstruction to sampling.

FIGURE 5. Computer Printout of 100-m² Block Average Radium-226 Concentrations Above Background in Surface Soil from Kriging Cottonwood Study Area Data, 10-m Grid

FIGURE 6. 100-m² Block Average Radium-226 Concentrations Above Background in Surface Soil from Kriging Cottonwood Study Area Data, 10-m Grid

FIGURE 7. 100-m^2 Block Average Radium-226 Concentrations Above Background in Surface Soil from Kriging Cottonwood Study Area Data, 50-m Grid

Data plots similar to Figures 5, 6 and 7 can be produced for total surface inventory or inventory above an average background. From the tabular data, each 100-m² block can be identified by north-south and east-west coordinates, and for each, the estimated average inventory and its standard error are given by the computer program. These averages can be multiplied by the size of the blocks and summed to yield total inventories. The associated standard errors can be used to calculate confidence intervals on the inventory estimates (see Appendix).

The EPA regulations state that any 100-m² area with an average value greater than 5 pCi/g over background must be cleaned up. These regulations do not discuss how an area is to be divided into 100-m² blocks. Mathematically, in any particular area there are an infinite number of overlapping 100-m² blocks of all shapes. One must assume that EPA will accept the use of a regular pattern of simple geometric shapes. McBratney, Webster and Burgess (1981) showed that the square grid is the most practical sampling protocol. Rendu (1979) examined a sequence of overlapping square blocks laid out over an area, and concluded that the placement of a grid work on an area does not significantly alter any results. The block estimates produced by Kriging are unbiased estimates of the average (mean) concentration in each block (estimates of the median concentration for logarithmic Kriging). If Kriging is done to get block averages with the size of the blocks 10-m square, then the results are applicable to the EPA regulations.

RECOMMENDED SAMPLING PROTOCOL

The discussion so far of the Cottonwood data analysis by Kriging indicates that the amount of detail obtained in defining concentration boundaries is roughly proportional to the number of sampling locations used. The EPA regulations, which require areas of 100 m² to be defined, suggest a large number of samples is required. A two-stage sampling plan can minimize sampling by providing accurate concentration contours only in those areas containing the boundaries of the properties requiring remedial action. Such a sampling plan is similar to the Holoway et al. (1981) concept of a preliminary survey.

The first-stage sampling uses a large size grid to identify areas where more intensive sampling should be performed. Figure 7 shows the result of Kriging on the Cottonwood data when all data except points on a 50-m square grid were deleted. Samples would be taken at the center of each 2500-m² block. In this figure the upper-left part shows contours that are above the EPA criteria of 5 pCi/g above background. We propose the following arbitrary sampling rules. These rules do not use all the information provided by sophisticated Kriging computer programs. However, they can be used by field crews without the services of a Kriging specialist. Also, these rules are appropriate for simple versions of Kriging which could be used with portable computers in the field. Any area that is shown to be above one-half the EPA standard by the first stage of sampling should be subjected to second-stage sampling. However, any area found to be above twice the EPA standards during the first-stage sampling could reasonably be assumed to be candidate for remedial action without more detailed contours. Note that the 50-m grid basically averages over 2500 m², thus such a large grid can only be used to identify trends that might indicate where 100-m² areas above EPA standards might be located.

The second-stage sampling is intended to accurately identify the 100-m² areas that require remedial action. Figures 5 and 6 are a representation of the Kriging results of the Cottonwood data using a 10-m grid for sampling. A compari-

son of Figures 6 and 7 shows that two-stage sampling would find all areas above 5 pCi/g even though the first stage did not accurately show contamination concentration contours. The three small contaminated areas in the lower right of Figure 6 are all much smaller than a 50-m grid and thus in general such areas would not be found during first-stage sampling. The first-stage sampling, on a 50-m grid, used 49 samples and indicated that 61% of the area should be intensively sampled in a second stage. Sampling 61% of the Cottonwood tract (119000 m²) on a 10-m grid would require 684 samples, for a total of 733 samples in both stages. Sampling the entire study area on a 10-m grid requires 1122 samples, thus the two-stage sampling results in a 35% savings in sampling effort (number of samples). The second stage found that 544 of the 100-m² blocks exceeded 5 pCi/g. This represents 46% of the area in the Cottonwood tract.

For visually evaluating the spatial scale of Figures 5 and 6, the small circular area in the top center that is between 20 and 40 pCi/g is 800 m² in size, the area within a baseball diamond is 752 m². There are three small areas in the lower-right quarter of these figures that are between 5 and 10 pCi/g. The lower-center area is 300 m², the center right is 1200 m², and the lower right is 600 m². Figure 7 has the same horizontal and vertical scales as Figures 5 and 6.

In review, our recommended protocol consists of three phases.

- 1) Use the general spatial-sampling principles discussed above to design an initial survey and to estimate the necessary equipment, time, and manpower required.
- 2) Analyze the initial survey data using both exploratory data analysis and Kriging to see if they can produce the desired level of accuracy and precision; if not, collect more data.
- 3) Compute the concentration contours to define the areas requiring remedial action.

KRIGING REQUIREMENTS AND ALTERNATIVES

Kriging is not a methodology that is readily available in the field. The computer programs require large computers and trained personnel are needed to set up and interpret computer runs. However, since the computer runs take only a few minutes of computer time, it is possible to use remote terminals in the field connected via telephone to a computer. Simplified versions of Kriging could be programmed into commercially-available battery-powered portable computers. Unless it is necessary to perform remedial action immediately after sampling, the use of Kriging by field crews will not in general be necessary.

Kriging is not universally accepted as the best methodology for performing spatial statistics. For example, Sanathanan et al. (1981) criticized Kriging because the authors' data after Kriging didn't give well-defined boundaries for areas requiring remedial action. Their argument ignores the real possibility that this situation resulted because the boundaries were, in fact, poorly defined. The authors further criticized Kriging for requiring the assumption of regularity and requiring the user to provide the algorithm information about spatial autocorrelation. These authors did not comment upon whether or not the assumption

of regularity is reasonable, even though most investigators believe it is. Without regularity it would be necessary to totally sample an area since the concentration at any particular point would be independent of all other points. While the authors criticized this assumption with Kriging, they in fact used it in their recommended method. The necessity of finding autocorrelations before Kriging can take time and requires considerable work, but it is the reason Kriging yields unbiased concentration estimates and provides standard deviations of those estimates.

Sanathanan et al. (1981) described and recommended a methodology based on clustering. Their method can neither estimate total inventory nor interpolate, thus the placement of remedial action boundaries between clusters of data points is subjective. Different investigators using clustering at the same site will get different boundaries. Since Kriging is not basically subjective, different investigators will get the same answer. Furthermore, clustering itself is a very subjective art. This was not mentioned in the paper. Clustering algorithms typically output a hierarchy of clusters, starting with all the data in one cluster and successively dividing the data into smaller and smaller clusters until at the final step each cluster contains one data point. This output is usually called a cluster tree because the trunk represents all the data, the limbs smaller clusters, and the leaves individual data points. With clustering the user must subjectively decide how far up this clustering tree to go to get a useful division of his data. Different users given the same data and objective will choose different levels on a cluster tree, and thus get different results.

HOT SPOTS

Kriging cannot be used for all types of spatial analysis problems. A common problem that cannot be analyzed using Kriging is the detection of hot spots. This situation has been studied by Singer (1975) and reviewed by Gilbert (1982). The sampling protocol required to find, with high (greater than 75%) probability of detection, a circular or elliptic hot spot can be summarized as follows: a regular grid of sampling points must be used with the grid spacing equal to approximately the longest dimension of the hot spot. The sampling and statistical theory of detecting hot spots is well worked out and precise. This theory is discussed in detail in the references given above.

A troublesome question is the definition of a hot spot. The statistical and sampling theory assumes that a hot spot is a well defined cylindrical or elliptical object buried close to or on the surface, such as a metal drum containing an easily detectable material that is substantially different from its environment. In radionuclide-contamination cleanup work, the term "hot spot" can assume a variety of meanings. It is difficult to obtain a consensus on the size of a hot spot and generally the size seems to vary with contamination level: small and very hot to large and slightly hot. Such a conditional definition can be used if a non-collimated survey instrument is used, but it is useless when soil samples are laboratory analyzed. An ideal definition of a hot spot is a well defined simple geometric shape with a clearly defined perimeter and a true-false type of rule for classifying such an area as hot. However, the implementation guidance in 40 CFR 192 indicates that "In most circumstances, no significant harm would be caused by not cleaning up small areas of land contaminated by tailings." This is clearly not a quantifiable definition of a hot spot. Holoway et al. (1981, Appendix VII) define hot spots in a different but equivalent way.

Clearly, a hot spot must be a rather large area if it is to be detected with high probability and with any reasonable sampling effort. The references above give rules for determining what proportions of small hot spots will be found with a large grid size if multiple hot spots exist. If a decontamination protocol could be written with the goal of finding some but not all hot spots, the theoretically required grid spacing for sampling would increase substantially in distance between samples. However, the only economically reasonable way to find isolated hot spots is to seek them out with survey instruments. Qualitatively any economically reasonable grid of soil samples cannot be expected to find small isolated hot spots.

CLEANUP CRITERIA

A statement such as "cleaning up 95% of the total inventory above background" is statistically vague. Two possible interpretations of this statement are as follows. If it means cleaning up all of the 100-m² blocks in which the lower 95% confidence limit of the median concentration includes 5 pCi/g, then 898 of the 1190 blocks (75% of them) in the Cottonwood tract would be cleaned up and the estimated inventory in these blocks is 0.17 Ci. The median rather than the mean was used because the data are lognormally distributed. (The mean can be used if an appropriate "lognormal correction factor" is included in the data analysis.) If blocks with median values of 5 pCi/g (the 1 μ Ci/m² equivalent was actually used) or greater are cleaned up, then 543 of the blocks, 46% of the total blocks, would be cleaned up and the total inventory in these blocks is 0.104 Ci with an approximate 95% confidence interval of 0.083 to 0.125 Ci. The average standard error of the medians is 0.622 μ Ci/m² (3.11 pCi/g). The confidence interval and average standard error are obtained by deriving the sum and average median as a linear combination of block medians and block variances.

Since all the Edgemont data error estimates are on concentrations in fixed areas, statements such as "clean up 95% of the contaminated area" cannot be used. To a statistician this implies that the contamination is a known quantity but the areas are measured with some error. These comments show how carefully the probability statements have to be worded in order to be useful to the statistician. Cleaning up 95% of the contaminated area is not the same as cleaning up the area with a 95% chance of being contaminated.

The EPA standards are statistically precise in defining the criteria for cleanup (5 pCi/g averaged over the first 15 cm in a 100-m² area). However, to implement this criteria with "reasonable assurance" is a totally non-informative specification. For the purposes of survey protocol, reasonable assurance must be replaced with three specifications:

- 1) What is the allowable chance of missing an area that should be cleaned up?
- 2) What is the allowable risk for incurring the expense of cleaning up an area that does not have to be cleaned?
- 3) How accurately must the mean values be known?

These three specifications are usually stated in a more rigorous form in the statistics literature:

- 1) The maximum permissible probability of declaring that an area meets the criteria when in fact it does not.
- 2) The minimum permissible probability of declaring that an area does not meet the criteria when in fact it does.
- 3) The maximum permissible value of the standard error of the mean of the area.

Typically one's intuitive quantification of "reasonable assurance" in terms of these three specifications is to make the probability of missing an area that should be cleaned up very low (e.g. 1%), the probability of wasting remedial effort on a clean area not quite as low (e.g. 5%), and requiring that the estimated mean be within 10 to 15 percent of its true value. In the actual soil sampling situations with which we have so far been associated, such specifications of reasonable assurance would result in the need for thousands of samples. Thus, the number of samples collected and the sampling protocol are invariably limited by available resources. Typical available resource sampling results in around a 50% chance of missing an area that should be cleaned up, a 50% chance of performing unnecessary cleanup, and a relative error of the mean around 100%. The biggest contribution to these disappointing specifications seems to be small scale spatial variability; concentrations at points just a few tens of centimeters apart can easily differ by a factor of 2.

This type of situation usually leads one to try to find a better statistical technique than Kriging. However, Kriging has been theoretically shown to be a statistically optimal procedure (Matheron 1971). The 50% probability of making an incorrect decision is a consequence of having data with a 100% relative error. In other types of spatial analysis, such as estimation of some types of ore-body locations and inventories, relative errors of 10% have been achieved and then the probabilities of an incorrect decision are low. This indicates that the most productive route for spatial analysis of radioisotope research is to perform research to investigate why spatial correlations are very low. Once the mechanism causing low spatial correlations is understood, steps can be taken to compensate for such phenomena.

IDENTIFICATION OF RESIDUAL RADIOACTIVE MATERIAL

The Uranium Mill Tailings Radiation Control Act of 1978 very carefully defines residual radioactive material and uranium byproduct material to differentiate between them and also between them and native deposits of materials naturally enriched in uranium decay-chain products. It is therefore essential, before remedial actions are performed, that the classification of the material in question be determined. "Residual radioactive material" (loosely paraphrased) means waste which the Secretary of Energy determines to be radioactive in the form of tailings which resulted from the processing of ore for the extraction of uranium and other valuable constituents of the ores, and other wastes which relate to the same extraction processes including any residual stock of unprocessed ores or low-grade materials. "Byproduct material" means any radioactive material, except special nuclear material, resulting from its exposure to the radiation from the process of producing or utilizing special nuclear material and also tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content.

Thus it is important to not only know the isotopic composition, but one must also ascertain the source of the radioactive material to properly categorize it. Residual radioactive material relates only to the designated inactive processing sites, and so no confusion should be likely between it and byproduct material which would generally be associated with active sites.

Differentiation between residual radioactive material (displaced tailings and ore) and natural deposits of materials containing enhanced uranium decay chain products requires more information. If soil samples are analyzed for ^{226}Ra content using an intrinsic germanium detector (Perkins and Thomas 1978; Young, Jackson and Thomas 1983; others), the uranium-daughter concentrations and therefore the daughter-to-uranium equilibrium ratio for each sample can be estimated. Comparison of this ratio to that of surrounding native soils will give important information to help classify the material. If the soil sample contains similar activities of uranium daughters ^{234}Th , ^{230}Th , ^{226}Ra , and ^{210}Pb , the soil activity is more likely due to natural uranium. Material containing uranium mill tailings on the other hand should have much less ^{234}Th activity than those of ^{230}Th , ^{226}Ra , and ^{210}Pb . Further guidance is given for interpretation of equilibrium ratios by Young, Jackson and Thomas (1983).

Differentiation between natural uranium-bearing deposits and displaced ore is best made by visual examination of the radioactive material in relation to the surrounding native geological structures and soil matrix. Many ores have characteristic physical features and colors which can be an aid to a trained observer. In any case judgment will have to be exercised to decide whether a deposit of uranium ore is natural or displaced.

CONCLUSIONS

Although there is an abundance of literature describing, recommending, and reporting survey techniques and results from radiological surveys and sampling for a variety of purposes, the literature describing sampling and surveys contains very little material that can be used to statistically establish the geographical or spatial locations to be used in a sampling plan for the measurement of residual radioactivity around uranium mill tailings piles.

Gamma-ray measurement techniques have been used and are still recommended by some investigators to estimate cleanup required near tailings piles. However, the radiation "shine" from a tailings pile contributes to the gamma measurements being made near the pile, thereby requiring special techniques or corrections to be applied to the measurement process or data collected. Promising innovations have been proposed by some investigators and are now being evaluated. Probably the most ambitious protocol is being pursued by TMC in Grand Junction, Colorado, and will soon be published.

Results of a PNL field test of a modified micro-R-meter performed as part of the Edgemont Remedial Action Program were reported. Linear regression analysis was applied to comparison measurements made using the modified micro-R-meter in situ and laboratory gamma analysis of 15-cm core samples of soil for ^{226}Ra concentrations. The low coefficients of determination obtained near tailings piles indicated that the modified (collimated) micro-R-meter readings may be used to evaluate the presence or absence of contamination above the average background for properties in the vicinity of tailings piles; but for those within approximately a block (300 to 500 ft) of the storage areas, another technique would be more appropriate. However, should the gamma measurement techniques being developed by TMC for use near tailings piles prove to be technically and economically attractive, the use of gamma-ray measurements to supplant most soil sampling followed by application of Kriging techniques to the gamma-ray measurements would appear to be an optimal survey procedure.

In the absence of proven available gamma measurement techniques for detection and measurement of residual radioactivity near tailings piles to meet the requirements of 40 CFR 192, this study concentrated on the statistical analysis of soil sampling data collected near tailings piles in the Cottonwood district of Edgemont, South Dakota. These data were readily available, having been collected by PNL over the past two years in support of the Edgemont Remedial Action Program. Kriging was the statistical technique key to this analysis.

Kriging is a theory of spatial interpolation and spatial statistical analysis. Although it is more difficult to use than alternatively proposed methods, it offers many theoretical and practical advantages for the analysis of radioisotope data collected with a spatial component, such as soil samples. Several methods and algorithms derived from this theory are useful in the design and analysis of soil sampling projects. These were illustrated using data from the Cottonwood district. The result of these analyses is an assessment of the inventory and distribution of radionuclides in the area. Kriging can be made available to investigators in the field through telephone links between portable computers or terminals and a large computer, but a specialist is required for the efficient use of such capability.

The conclusion reached concerning spatial sampling design work is that for situations like that at Cottonwood, the accuracy of the spatial pattern derived by Kriging is only slightly affected by the number of samples used, and thus a small number of samples taken on a widely spaced grid is the most efficient sampling plan. There is the possibility that a large-grid (i.e. widely spaced sample sites) sampling plan alone will not give "sufficient detail" for planning cleanup activities. A second stage of sampling using a smaller grid spacing on areas selected from the results of the large-grid sampling can be used to provide this detail. There are no available algorithms for answering questions about "sufficient detail," so the results of an empirical evaluation were reported.

Not all statistical topics related to spatial data can be analyzed using Kriging theory. Methods for the detection of hot spots are one such topic that was discussed. Another is the unambiguous specification of the statistical criteria upon which cleanup decision rules are based. The term "reasonable assurance" must be replaced by three statistical criteria in order to eliminate ambiguity. These three criteria are: 1) a statement defining the allowable probability of missing an area that should be cleaned up, 2) a statement defining the allowable probability of incurring the expense of cleaning up an area that does not have to be cleaned, and 3) a statement defining the required accuracy and precision of the data.

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APPENDIX

STATISTICAL METHODOLOGY

The theoretical basis of Kriging is well documented in the literature and textbooks of geology. A monograph by Rendu (1978) is entirely devoted to Kriging. Two recent texts that have major sections devoted to Kriging are those by David (1977) and by Journel and Huijbregts (1978). The computer program used to perform Kriging on the Cottonwood data was BLUEPACK, a proprietary spatial analysis computer package obtained from Ecole Nationale Supérieure Mines de Paris (French National School of Mines).

The methodology and computer programs used in the discussion of survey design are given in the three-part paper by McBratney, Webster and Burgess (1981). The analysis of the hot-spot problem is based upon the work by Singer (1975) as developed by Gilbert (1982). The remaining sections, such as the specification of criteria, are based upon general graduate-level statistics.

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