OAK RIDGE BACKGROUND SOILS PROJECT PROVIDES DATA FOR REMEDIAL ACTION EVALUATION

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ABSTRACT

Many constituents of potential concern for human health occur naturally at low concentrations in soils. The primary objective of the Background Soil Characterization Project (BSCP) was to provide fully validated and defensible, reservation-wide background concentration data on significant potential contaminants of concern (organics, inorganics, and radionuclides) in natural soils on the Oak Ridge Reservation (ORR). The data are particularly significant for remedial action projects; the data can be used to establish technical guidance and the basis for realistic cleanup requirements at hazardous waste sites on the reservation. Other objectives included providing baseline data for conducting contaminated site assessments and quantifying estimates of human health risk associated with background levels of potentially hazardous constituents in soils.

All results were required to adhere to the most rigorous Environmental Protection Agency (EPA) protocols and requirements (EPA Level IV) for analytical procedures, quality control, data validation, and data record documentation. Data validation required the use of existing validation criteria and procedures from the EPA Contract Laboratory Program (CLP) for chemical data and development of criteria and new procedures for non-CLP chemical and radiological data. These procedures provided data that are both technically and legally defensible. All analytical laboratory results were fully validated and peer reviewed, and were verified as being representative of and corresponding to the formations of interest. Statistical analysis was used to establish data validity in meeting project objectives and provided the summary statistical parameters necessary for application of the data to subsequent assessment of risk. Field data were integrated with analytical data to provide overall technical interpretation to determine the meaning and implications of the results.

The main conclusions drawn from analysis and interpretation of the data were (1) there was general consistency in background concentrations of soil constituents of interest and in the levels of risk associated with background soil concentrations among sampling sites on the ORR and in two off-site areas, (2) all results reported in the project were confirmed to be representative of background concentrations, and (3) natural background risk levels calculated for the constituents of interest were above the EPA's conventional range of acceptable risk but well below generally accepted levels of lifetime risk from background radiation across the United States.

INTRODUCTION

The BSCP was undertaken to provide the necessary data to make informed decisions based on extensive characterization of natural background soil concentrations of organics, inorganics, and radionuclides found on the ORR. Standards and cleanup criteria for contaminated soils on the ORR will be based on concentrations above those established as background in this project for typical soil constituents. These data will also provide the basis for discussion and negotiation with regulators. The availability of rigorous and defensible background soils data will expedite the decision-making process and enhance public acceptance of proposed remedial actions.

To evaluate realistically the level of contamination (with implications for risk and remedial actions), it is necessary to know with confidence the background levels of contaminants from fully validated and defensible baseline data that would be expected at a specific site. To understand the geologic soil environment, the BSCP addressed variability of concentration levels in terms of 1) taxonomical types (soil series) occurring in different geologic formations, 2) soil sampling depths (horizons) within a specific soil profile, and 3) natural areal variations in soils both on-site and off-site developed from the same geologic formation.

The ORR is located in East Tennessee and lies in an area characterized by elongated ridges and valleys that run northeast to southwest. The hydrologic system on the ORR, including both surface water and groundwater, is dominated regionally by the Clinch River. The climate of the area is generally temperate with warm, humid summers and cool winters, and the average annual rainfall in the Oak Ridge area is approximately 136 cm.

Geologically, the areas sampled contain three principal rock groups that include formations in the Conasauga (shale), Knox (dolomite), and Chickamauga (limestone/carbonate) groups. There are two major categories of soils: residual soils

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developed from in-place weathering of the geologic groups and soils developed in partially sorted colluvial and alluvial soil materials. Only the major formations within residual soil groups were considered in this investigation, because they represent the dominant soils at imminent remedial action projects on the ORR. These formations were the Dismal Gap and Nolichucky of the Conasauga Group, the Copper Ridge and Chepultepec of the Knox Group, and undefined formations of the Chickamauga Group in two different geographic areas (1, Vol. 1) (references appear in brackets).

Extensive site screening was performed to ensure that true background samples would be obtained in this project. The screening consisted of using a hand-held radiation detector over each sampling location, gamma scanning for cesium-137 fallout on surface soil samples, and laboratory analysis for volatile organic compounds (VOCs) to detect any evidence of gross contamination. In this project background was defined as concentrations of soil constituents that showed no evidence of contamination from activities at facilities on the ORR. Screening was done after preliminary examination and evaluation of each prospective sampling site with respect to physical and geologic factors, including extent of vegetative cover and evidence of land use practices, site accessibility, local soil erosion, and any other potential outside influences (1, Vol. 3). Finally, two off-site sampling areas (see Fig. 1) were designated to obtain corroborative data for comparison with ORR data.

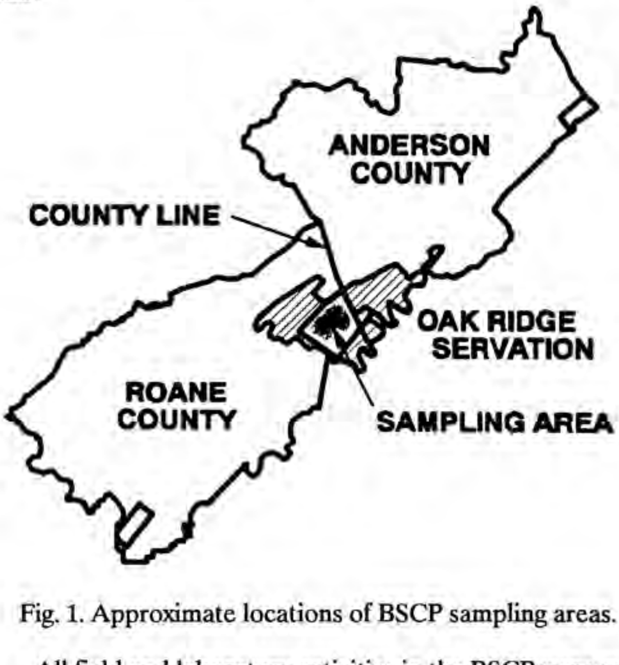

Fig. 1. Approximate locations of BSCP sampling areas.

All field and laboratory activities in the BSCP were consistent with the data quality objectives (DQOs) established as part of the project planning process. The BSCP Plan (1, Vol. 3) specified the full range of project requirements and assumptions and included field crew training requirements, quality assurance audits and surveillances, and data management. In addition, the basic requirements of precision, accuracy, representativeness, completeness, and compatibility for integrating and evaluating analytical and field data were met. These requirements were developed as a result of applying the DQO process in structuring the project.

OBJECTIVES AND APPROACH

The objectives of the BSCP were to:

- determine background concentrations in undisturbed soils of organics, inorganics (metals), and radionuclides that are key to environmental restoration projects on the ORR;
- provide fully validated and defensible, reservation-wide baseline data for contaminated site assessment;
- provide the basis for establishing remedial action cleanup levels; and
- quantify baseline potential health risk from background constituents for comparison and evaluation of risks associated with contaminated sites.

The approach detailed in the BSCP Plan (1, Vol. 3) is summarized as follows:

- identification of the most important geologic formations underlying potentially contaminated sites on the ORR;
- identification of the dominant residuum soil type corresponding to each selected formation;
- randomized selection of candidate soil sampling sites on the ORR, and in two remote areas in western Roane County and in eastern Anderson County;
- basing of analyte lists on site history, risk assessment requirements, and EPA target analytes;
- field screening for site acceptability followed by soil sampling;
- chemical and radiological analyses by commercial analytical laboratories;
- full (100%) data validation, verification, statistical analysis, and interpretation; and
- transfer of data to the Oak Ridge Environmental Information System (OREIS) for availability to users.

SAMPLING SITE SELECTION

ORR sampling sites were confined mostly to the Roane County portion of the ORR, but some ORR Bethel Valley Chickamauga sites were located in Anderson County (Fig. 1). Recent digitized soil maps (available from OREIS), where residual soils had been related to the underlying geologic formations, provided the base map for selecting most potential ORR sites. A total of 120 sites (with three soil horizons) was required. The statistical sampling program developed for this project was used to randomly select grid coordinates that fell on predetermined soil map delineations of the most commonly occurring soils with the stipulation that no two sites be less than 250 ft apart. This methodology produced a working base map containing a unique number of sites in each sampling area for Dismal Gap, Nolichucky, Copper Ridge, Chepultepec, and Chickamauga soils.

In addition, the statistical program determined primary and secondary sampling sites. Secondary sites were alternate site locations in case the primary sites were unacceptable in terms of the selection criteria. In several cases on the ORR, however, both primary and secondary sites were unacceptable, resulting in the soil scientist evaluating nearby potential sites that would meet the criteria. The majority of ORR Chickamauga sites were selected by the soil scientist in this way because of extreme soil variability. Potential sampling sites in southwestern Roane County and northeastern Anderson County (Fig. 1) were selected somewhat differently because of land ownership, vegetation cover, and past land use disturbance constraints. Anderson County and Roane County designated sites were located within the shaded off-site areas, shown in Fig. 1. More than 48 potential sites were located in these off-site areas. Those remote sites chosen were located throughout the areas evaluated and had to meet the same site accessibility, vegetation cover, and land disturbance requirements of the site selection criteria (1, Vol. 3) as ORR sites.

ANALYTICAL PROGRAM

The objective of the analytical program was to determine the background concentration levels of selected organics, inorganics, and radionuclides in soil samples by applying consistent analytical methods and quality control requirements.

The assumptions used to select the analytical parameters follow.

- Background concentrations of naturally occurring organic, inorganic, and radiological parameters or analytes of interest were those normally found in soils and sediments of natural origin. Contamination was indicated when levels were found to be above natural background. These included organic compounds, heavy metals, and radionuclides used in or generated by industrial and agricultural activities in the area or research activities within the ORR.
- Parameters or analytes not occurring naturally were assumed to have an a priori concentration equivalent to zero background, which would be below the analytical detection limits. Some of these included man-made compounds, such as volatile organics and some semivolatile organics. Radionuclides were considered an exception due to nuclear activation and fission products that may have been added to background by natural processes, such as atmospheric deposition, from external sources.

The analytical methods and data documentation requirements used in this project are those consistent with EPA analytical Level IV (1, Vol. 3). EPA CLP methods and procedures were used where appropriate and SW-846 methods, as well as HASL 300 methods, were used for non-CLP parameters. Due to the nature of the project, the contract-required detection limits were determined to be too high, so the analytical laboratory adapted detection limits to adequately address background levels.

Analytical data quality control was maintained by analyzing 1) laboratory blanks to assess contamination levels in the analytical process; 2) laboratory control samples to assess analytical method bias, precision, and comparability; 3) matrix spikes to assess bias of the method for the matrix, as well as precision of the method when performed in duplicate; and 4) duplicates to assess precision of the sampling process and/or the analytical methods.

DATA VALIDATION

Data validation was an important component of this project. The BSCP's requirement for 100 percent fully validated and defensible analytical laboratory data (EPA analytical Level IV) necessitated development of select project-specific validation guidelines and criteria where existing EPA CLP data validation guidelines were not available (1, Vol. 3). Project data included organics [pesticides, polychlorinated biphenyls (PCBs), herbicides, and polynuclear aromatic hydrocarbons (PAHs)]; inorganics (metals and cyanide/sulfate); and radionuclides, both naturally occurring and fallout components, determined from the EPA target analyte list and target compound list and from site program risk assessment considerations. Analytical methods included a wide range of EPA CLP methods and non-CLP methods, as well as the neutron activation analysis (NAA) and inductively coupled plasma/mass spectroscopy (ICP/MS) methods.

Available CLP validation criteria were used for pesticides, PCBs, metals, and cyanide consistent with EPA analytical Level IV where applicable, but new criteria and procedures had to be developed to validate herbicide, PAH, sulfate, radiochemical, NAA, and ICP/MS results. The objective was to produce a consistent set of validation criteria and procedures.

The BSCP's overall data validation process was conducted in three distinct steps. The first step consisted of technical validation of data according to specified criteria and laboratory action items issued to address specific validation concerns. The second step was a peer review of the previously validated data packages and included checking flagged data, assessing the rationale for professional judgments, and verifying reasonableness with respect to project data quality objectives. The third step consisted of the validation oversight function and included assessing the overall rationale of the approach and the reasonableness of the validation of each data package as a whole. After the initial phase, this three-step validation process was conducted by three independent groups, which afforded an exceptionally high level of reliability, comprehensiveness, and consistency in the data of this project.

In addition, computerized data validation procedures were developed to permit validation to be done in near real time 1) to allow for quicker response times and interaction with the analytical laboratories so that changes and adjustments in procedures and data records could be made in pending analyses and data packages, and 2) to shorten the time required for producing project data reports.

STATISTICAL ANALYSIS

Statistical analysis established data validity in meeting project objectives by providing summary statistics for analytes, error analysis, and comparison data for data interpretation and assessment of background risk, and included detection frequencies, median estimates as measures of central tendency, upper 95th quantile estimates as measures of the upper ends of the normal background range, and confidence bounds for these estimates. Detection probability confidence bounds were also given for "nondetects." The statistical methodology for "detect" data assumed that data follow the lognormal distribution, with possibly different means but with the same variance for each formation–sampling site combination. The statistical methodology incorporated each nondetect as "between zero and the detection limit" (using the lognormal model) without resorting to approximation, such as setting its value to the detection limit.

Comparisons were made across individual sampling areas and among soil horizons, and some significant differences were determined among the ORR and Anderson County and Roane County sampling sites. Laboratory and spatial area variances were also estimated and compared. All data were examined graphically to assess the lognormal assumption and to check for outliers. With the exception of a few outliers, the data appeared consistent throughout.

RESULTS

Analytical results were evaluated and qualified during the laboratory review process and the independent data validation process. The vast majority of the data proved usable. Relatively few analytes produced lower utilization rates. Among the organic results obtained, PAH data were 75 percent usable. Among radionuclides, 70 percent of the neptunium-237 and only 43 percent of the curium-244 data were usable. All other data met project data quality requirements. Typical results obtained for radionuclides and inorganics presented in Tables I and II show the degree of consistency in the data from the two formations sampled in each of the three principal soil sampling areas. Data exhibit similar background concentrations both within the same formation and across all formations on the ORR and between the reservation and off-site sampling areas. Full data have been reported (1, Vol. 2).

DATA INTERPRETATION

The objective of data interpretation was to confirm that true background data were indeed obtained. There are several factors that can influence the effectiveness of data interpretation efforts in this area. Organics, inorganics, and radionuclides in soils can have several sources. Sources are an important consideration in interpreting data derived from geologic media. Also, current extraction procedures that are part of EPA-mandated analytical methods remove differing amounts of various soil constituents and can possibly bias the results. The location of the soil in the landscape can also affect the data. Interpretation of soils data, therefore, must be done systematically and very carefully to ensure validity and consistency.

Screening sampling sites on the ORR using a radiation detector, gamma scanning, and laboratory analysis of VOCs did not reveal any gross contamination. Any VOC screening data detects at prospective sampling sites were taken as a sign of contamination and warranted prompt rejection of the site for actual soil sampling. Most of the ORR sites, along with Roane County and Anderson County sites, had some detects of organic contaminants, particularly PAHs. All data in this project were obtained under these restrictions to ensure that they represented background values.

Some organics (PAHs) were distributed either uniformly across most sampling areas on the ORR or randomly throughout all the sampling sites. The presence of PAHs, then, was considered as background for purposes of data comparison with contaminated sites. Some PAHs may have a soil origin, but most PAHs were probably from several local industrial sites.

Inorganic compounds in soils are much more difficult to interpret. Some inorganics are definitely inherited from the underlying geologic formations. Others have both an anthropogenic source (either from global fallout or from local and regional sources) and a geologic source. For example, lead and arsenic can have both geologic and anthropogenic sources, while nearly all mercury can be considered a surface anthropogenic contaminant because of elevated levels in the A soil horizon, which is the upper soil level. Several metallic elements, including cadmium, osmium, and silver, were not detected in any BSCP soil samples. The presence of any of these in the A horizon at higher levels than in the B or C soil horizon beneath could be considered an indicator of possible contamination. Some small amounts of these elements may be derived from the underlying rock.

Concentrations of inorganic compounds or metals that are higher in the A horizon than in the B or C soil horizons or in the on-site than in the off-site areas would be an indicator of anthropogenic contamination of the ORR soils and false background levels. An exception to this statement may be biocycled elements that are used by plants and become concentrated in the A horizon. In general, anthropogenic metals (heavy metals) were not detected in the A horizon at levels significantly higher than in the B or C horizon, or their levels were not significantly different than in off-site areas.

Selected metals were analyzed by the current EPA-mandated atomic absorption (AA) spectroscopy method and ICP method and compared with ICP/MS. The ICP/MS analytical method exhibited lower instrumental detection limits than the AA and ICP sweep (AA/ICP). Correlations between ICP/MS and AA/ICP were fairly consistent for all metals except thallium and selenium, suggesting that analytical methods did not bias the results, as long as the soil samples were extracted by the same procedure.

The NAA method is a whole soil analytical technique in contrast to the AA/ICP and ICP/MS analytical techniques, where an acid extract of the soil is analyzed. The levels measured by AA/ICP methods were not different from those by NAA when the compounds were acid soluble or were on soil clay or organic matter exchange sites. NAA results were higher than AA/ICP when the elements were part of the soil mineral or were more resistant to dissolution.

Many radionuclides have two primary sources: the underlying geology plus both global and regional anthropogenic (fallout) sources. However, a third possible source of certain radionuclides (curium-247, tritium, technetium-99, cesium-137, and strontium-90) from local research-oriented activities on the ORR cannot be ignored. The presence of these isotopes above background on the ORR can be interpreted as a sign of local contamination. Uranium isotopes can have a local source as well as a geologic source. Some important radionuclides, such as thorium isotopes and potassium-40, have an entirely geologic source. Concentrations of these local source radionuclides above background levels should be taken as indications of potential contamination from local sources.

Several trace and rare earth elements that were analyzed are not evaluated typically in risk assessment, but they can be important in geochemistry investigations and in tracing sediments to their source geologic formation. Cerium, europium, and terbium had higher concentrations in the Chickamauga Group than in soils from the Conasauga or Knox Group. Several trace elements were highly depleted in the Knox Group. These included hafnium, lanthanum, lutetium, and scandium. Titanium and ytterbium were fairly evenly distributed across all geologic groups.

BACKGROUND RISK EVALUATION

Background soil data, collected from A horizon soils of the Dismal Gap, Nolichucky, Copper Ridge, Chepultepec, and Chickamauga formations on the ORR and from Anderson and Roane counties, were evaluated in terms of potential adverse effects to human health. This background risk evaluation provided the context for the discussion and comparison of risks associated with site-related contamination and for determining *contaminants of potential concern* for a suspected or contaminated site.

Three primary pathways of exposure were evaluated for organic, inorganic, and radionuclide analytes, which included 1) direct ingestion of soil, 2) dermal contact with soil, and 3) external exposure to radionuclides in the soil. Background risks for individual analytes, total pathway risk estimates (i.e., the sum of the background risks of all analytes within a pathway), and cumulative risk estimates (i.e., the sum of the total pathway risks) were determined.

Constituents detected in uncontaminated background soil samples were evaluated within the context of the EPA-approved guidelines for contaminated soils in which there are three regions of carcinogenic risk (risk $< 1.0 \times 10^{-6}$, no concern; risk between 1.0×10^{-6} and 1.0×10^{-4}, range of concern; and risk $> 1.0 \times 10^{-4}$, unacceptable risk) and two areas of systemic toxicity (hazard index < 1.0, no concern; and hazard index > 1.0, concern). The background risks were reported in this manner, but the results were intended only for comparison with risks determined for contaminated sites; these results themselves do not pertain to remediation decisions.

Carcinogenic risk and noncarcinogenic hazard indices determined for individual analytes (found in the A soil horizon) were similar for the three sampling areas (ORR, Anderson and Roane counties). The cumulative pathway background risks (i.e., risks from ingestion of soil plus risks from dermal contact with soil plus risks from external exposure to radionuclides in the soil) were determined for the formations sampled and ranged from 3.1×10^{-4} to 1.2×10^{-3}. Results are presented in Fig. 2, which shows the cumulative risk resulting from using the upper and lower 95 percent confidence bounds on the median concentration (UCB95 and LCB95). The main contributors to risk for the ingestion pathway were beryllium and several PAHs; contributors to the cumulative risk from the dermal contact pathway were relatively small. Cesium-137, potassium-40, radium-226, and thorium-228 were the main contributors to risk for the external exposure pathway.

Total pathway hazard indices were determined also for ingestion of soil and dermal exposure to Dismal Gap Formation soil and Copper Ridge Formation soil and ranged from 0.06 to 1.7. Arsenic and manganese were the major contributors to the hazard indices for the ingestion pathway, and for the dermal exposure pathway they were arsenic, chromium(VI), manganese, and vanadium.

These background risk estimates should be considered only in the context of comparison with site-related risk. The EPA conventional action level for remediation, 1.0×10^{-4}, refers only to *risks related to hazardous waste sites*. Background risk results themselves are not indicative of concerns or actions that would be identified with similar potential risks from a contaminated site, and care should be taken not to misinterpret these results as pertaining directly to remediation decisions.

CONCLUSIONS

The BSCP provided the needed comprehensive sitewide, fully validated, and consistent background soil data base for contaminated site assessment, for establishing remedial action cleanup levels and design criteria, and for use in evaluating waste site treatability studies, as well as providing needed baseline estimates of human health risk associated

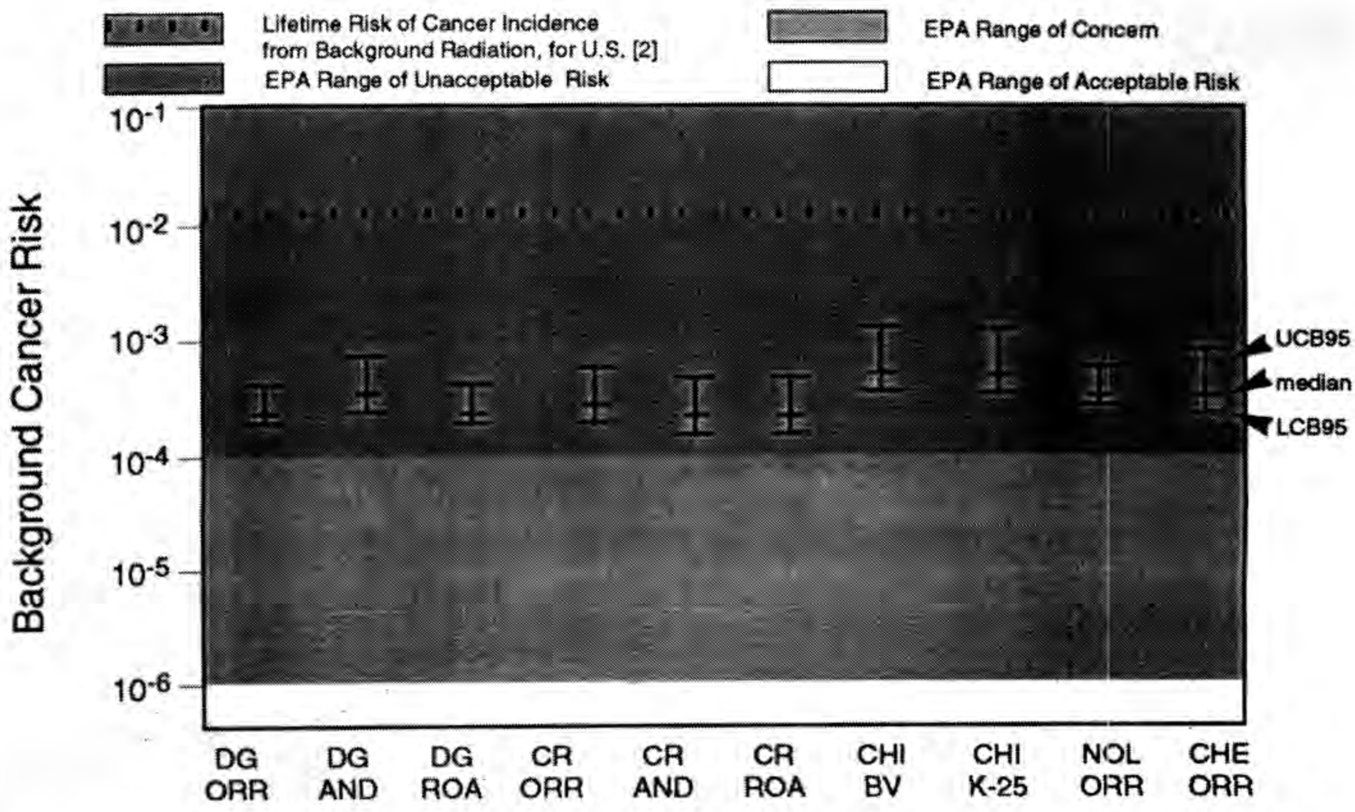

Fig. 2. Comparison of total background cancer risks calculated from soil samples from the Dismal Gap (DG), Copper Ridge (CR), Chickamauga (CHI), Nolichucky (NOL), and Chepultepec (CHE) formations in Anderson County, Roane County, and on the ORR.

with background concentration levels of potential hazardous substances and other constituents for comparison with contaminated sites on the ORR.

In summary, none of the ORR sites exhibited any indication of disturbance in approximately the past 50 years. For this reason, the data can be considered as background and used as a basis of comparison with similar areas on the ORR where contamination is known or suspected. Estimated risk levels were similar across all formations and very similar within the same formation on the ORR and between the ORR and off-site sampling areas; calculated risk estimates were above the EPA's conventional range of acceptable risk but well below the U.S. average continental background radiation level of 1.4×10^{-2} (2).

Analytical laboratory methods and data validation procedures developed and used met the project requirements specified in the BSCP Plan (1, Vol. 3). Many of the laboratory data concerns that surfaced during the early validation effort could have been averted by conducting a project-specific preaudit before the project began with reference to the BSCP Plan and the detailed method-specific contract laboratory statement of work including critical review of laboratory operating procedures and quality assurance/control requirements. Also, requiring processing of preliminary performance evaluation samples through the laboratory for each of the specific analytical methods requested would have revealed the laboratory's ability to perform the required analyses and indicated the type of data package that each laboratory could deliver before actual field samples were sent to the laboratory for analysis.

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