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THE LASL APPROACH TO URANIUM GEOCHEMICAL RECONNAISSANCE

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ABSTRACT

The US ERDA, as part of the NURE program, has initiated a nationwide Hydrogeochemical and Stream Sediment Reconnaissance (HSSR). The aims of the NURE program are to provide data on which to base more accurate estimates of US uranium reserves for long-range planning and to aid in meeting the nation's projected uranium demands into the next century. The HSSR objective is to complete, by 1980, a reconnaissance of the nation's surface waters, ground waters, and stream and lake sediments, to aid in assessment of uranium reserves and identification of areas of interest for exploration. Responsibility for carrying out the HSSR across the nation is divided among four ERDA Laboratories: Lawrence Livermore on the west, the Savannah River Laboratory on the east, the Oak Ridge Gaseous Diffusion Plant in the central states, and the LASL in the Rocky Mountain states of New Mexico, Colorado, Wyoming, and Montana and the state of Alaska. To be most effective in meeting its objective, data from the HSSR must be acquired and reported in a timely fashion, evaluated in conjunction with all pertinent geological, geochemical, and geophysical information available, and put to use with all methods of uranium resource assessment and exploration.

Based on extensive review of world literature on the subject, and results of pilot work, the LASL has concluded that, in keeping with the existing time and funding constraints, the most effective geochemical reconnaissance for uranium over the region for which it is responsible should follow strict, standardized sampling procedures and concentrate essentially on determination of the uranium levels in both natural waters and waterborne sediments wherever possible. Also, it is considered necessary that at least one well-chosen water sample and/or sediment sample be obtained from each nominal 10 km^2, except in localities (such as lake areas of Alaska) where abundant water in contact with organic sediment should make one sample location per 20 km^2 or so suffice. This indicates that sampling ~ 240 000 locations will be required over the ~ 2.7 million km^2 land area assigned the LASL.

Patterned after extensive uranium reconnaissance done in many other countries, the LASL project is comprised of the following five components: (1) Organization and planning, which includes management, design, and execution; (2) Field sampling, which includes orientation studies, generation of specifications, and contracting and inspection of field work; (3) Sample receiving and analysis, which includes development of methods and hardware, quality assurance, and archival storage; (4) Data handling and presentation, including verification, storage, output, and plotting; and (5) Data evaluation and publication, which incorporates geochemical, geological, statistical, and empirical evaluation and report writing. The LASL approach to each component and the current status in each state are described.

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I. INTRODUCTION

The US Energy Research and Development Administration (ERDA), as part of the National Uranium Resource Evaluation (NURE) program, has initiated a nationwide Hydrogeochemical and Stream Sediment Reconnaissance (HSSR) for uranium.[1] Over the past 20 years, such work has been sponsored by the governments of many countries around the world, both in the East and the West.[2-20] The ultimate aims of the NURE program are to provide the Federal Government with data on which to base more accurate estimates of US uranium reserves and to aid private industry in meeting the nation's projected uranium demands into the next century. The objective of the uranium HSSR is to complete, by late 1980, a reconnaissance of the nation's surface waters, ground waters, and stream and lake sediments, to aid in the assessment of uranium reserves and the identification of areas of interest for uranium exploration. Except for Hawaii, all states of the US are to be covered.[1]

Responsibility for carrying out the HSSR for uranium across the nation has been divided among four ERDA Laboratories (Fig. 1). The Los Alamos Scientific Laboratory (LASL) of the University of California, located at Los Alamos, NM, is responsible for the coverage throughout the Rocky Mountain states of New Mexico, Colorado, Wyoming, and Montana, which form a tier stretching northward from Mexico to Canada, and for the state of Alaska, which is larger by itself than the other four combined.[21] The LASL approach to the HSSR for uranium is the subject of this paper.

The HSSR throughout the remainder of the continental US is the responsibility of:

(1) The Lawrence Livermore Laboratory (LLL), also of the University of California, located at Livermore, CA. The LLL has the seven conterminous states west of those to be covered by the LASL.[1]

(2) The Oak Ridge Gaseous Diffusion Plant (ORGDP), operated by the Union Carbide Corporation, and located at Oak Ridge, TN. The ORGDP has the responsibility for twelve central and northeast-central states located next eastward of the four-state tier to be covered by the LASL.[1]

(3) The Savannah River Laboratory (SRL), operated by E. I. du Pont de Nemours and Company, and located at Aiken, SC. The SRL is responsible for a twenty-five state region stretching along the eastern seaboard and Gulf Coast from Maine to Louisiana.[1]

To be most effective in meeting its objective, data from the HSSR for uranium must be acquired and reported in a timely fashion, evaluated in conjunction with all pertinent geological, geochemical, and geophysical information available or becoming available from related projects (such as the aerial radiometric reconnaissance, other NURE projects, and the USGS uranium-thorium research),[22-26] and put to use with all of the various techniques of uranium resource assessment and methods of exploration.[27,28]

II. AREA OF RESPONSIBILITY AND APPROACH TAKEN

The LASL first obtained funding for the uranium HSSR in April 1975, at which time the project design work began.[21] As shown in Table I, the total area of the five states assigned to the LASL is approximately 2.7 million km^2, or

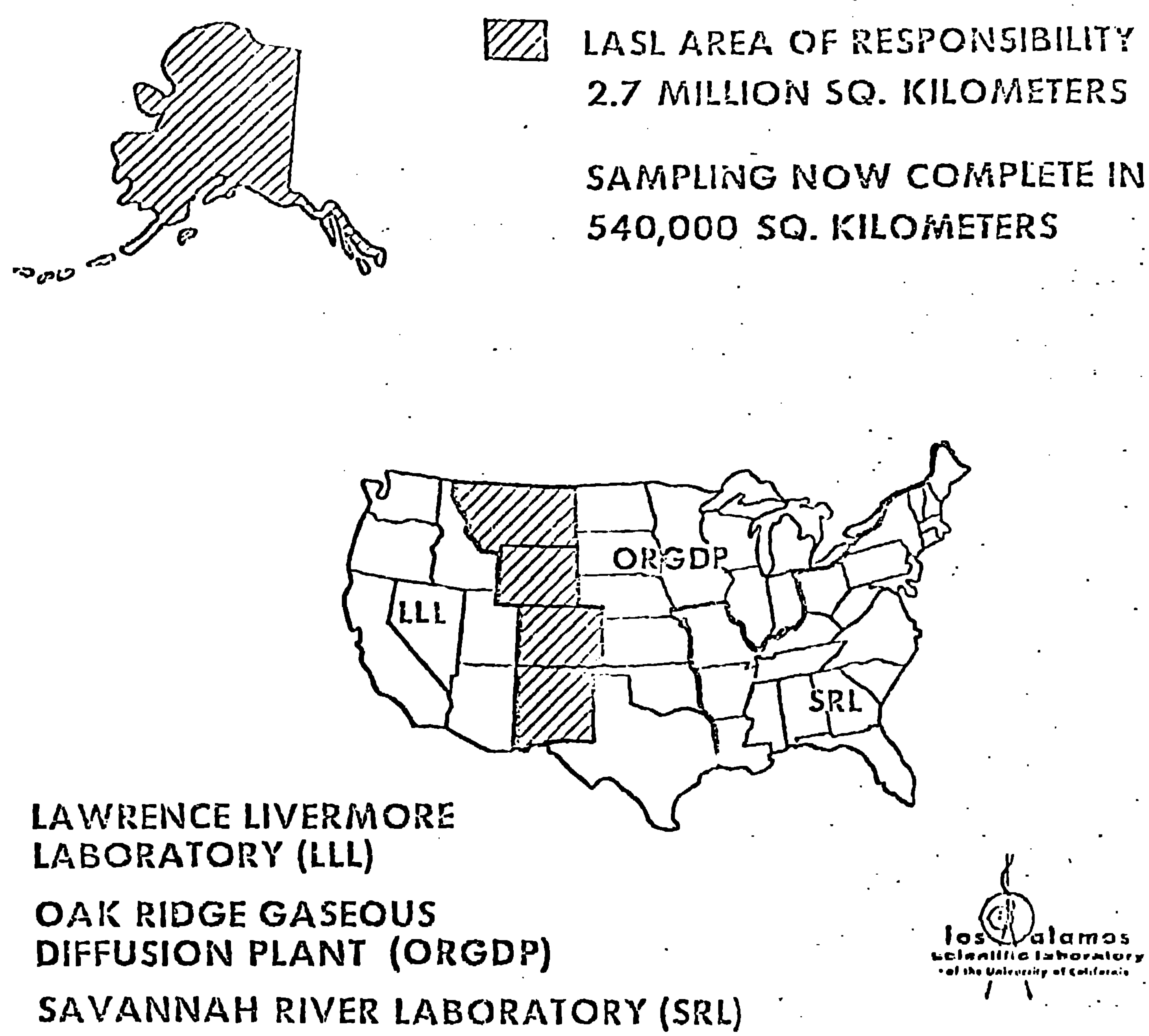

Fig. 1

Areas of responsibility for carrying out the HSSR across the nation.

TABLE I

AREAS OF THE STATES FOR WHICH THE LASL IS RESPONSIBLE
IN THE URANIUM HYDROGEOCHEMICAL AND STREAM SEDIMENT RECONNAISSANCE

Name	Land Area km^2 (mi^2)	Inland Water Area km^2 (mi^2)	Total Area km^2 (mi^2)
New Mexico	314,456 (121,412)	658 (254)	315,114 (121,666)
Colorado	268,753 (103,766)	1,246 (481)	269,999 (104,247)
Wyoming	251,755 (97,203)	1,841 (711)	253,596 (97,914)
Montana	377,069 (145,587)	4,017 (1,551)	381,086 (147,138)
Alaska	1,467,052 (566,432)	51,748 (19,980)	1,518,800 (586,412)
Totals	2,679,085 (1,034,400)	59,510 (22,977)	2,738,595 (1,057,377)

Approximately 35% of the area of the continental U.S.

U.S. Area*	7,676,719* (2,963,998)	150,898 (58,262)	7,827,617* (3,022,260)

*U.S. Area excludes Hawaii

about 35% of the continental US. The total time between April 1975 and October 1980 amounts to 5.5 yrs. The total funding anticipated for the entire nationwide project was about $40 million. Consequently, based on an extensive and detailed review of world literature on the subject,[2-20,22-60] and finally, on results of pilot work,[61,62] the LASL has concluded that—in keeping with the existing time and funding constraints—the most effective geochemical reconnaissance for uranium over the region for which it is responsible should follow strict, standardized sampling procedures and concentrate essentially on the determination of the uranium levels in both natural waters and waterborne sediments wherever possible.[3,9,13,14,32,40,50,57]

Again, on the basis of experience reported in various geologic regimes and climatic zones from around the world, it is considered necessary that at least one well-chosen water sample and/or sediment sample be obtained from each nominal 10 km^2, except in localities (such as lake areas of Alaska) where abundant water in contact with organic sediment should make one sample location per 20 km^2 or so suffice. While it has been shown in some areas that such a sampling density is not particularly promising,[5,7,19,29] in a considerable number of others it has proven quite adequate for uranium reconnaissance work aimed simply at delineating areas favorable for exploration.[12,13,30-40]

Also, since the dispersion trains of uranium in water are sometimes much greater than in sediment,[9,12,41] as is generally the case in carbonate terranes,[9,39] or vice-versa as has also been reported,[6,26,36,41] particularly in areas of bituminous rocks, the sampling of both waters and sediments wherever possible should enhance the adequacy of coverage as well as give better insight into the uranium geochemistry of any specific area.[8,19,39,56,57]

Furthermore, while the detailed sampling of water alone has been credited with playing a major part in the discovery of actual uranium deposits,[3,14,28,42] the sampling of sediment often holds greater promise,[11,26,57] particularly in areas where there is a large component of organic material[12,37,39,45,51] or where only dry stream beds are generally available.[8,19,41] Under advantageous conditions, which are not uncommon, the benefit of the depth of penetration to be accrued from water sampling (whether wells are actually sampled or not)[14,26,31] can also come from sampling sediment—particularly where organic material serves to integrate over the long term the uranium levels in the water with which it is in contact.[10,39]

At any rate, the sample densities decided upon indicate that the sampling of about 240 000 locations will be required over the total land area of approximately 2.7 million km^2 assigned the LASL. In order to meet this requirement, and effectively put the samples to use, it was recognized from the outset that a complex array of activities, involving numerous people, would have to be prepared for, scheduled, expedited, and carried out. A major portion of these activities are identified in Fig. 2 and described below.

Organization And Planning

This is the most critical portion of the survey because certain portions of any original plan can prove irreversible once instigated. Close teamwork is required between all the principals. To insure prompt action, a Survey Design and Evaluation Working Group was formed from within the LASL to give their full attention to organization and detailed planning of the project. Working closely with the contract administrators and utilizing consultants as deemed prudent, the Working Group included the Project Manager and an adequate number of Staff Members to bring to bear the laboratory-wide proficiency in geologic science, nuclear and conventional analytical chemistry and physics, electronics, and statistics. A cross-referenced map and document library was developed, containing all USGS quadrangle, topographic, and geologic maps; satellite, air photo, and infrared coverage; and hydrologic, magnetic, gravimetric, radiometric, mineral, ore occurrence, and land management maps available throughout the entire LASL region.

The LASL administrative chain supporting the HSSR comes through the Director's Office and the Associate Director for Research, on through the Geosciences Division Office and the G-Division Leader, and to Group G-5, the LASL Geochemical Applications Group. This Group, with the help of the aforementioned Working Group, has the responsibility for designing and carrying out the LASL HSSR project. Upon being formed, Group G-5 took over the map and document library and began organizing and planning the various aspects of the reconnaissance work. In addition to the writing of specifications for all contract work, the ordering and testing of all necessary field equipment and supplies and the development of field data collection, handling, verification, evaluation, and reporting formats were addressed. The aim throughout has been to standardize and automate

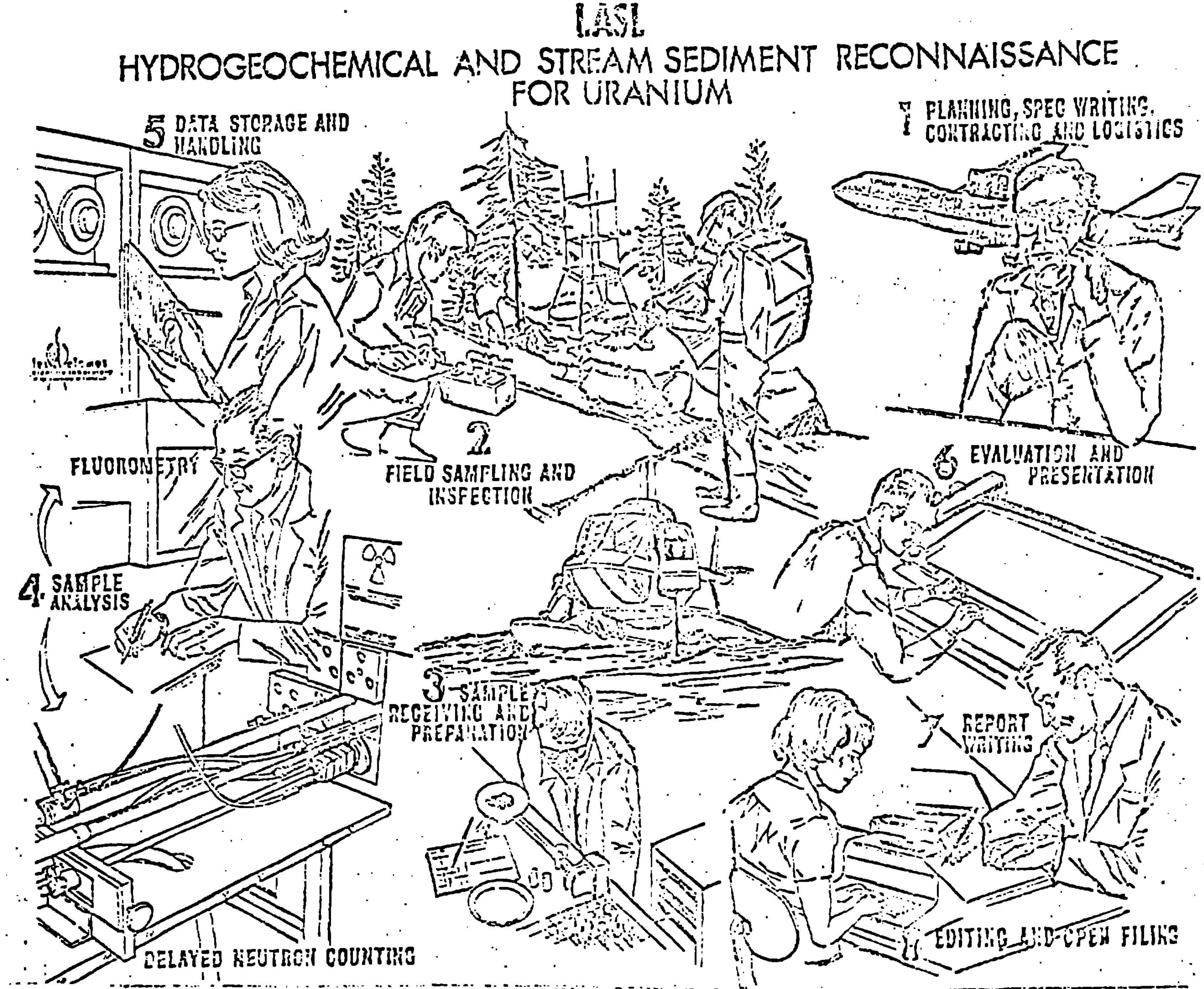

Fig. 2

Activities involved in the [illegible]

wherever possible. One of the first major items of business was development of the field data form shown in Fig. 3. Ultimately procured as a collated fire-, water-, and tear-proof front sheet, with three differently colored, carbonless-copy sheets and a sheet of twelve preprinted sample labels behind, the field data form is designed to allow versatility and take account of those parameters most commonly measured and put to use in uranium reconnaissance work around the world. Important interpretive factors to which these measured parameters relate are described below.

pH

Uranium is soluble over a wide range of pH,[8] and it is only when extreme high or low values are encountered that it becomes important to interpretation.[41] As pH decreases, uranium content increases.[30,31]

Specific Conductance

An increase in total dissolved solids (approximated by an increase in specific conductance) will usually be accompanied by an increase in uranium in natural waters.[37,39,43,48]

Equivalent Uranium

This is a measurement of surface radioactivity (uranium and thorium daughter products) at each location and is provided for use alone or with NURE airborne radiometric results when they become available. An abnormally high value is indicative of local radioactive mineralization.[19]

Temperature

The temperature of the water may influence the measured concentration of uranium to some extent since it controls the rate of certain chemical reactions and biological activity.[31,41]

Geology

Uranium content is generally higher in waters draining certain lithologies (such as acid igneous types).[28,50] Oxidizing ground waters often circulate in highly fractured, fissured, and faulted terranes, and often act to introduce dissolved uranium into the surface waters in favorable areas.[10,14,19,31,35,39]

Organics

These can be approximated from water description, density and type of local vegetation, and temperature. High organic content tends to rapidly adsorb uranium from water and correspondingly increase uranium in sediment.

Relief

Surface waters draining mountainous regions tend to have relatively short dispersion trains of uranium.[5,7]

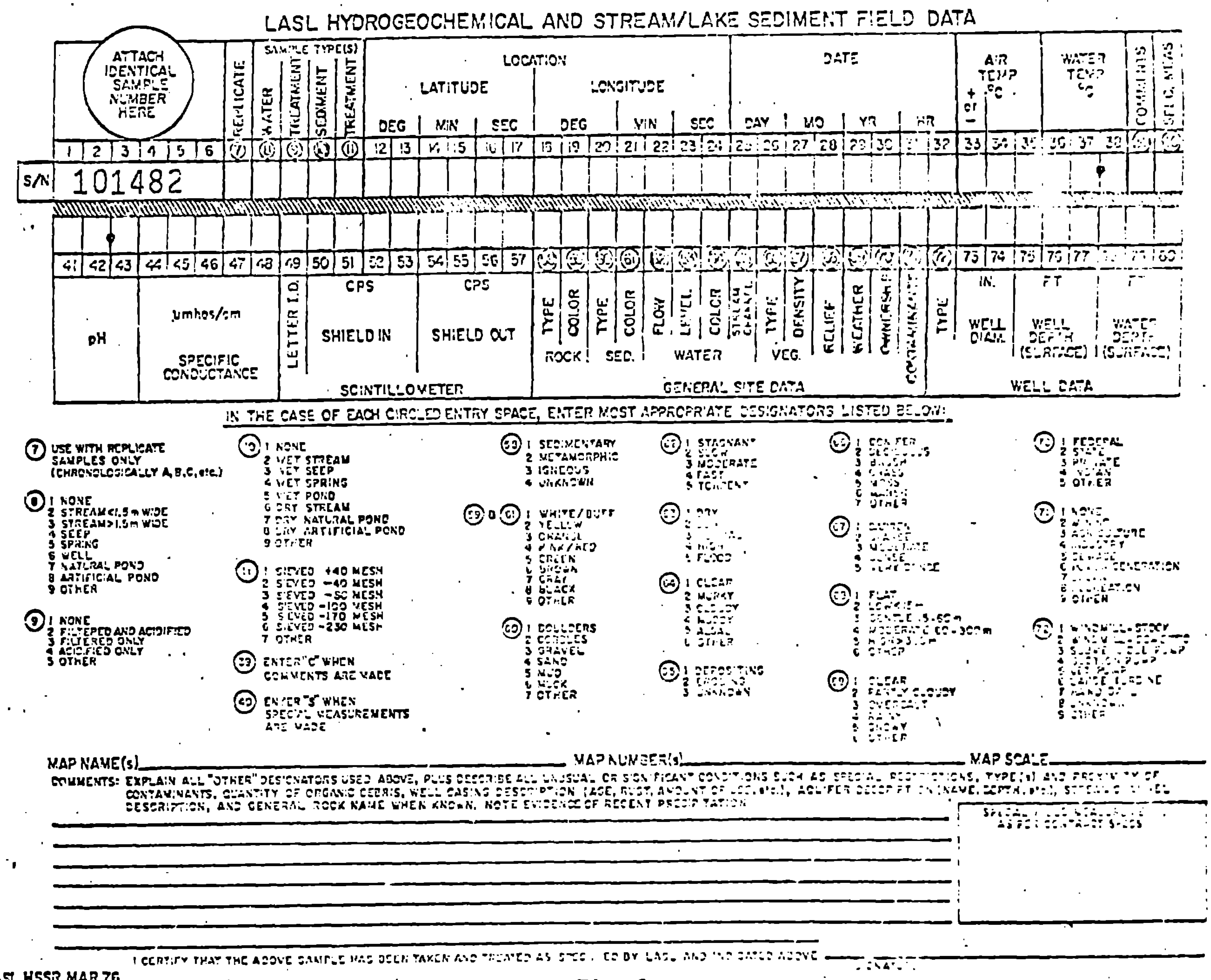

Fig. 3

LASL HSSR field data form.

Weather

Seasonal and weather conditions tend to change the uranium concentrations in surface waters and, to some extent, in sediment.[31,44] During periods of rapid snowmelt and high runoff the normal uranium concentration may be diminished by a factor of 1 to 3 times, due to dilution. Following periods of drought, the uranium content in rainwater runoff has been found to sharply increase for a brief period.[30,44] In standing waters (lakes and ponds), where evaporation is prevalent, the uranium content tends to increase.[3,31] Hydrogeochemical surveys should be completed over a short period of time to minimize the influence of seasonal change.[3,10]

Contaminants

The identification of potential sources of contamination such as metallic mines, dump residues, and acid mine waters can be important to making a proper interpretation of the uranium data.[5,22,30] The use of phosphate fertilizers might increase the uranium concentration in certain agricultural areas.[56]

Field Sampling

Except for orientation studies and check samples, all field sampling is done by commercial contractors according to the LASL specifications. All contracts are let by competitive bidding. Each contractor is supplied field maps with the desired sample types and locations symbolically premarked at the LASL. The maps are normally USGS quadrangles (either 7.5' or 15') but, where not available, Forest Service, State Highway, or other reasonably detailed maps are provided. As each location is sampled, a unique sample location number, preprinted on the transparent adhesive labels provided with the identically numbered field data forms, is pasted over the precisely marked site on the field map. The latitude and longitude of each location is computed by the sampling contractor. Every location is later checked (and corrected if necessary) at the LASL by overlaying computer-produced location plots on the field maps used. The latitudes and/or longitudes are corrected if the overlay locations are displaced by more than ~ 50 m from the locations marked on the field maps. When a desired location cannot be sampled as specified, an alternate sample type or location as near as possible to the original one is picked, and the new sample type and/or location is/are marked on the field map and properly labeled as above. Areas are normally sampled in approximately 20 000 km^2 blocks to match USGS 2° map sheet boundaries.

Water Sampling

Water samples are taken directly from the source wherever possible, filtered through a 0.45-micron membrane filter into one each, prewashed and sealed, 41-ml reactor "rabbit" and 25-ml vial (both polyethylene). Both are then acidified to a pH ≤ 1 with 8N, reagent-grade HNO_3. All sample containers are doubly labeled with the preprinted, adhesive labels carrying the same sample location number preprinted on the field data form. Springs are sampled as near to their point of emergence as possible; stream waters are taken from fast-flowing current away from the bank; ponds (including small lakes and reservoirs) are sampled from just below the surface, away from the bank; and well waters are taken near the wellhead if the well is pumping or from a holding tank if not. Because of the high costs involved in Alaska, the time consuming operation of filtration is deleted.

Sediment Sampling (Wet or Dry)

Enough fine-grained, organic-rich, water-transported sediment to yield a composite sample of ~ 25 g after processing (as indicated below) is taken from beneath the water level (where water exists) at three closely adjacent spots at each location. This is done with a polyethylene scoop, after the water sample (if any) is taken. The sediment is put into a new, clean, and originally sealed, zip-top polyethylene bag and properly double-labeled for delivery (with the field data form) to the contractor's drying facility. After drying at ≤ 100° C, each sample is sieved through stainless steel sieves to -100 mesh. The -100 mesh fraction is put into a prewashed, 25-ml polyethylene vial, appropriately double-labeled (using labels from the data form), and sealed for shipment to the LASL. In lake sampling campaigns in Alaska, the sediment sample is taken with a specially designed, suction-operated bottom sampler dropped from a helicopter.

Field Measurements

The air temperature, taken in the shade at the time of sampling, is recorded to the nearest whole degree centigrade. The water temperature is measured in the source water and recorded to the nearest one-half degree centigrade. All temperature measurements are made with quality, precalibrated thermometers. The pH of the source water is measured with a calibrated, portable pH meter or multi-range pH paper, and recorded to the nearest one-tenth of a pH unit. The specific conductance (μmho/cm) of the source water is measured with a calibrated, temperature compensated (25° C) portable meter after the attached sample cup or probe has first been rinsed three times in the source water. The scintillometer readings, taken on a flat, dry spot within a few meters of the sample location, are measured with a portable scintillometer. Two readings are recorded, the first with a radiation shield in place (blocking out ground radiation), and the second with the shield removed. The readings (in counts/s) are converted by computer to give the equivalent uranium (eU) value set forth in the data listings. No scintillometer readings are taken when lake sampling in Alaska.

Field Observations

These represent the best subjective judgment of the field sampler on location and include very general descriptions of the local bedrock, sediment, water, vegetation, terrain, weather, possible contaminants, and water well configuration, if applicable.

Sample Receiving and Analysis

Samples are received from the field along with the location maps and all but the last carbonless copy of each completed field data form. They are shipped or delivered in special LASL-furnished, insulated shipping containers which will hold the samples obtained from nominally 200 sample locations. After inventorying all samples against the data forms, the water samples in the 25-ml vials are sent to Group CMB-1, the LASL Analytical Chemistry Group, where determination of the uranium content is done by fluorometry.[64] The water samples in the 41-ml reactor rabbits, as well as the dried and sieved sediment samples, are sent to Group P-2, the LASL Reactor Research Group, where all delayed neutron counting (DNC) for uranium and any multielement work by neutron activation analysis (NAA) is done. The use of DNC for determining total uranium in sediment[11,16,17,25,55,63] and uranium in water,[17,25,41] as well as the use of NAA for multielement analyses in general geochemical surveying, offers great advantages.[20,25,41,55,63]

Water Samples Analyzed for Uranium by Fluorometry

In a controlled laboratory environment, a NaF (98%)-LiF (2%) flux pellet is prepared and placed on a platinum dish. The 25-ml water vial is vigorously shaken and a 0.25-ml aliquot of water is withdrawn, dropped onto a flux pellet, and then evaporated under a heat lamp. The sample flux is then heated until fused. After it cools, it is excited with ultraviolet radiation in the fluorometer, and the measured fluorescence is read, recorded, and put through a computer routine using standards and blanks run at the same time to obtain the uranium concentration.[64] Water samples which have uranium concentrations in excess of 10 ppb (the upper limit of detection of the fluorometry system without recalibration) are reanalyzed using the delayed neutron counting technique described below. Those with < 10 ppb have their uranium concentration automatically entered in the data base.

Water Samples Analyzed for Uranium by Delayed Neutron Counting

Only waters with > 10 ppb uranium are assayed using DNC. Samples taken in the 41-ml rabbits are thoroughly cleaned (exterior) before analysis. Samples received in 25-ml vials (used exclusively in some of the early work) are transferred to clean, labeled, 41-ml rabbits before being analyzed. Each water sample is weighed, and its weight (less that of the rabbit) and location number are recorded. The rabbits are then loaded into a 25-sample transfer clip. The reactor pneumatic transfer system and background radiation levels are checked, and the system is calibrated using four standards. The transfer clip is installed on the pneumatic feed line, and the count control is set (typically, a 60-s irradiation, a 30-s delay, and a 60-s count is used, but this can be changed to accommodate abnormally high or low uranium concentrations). The samples are automatically cycled through the system and the uranium concentration is automatically computed in ppb and entered into the data base.

Uranium Analysis of Sediment Samples

All sediment samples are analyzed for total uranium by DNC. A split of each sample (dried and sieved as described) is transferred to a clean 4-ml rabbit, weighed (less the tare), and recorded along with the appropriate location number. The readied rabbits are loaded into a 50-sample transfer clip. The reactor pneumatic transfer system and background radiation levels are checked, and the system is calibrated as above. The transfer clip is installed and the count control is set (typically, a 20-s irradiation, a 10-s delay, and a 20-s count is used). The samples are cycled through the system and the uranium concentration is automatically measured, computed in ppm, and entered into the data base.

Data Verification, Handling, and Presentation

Once the incoming samples have been inventoried, the field data forms are manually checked for incomplete or inconsistent information. Once this is done, the original front sheet is torn off and sent to Group Q-12, the LASL Energy Division's Statistical Analysis and Assessment Group, for entry into a computer data base. A separate data base is established for each individual area of approximately 20 000 km^2. As the data is entered, it again goes through a verification routine, this time programmed into the computer. Once all the field data for an area are entered, overlays are plotted of all sample locations at the same scale as the maps used in the field and carrying the location numbers. The overlays are checked against these maps and all necessary corrections are made. By the time this is done, the uranium determinations on both the waters and the sediments are usually completed and fed into the data base. After a cycle of data-base recheck and cleanup (to insure that there is a uranium value for each and every water and sediment sample), the first set of a standardized sequence of normal and log-normal frequency histograms of the uranium concentrations for each individual sample source (i.e., springs, streams, wells, etc.) and uranium concentration overlay plots (distinguishing between water and sediment sample sources) is automatically produced. By examination of this output and the data listings through no less than two (but no more than three) iterations, the final versions of the histograms and uranium concentration overlay plots for the HSSR report on an individual area are derived. These, coupled with the computer data output listings and the results of geological, geochemical, and any statistical evaluations that have been carried out, will go to form the standard LASL HSSR reports as briefly described below.

Data Evaluation and Presentation

Neither time nor funding will allow in-depth evaluation of all the field and analytical data. The principal aim will be to see that all data is standardized, consistent, and valid as far as the described sampling techniques and analytical results are concerned. The LASL does, however, plan to include a geologic base map at 1:250 000 scale whenever possible, unless one is publicly available with the same boundaries as the area covered in a specific report. In addition, a limited amount of geological, geochemical, and hydrological interpretation and/or statistical analysis will also be included as part of the standard LASL reporting format as outlined below.

LASL Reconnaissance Report Format

A. Abstract

B. Text (~ 10 pages), to include sections on:

1. Geography, climate, weather (at the time of the survey),
2. Geology, hydrology, water quality, and geochemical considerations,
3. Uranium occurrences in the area,
4. Empirical and statistical evaluations,
5. Conclusions.

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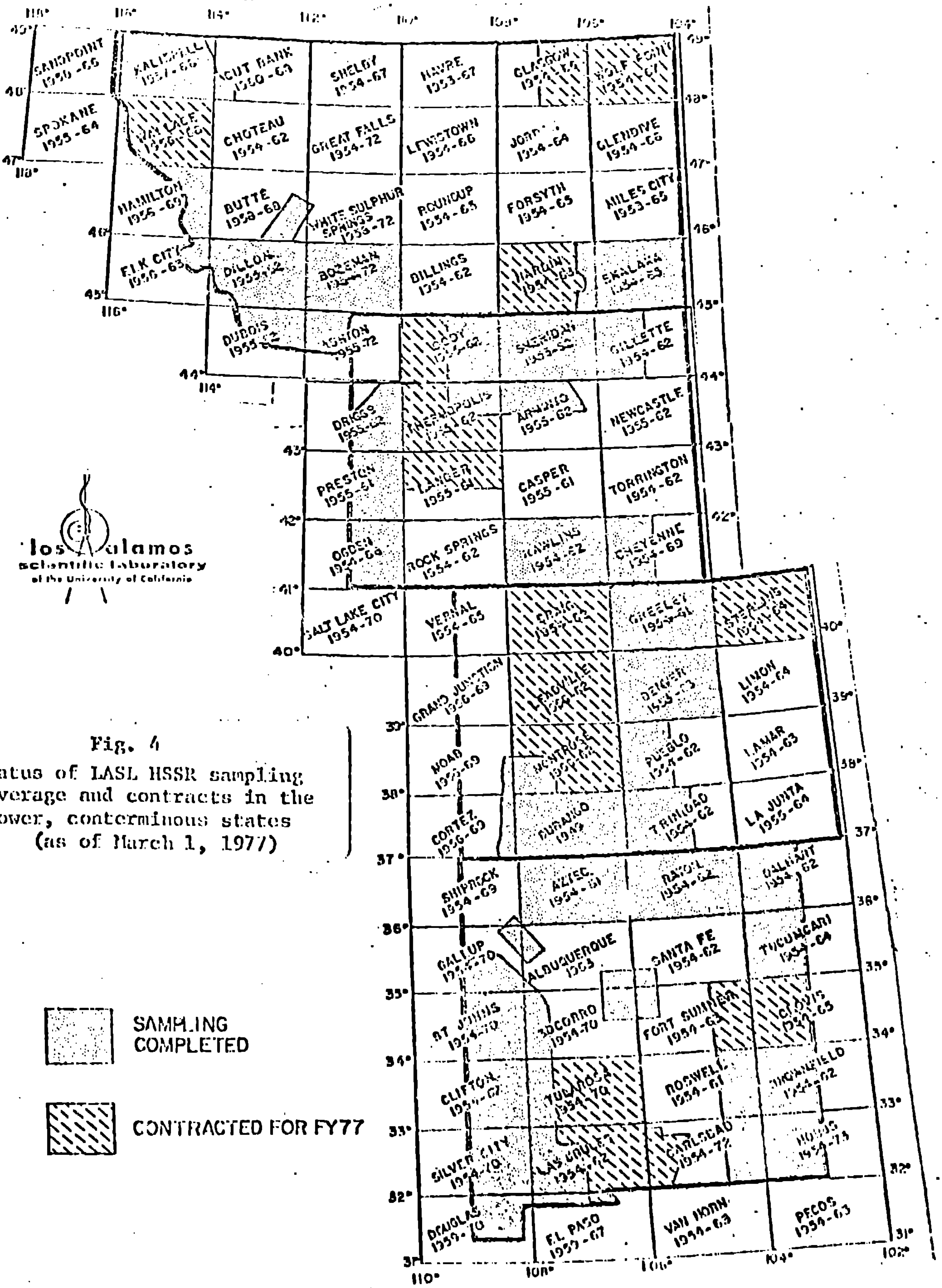

Fig. 4
Status of LASL HSSR sampling coverage and contracts in the lower, conterminous states (as of March 1, 1977)

C. Standard procedures used in taking, treating, and analyzing the samples.

D. Data listings (including sample information, field data, and uranium concentrations) for:

   1. Water samples.
   2. Sediment samples.

E. Key to codes used in data listings.

F. Maps and overlays (at 1:250 000 scale).

   1. Geology map.
   2. Sample location overlay.
   3. Graphic plot of uranium concentrations--water.
   4. Graphic plot of uranium concentrations--sediment.

G. References cited. It is planned that at least the most current and best 10 or 12 references on the geology, water quality (chemistry), and uranium geochemistry of the area will be referenced, such that the reader will have the complete bibliographic data for such reports at his fingertips.

III. PRESENT STATUS OF THE LASL HSSR PROJECT

The status of the LASL HSSR sampling coverage and sampling contracts in the states of New Mexico, Colorado, Wyoming, and Montana as of March 1, 1977, is shown in Fig. 4. The status and schedule of sampling in Alaska is shown in Fig. 5. At the present time, one of the five sampling seasons has passed and the LASL has sampled about 19% of the total area for which it is responsible. If this rate can only be continued, the assigned HSSR for uranium throughout the LASL's region can be accomplished. Every effort is being put into insuring that the data accruing is valid and can be effectively put to use by the public sector as well as the ERDA.

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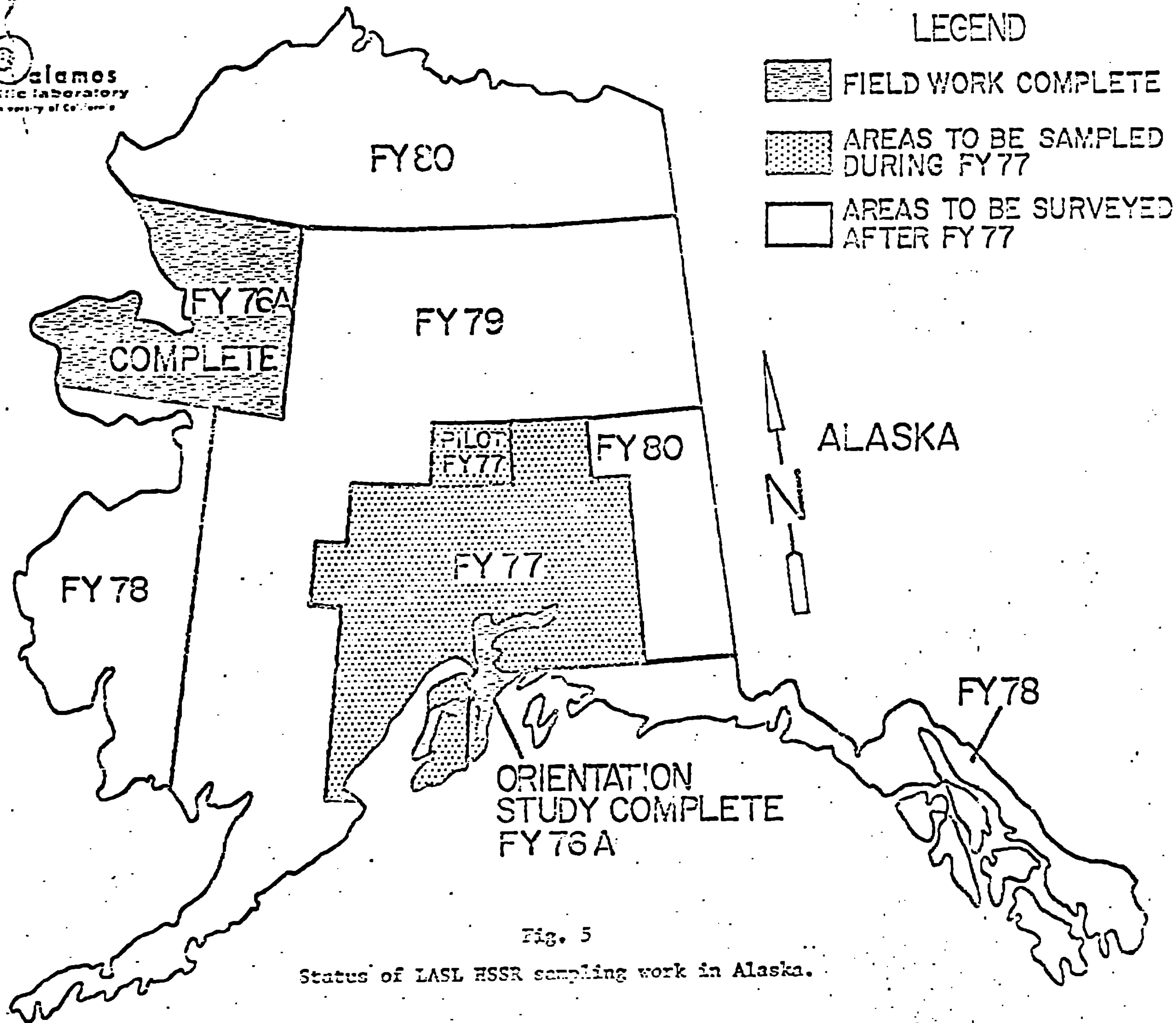

Fig. 5

Status of LASL HSSR sampling work in Alaska.

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