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DATA ACQUISITION AND ANALYSIS SYSTEM FOR EMERGENCY ENVIRONMENTAL SURVEILLANCE

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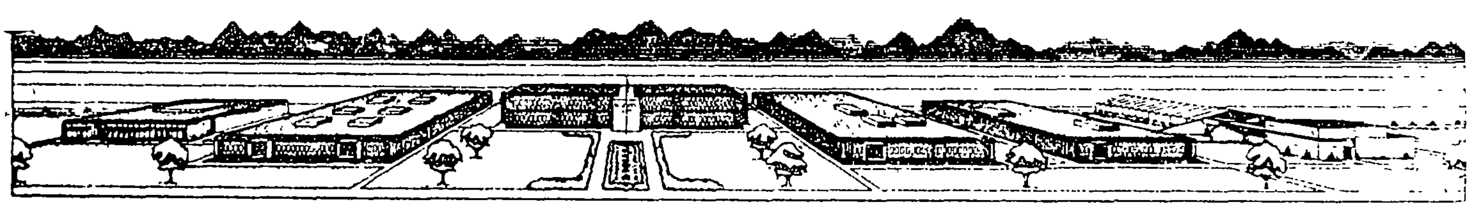

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INTRODUCTION

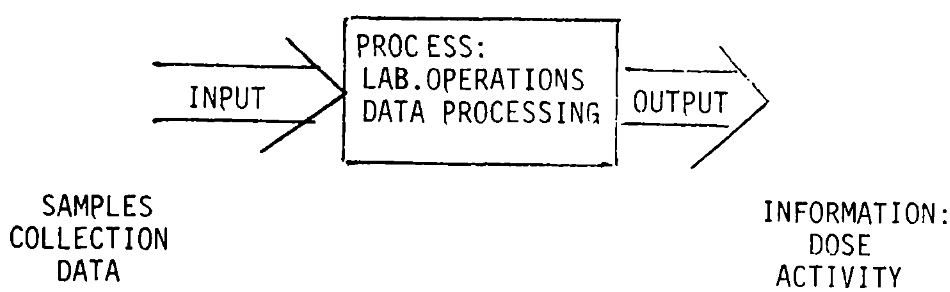
An environmental surveillance program, whether it be of a continuing nature or an emergency action, can be represented as an integrated system. Such a system, irrespective of its complexity can in turn be described in terms of three basic functional processes:

INPUT, which is the basic component on which the system operates;

OUTPUT, which is the end result of the operation; and

PROCESS, which is the activity which transforms input into output.

In the context of a laboratory operation supporting an environmental surveillance program, input may be explicitly defined as samples and collection data. Output will be (in the case of radiological surveillance) activity concentration or dose estimates. The process is the laboratory function of analyzing samples and processing the data generated therefrom. Such a system is shown below.



The system described herein is the "PROCESS" referred to above. It is an integrated system for the rapid analysis of large numbers of environmental samples.

GENERAL SYSTEM DESCRIPTION

Data acquisition and analysis is a production system designed to process routine as well as special samples.[1] The predominant routine sample types are: milk, water, air filters, and charcoal cartridges. Other types of samples received periodically are: vegetation, animal feed, animal tissue, soil, and gas. Sample load for gamma analysis and chemistry is typically 1000 samples/month. In addition, 100 air filters/day are received for beta counting. During peak periods as many as 200 samples per day are processed for gamma analysis and/or chemistry. The types of analyses performed on a sample (dependent upon the sample type and program requirements) are: gross and specific alpha and beta counting, specific radiochemistry and gamma counting with qualitative and quantitative spectral analysis.

For the routine continuing surveillance programs, a systematic procedure has been established for processing both samples and data. Special samples, notably those related to emergency situations require modification of the basic system in order to provide quicker through-put of data.

A computer system is utilized in the analytical process in three ways: it allows rapid conversions of data from one medium and format to another; it performs calculations and presents its resulting output in a form suitable for analysis by professional personnel; and it allows for the storage and retrieval of accumulated data.

Operationally, the processing of samples is complex. Many processes occur simultaneously and are mutually dependent. Successful processing of samples requires procedures that extend across operational unit functions. Hence, description of the system is based on a general breakdown according to processing function rather than operational unit. This breakdown is developed as follows:

1. Sample control system
2. Gamma analysis system
3. Air sample analysis system
4. Chemistry data analysis system
5. Data Management system

All samples are processed through sample control. Air filters and charcoal cartridges are then processed through the air system (possibly incorporating a loop through the gamma and/or chemistry system). All other samples are processed through the gamma and/or the chemistry system. Data from all samples are processed through the data management system. These functional sub-systems are shown schematically in Figure 1.

SAMPLE CONTROL

Sample control records, prepares, and distributes for appropriate analysis all samples received. Each sample which is to receive gamma analysis or chemistry is given a unique laboratory number. Collection information is transcribed from the sample collection data form to a four-part log-in

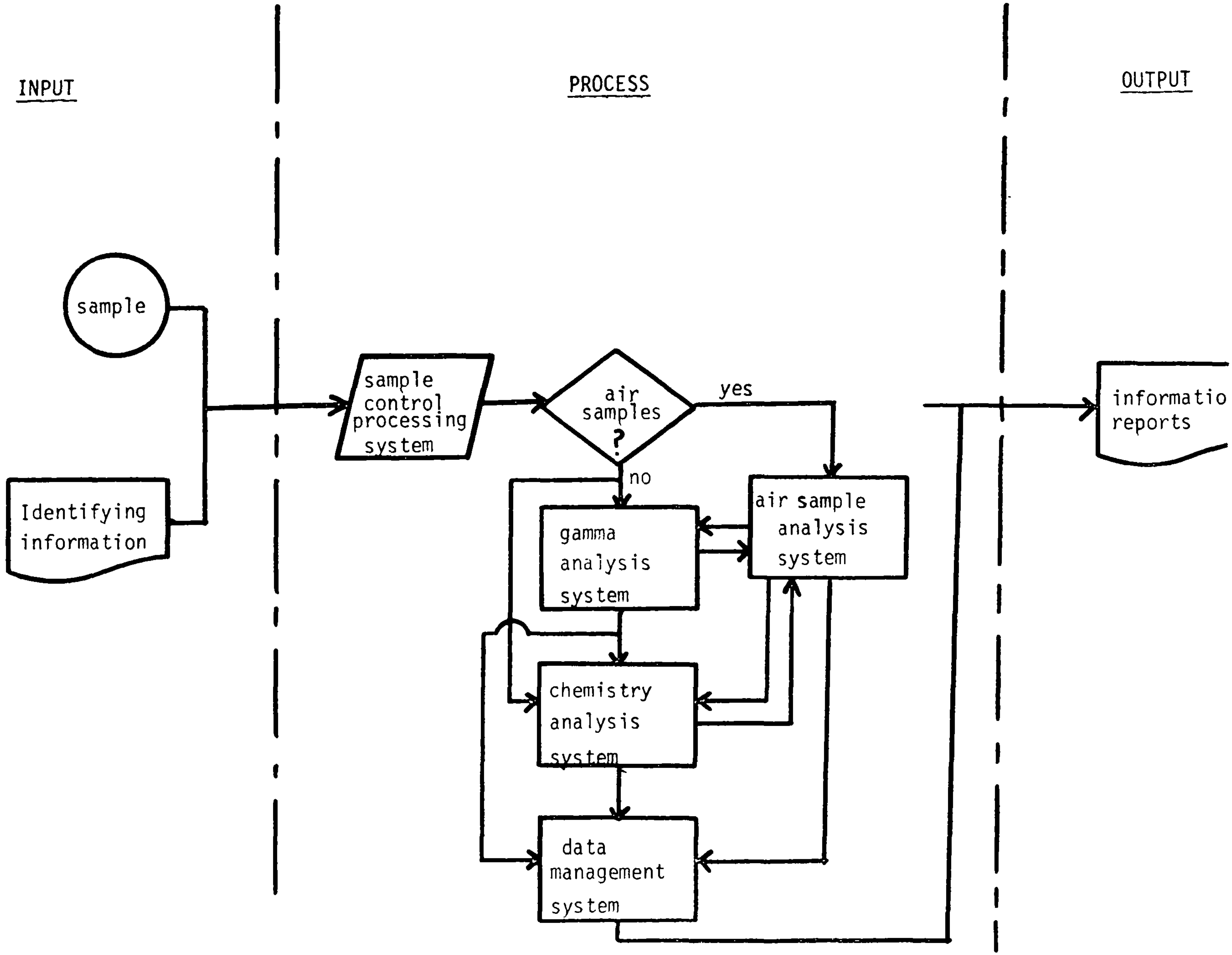


FIGURE 1

Data Acquisition and Analysis Processing Functions

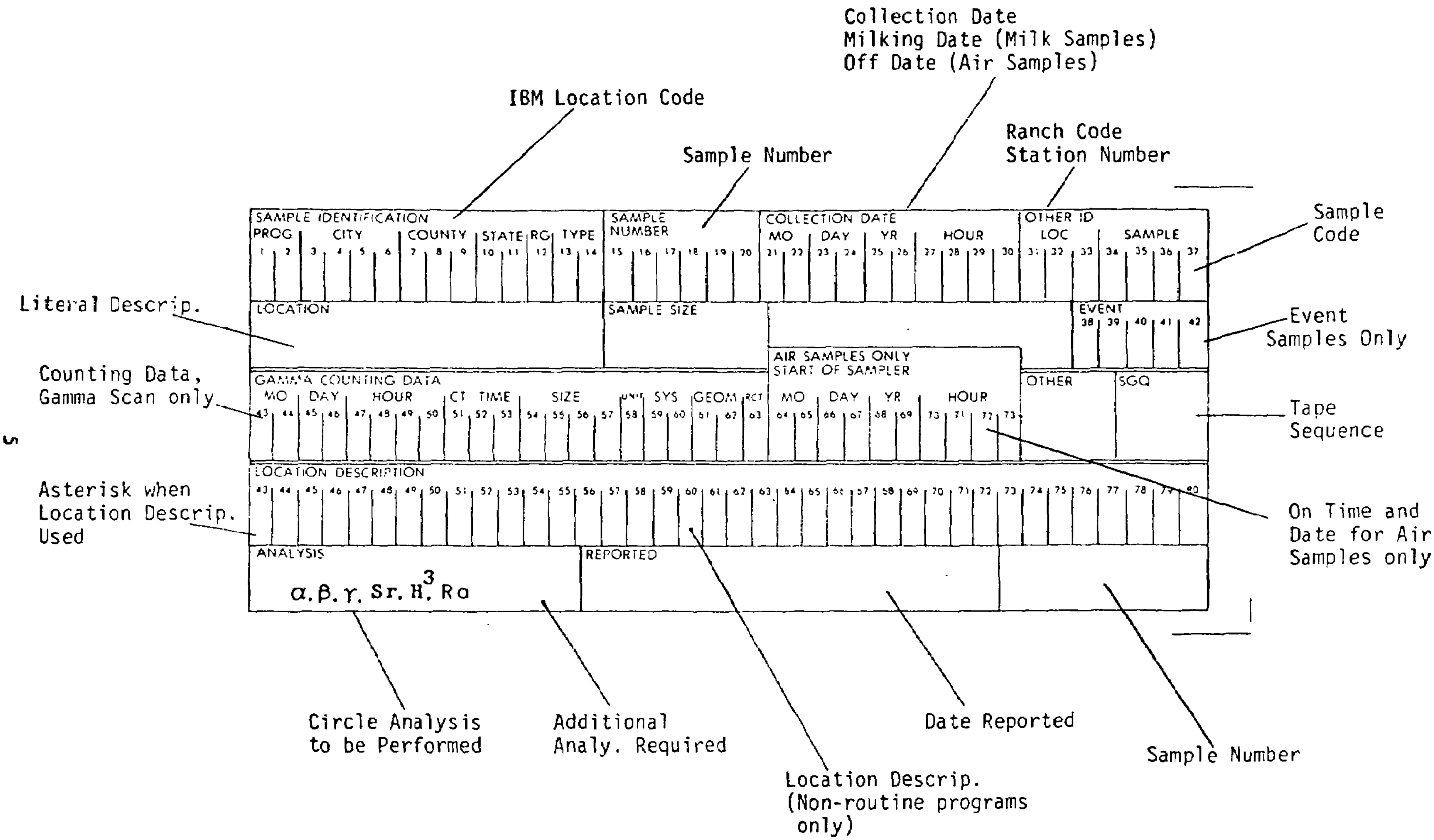


FIGURE 2
SAMPLE CONTROL LOG-IN FORM

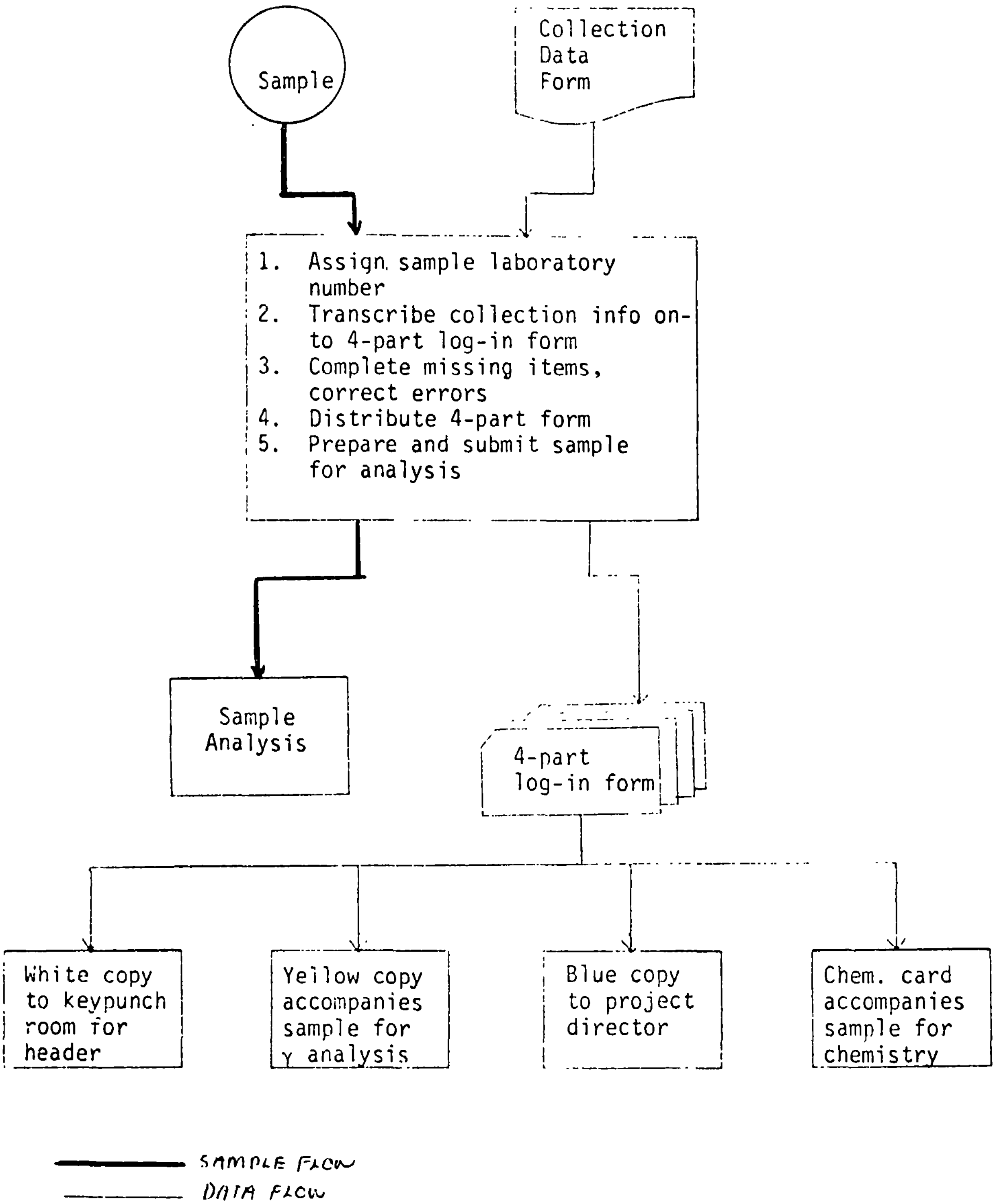


FIGURE 3
SAMPLE CONTROL PROCESS

form (Figure 2). An appropriate location code, sample type code, and program code are added. The sample is then forwarded with the appropriate copy of the log-in form for selected analysis (Figure 3).

Air filters and charcoal cartridges are not immediately logged in. They are posted as to date received and submitted for gross beta and gross gamma counting, respectively. If the gross counts exceed pre-set limits, they are returned to sample control for log-in and subsequent isotopic analysis (see Air System).

Sample control is also responsible for contamination control in the analytical laboratories. During emergency periods, all samples are appropriately repackaged and sealed prior to submittal to low-level counting facilities.

GAMMA ANALYSIS SYSTEM

General Description

The "gamma system" encompasses the sample handling, counting, data analysis and interpretation, and reporting procedures involved in quantitative gamma spectral analysis. In processing large numbers of samples by quantitative gamma spectral analysis, a number of assumptions must be made prior to the analysis. The most important are related to detector response characteristics, system reproducibility, and radioactivity standards library. An analysis is made and the results reviewed in order to validate the initial assumptions. If the assumptions are found to be incorrect, they must be altered and the analysis repeated.

Instrumentation

The gamma counting facility consists of five analyzers each operating in split mode with two thalium activated sodium iodide detectors. The analyzers are TMC Model 404C, 400 channels, with multiple input. The detectors are 4-inch-thick by 4-inch-diameter crystals and are manufactured by the Harshaw Chemical Corp. The crystal housing, hermetically sealed, is of 0.019-inch Type 304 stainless steel. A 3 1/2-inch diameter by 5/16-inch-thick Vycor optical window is coupled to a 5-inch-diameter, RCA, venetian blind, dynode multiplier phototube, Type 2065. The detector assembly is seated on a lucite shelf in a steel shield of 6-inch-thick wall. The chamber within the shield is 20 by 20 by 24 inches, lined with 0.1-inch lead, 0.03-inch cadmium and 0.015-inch electrolytic copper.

Readout from each analyzer is by means of perforated tape and typewriter (the perforated tape is processed by the computer for analysis and data storage).

Calibration and Quality Control

Routine samples are counted in one of four standard geometries. Four special purpose geometries are used for limited studies where sample quantity or processing may require a non-standard configuration.

TYPE	GEOMETRY	DESCRIPTION
Standard	01	2-inch-diameter planchet
Standard	02	4-inch-diameter planchet
Standard	03	400-ml polethylene container
Standard	06	3.5-liter Marinelli beaker
Special	12	250-ml polyethylene container
Special	15	1-liter cubitainer
Special	16	250-ml resin
Special	17	Soil

A radioactive isotope standard is counted on all detectors for each geometry and nuclide to be analyzed. An effort is made to recalibrate the more common long-lived isotopes on a yearly basis. A gamma efficiency vs. energy curve is used for quantitation of nuclides that are not readily available as standards.

A 400-minute background count is accumulated daily on each system. Both the standard spectra and background count are processed through appropriate computer programs and stored on disk for recall during data analysis.

In quantitative and/or qualitative analysis by gamma spectroscopy, the validity of the analysis depends on the satisfactory and reproducible operation of the instrumentation. The first level of quality control, then, is that applied to the instrument itself.

System response is checked daily by counting a ^{207}Bi reference standard. This nuclide has a 30-year half-life and two prominent gamma emissions at 0.570 MeV and 1.063 MeV. The source is counted for ten minutes and read out on punched paper tape. The tape is then run through a computer program (B1207)* and the following parameters calculated:

1) peak locations
2) difference between peak locations
3) sum of counts under photopeak
4) resolution
5) peak ratio

Peak location and interval are maintained within 0.5 channels of the theoretical. If both peaks are shifted equally, a zero shift is indicated. A gain change is indicated by a proportional shift. Daily corrections are made to maintain the energy calibration within the specified limits. Control charts are updated daily to evaluate long-term trends. These are maintained for each detector system.

*A name in capital letters designate the name of the computer code in the process. Full documentation of all computer programs used by the Western Environmental Research Laboratory is available at the Laboratory. Documentation includes a complete index of programs, one-page summaries identifying the nature of the programs, source language listing, flow charts, data set-up information and operating instructions.

The sum of counts within the photopeak provides a check on counting efficiency. Resolution provides a measure of energy separation. Charting of sum counts can detect long-term failure of the detector while resolution charging can indicate gross detector failure.

Peak ratio is the ratio of counts in the two peaks. Although not necessary as a quality control check, it does provide another sensitive indicator of change in detector response.

Background data are accumulated daily to check abnormalities that may occur on a long-term basis. After normal operations, the systems are set for a 400-minute background count. The gross gamma count is reported daily and plotted on a control chart. If the background is unusually high, the spectrum is checked to determine the reason for the increase. A background quality control chart is maintained for each system to detect long-term trends and fluctuations.

Sample Counting and Data Flow

Figure 4 shows the routine sample and data flow through the gamma system. The sample is received along with the yellow copy of the log-in sheet. Counting data are added to the log-in form. The sample is counted from 10 to 40 minutes depending on sample type, sample load, and desired data precision.

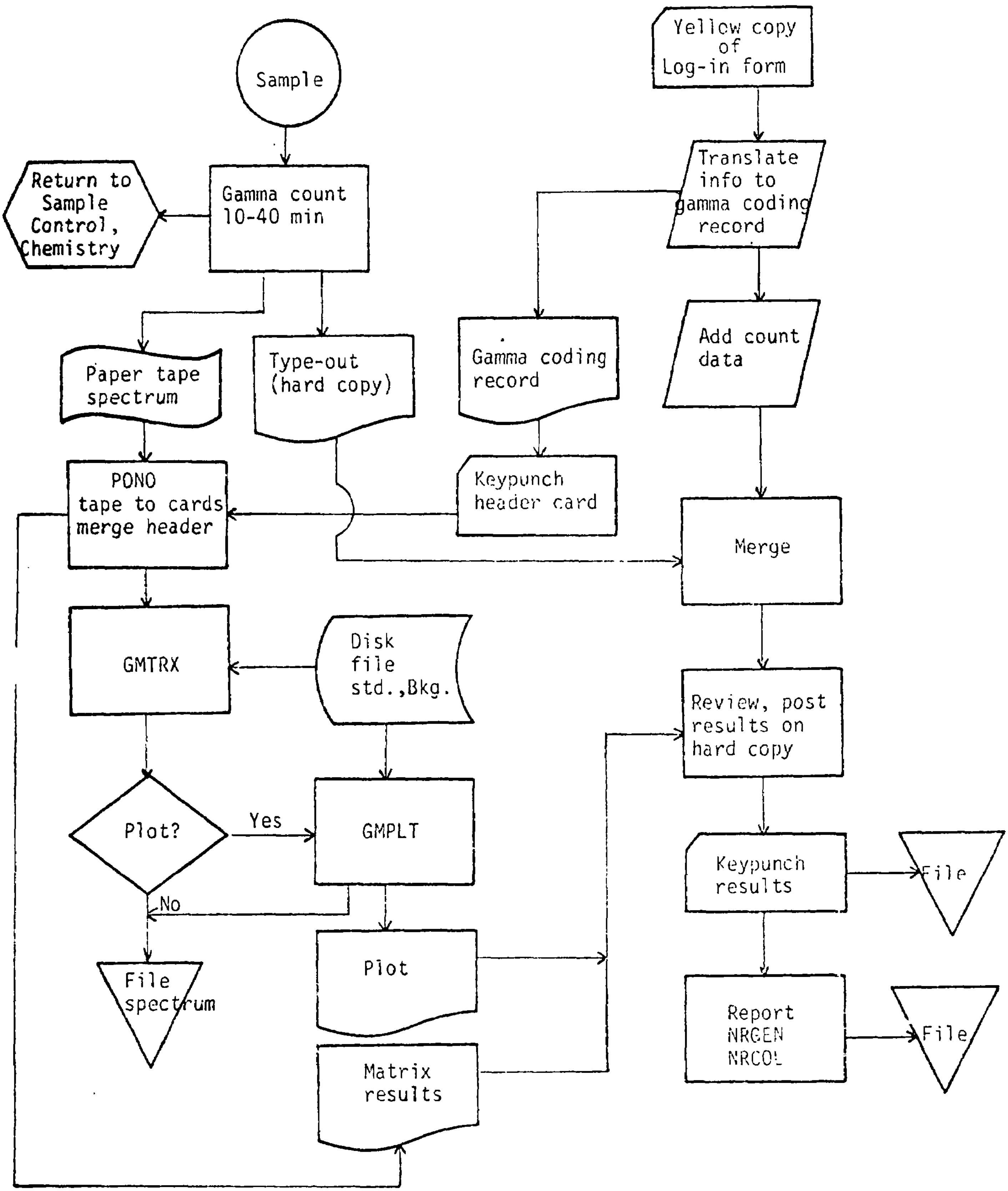


FIGURE 4
SAMPLE AND DATA FLOW GAMMA ANALYSIS SYSTEM

Count Time (Minutes)

Sample Type	Routine	Event-Related
Air Filter	10	10
Charcoal Cartridge	10	10
Milk	40	20-40
Water	40	20-40
Feed	40	20-40
Vegetation	10	4-10

Count data are read out on punched paper tape and typewriter printout. A gamma analysis coding record is used to identify the sequence of spectra on the punched paper tape. The yellow log-in form is attached to the typewriter printout to identify spectra. This becomes the hard copy of the raw data that is retained for future reference as needed.

The gamma counting information is keypunched, producing a gamma header card for each sample counted. The punched tape is converted to cards and merged with the appropriate header card (PONO). The data are then processed through a gamma analysis program (GMTRX) which utilizes the simultaneous equation technique to resolve the spectrum.[1] Radionuclide standards and background information are stored on disk for access by the gamma analysis computer program. The resulting data are reviewed and the results posted on the hard copy. A plot of the spectrum can be generated if necessary. The results are then keypunched onto cards for subsequent reporting.

Data Analysis

Quantitative gamma spectral analysis is performed by the simultaneous equation or matrix technique. Data relating to the interference coefficients among radionuclides are utilized in the matrix. Three files of data are maintained for routine analysis, each containing a library of eight radionuclides. These are grouped, according to whether the predominant activity is of long, intermediate, or short half-life as follows:

	Long-Lived		Intermediate Half-Life		Short Half-Life	
	Isotope	Peak Energy MeV	Isotope	Peak Energy MeV	Isotope	Peak Energy MeV
1.	^{144}Ce	0.13	^{147}Nd	0.09	^{141}Ce	0.14
2.	^{131}I	0.36	^{141}Ce	0.14	^{131}I	0.36
3.	^{106}Ru	0.51	^{132}Te	0.23	^{133}I	0.53
4.	^{137}Cs	0.67	^{143}Ce	0.29	^{137}Cs	0.67
5.	^{95}Zr	0.76	^{131}I	0.36	^{132}Te	0.23
6.	^{54}Mn	0.84	^{103}Ru	0.50	^{99}Mo	0.75
7.	^{40}K	1.46	^{95}Zr	0.76	^{135}I	1.28
8.	^{140}Ba	1.60	^{140}Ba	1.60	^{40}K	1.46

The appropriate data set is utilized according to the circumstances. Knowledge of event characteristics allows special data sets to be specified. These data can be assembled in any combination of up to eight radionuclides.

The program calculates the activity concentration of each of the nuclides at the time of count and at time of collection. If an isotope is determined to be absent, it is deleted from the matrix and a recalculation is executed. This process continues until the matrix is exhausted.

System Performance

1. Data Turnaround

   On a routine basis gamma matrix output is available within 24 hours of sample receipt. During an emergency situation, a batch of 50 samples can be logged in, counted, analyzed and reported within 6 hours. As many as 200 samples can be processed in a day.

2. Sensitivity

   It should be noted that in gamma spectral analysis, minimum sensitivity is dependent upon both isotopic mixture and relative isotope concentration as well as sample counting time.

   Those values stated below for gamma isotopic results refer to low level environmental samples.

Minimum Sensitivity, Gamma Spectral Analysis

Sample Type	Minimum Sens. PCi/L or kg.		
	40 Min Count	20 Min Count	10 Min Count
Milk (3.5L)	10	20	-
Water (3.5L)	10	20	-
Food (3.5L)	10	-	-
Feed	-	-	50
Veg.	-	-	50
Air (500M^3)	-	-	0.1

AIR SYSTEM

General Description

Approximately 100 air filters and 25 charcoal cartridges are received daily for analysis. The air filters receive a sequence of three gross beta counts and the activity is extrapolated back to the end of collection. Extrapolated gross beta activity is used to document trends in long-lived airborne radioactivity. Activity at time of count is used as a screen to detect sudden increases in gross activity. If the beta activity is above a preset level at time of count, the sample is submitted for gamma analysis. All the charcoal cartridges receive a gamma scan. If the gross gamma activity is above a preset guide, isotopic quantitation is performed. Processing of these samples follows the procedures established within the gamma analysis system description.

Instrumentation

The counting systems consist of three Beckman (Sharp) Widebeta counting systems. Each counter has a 5-inch-diameter thin window (100 mg/cm^2) gas flow, proportional detector which is incorporated in a 6-inch-thick lead shield to eliminate background from environmental radiation. The sample detector is operated in anticoincidence with a cosmic-ray guard counter which removes the cosmic-ray component of background.

The systems use pure methane (99.99%) as counting gas and are operated in the proportional region (3 KV) to provide for both alpha and beta counting based on pulse height discrimination. Simultaneous alpha and beta accumulation and readout are provided. The systems incorporate an automatic sample changer (60 sample capacity) and an automatic data readout capability. Readout is by means of IBM Model 026 Hollerith card punch.

The air filters are counted on 4-inch stainless steel planchets. The collection data pertaining to the sample are pre-punched on the Hollerith card which is placed in the card punch in the same sequence as the samples in the sample changer. Counting data for each sample are then automatically punched onto designated fields on the Hollerith card.

Calibration and Quality Control

Each system is calibrated over a range of beta energies and self-absorption. Typically, using $^{90}Sr/^{90}Y$ in equilibrium, with an average maximum beta energy of 1.40 MeV, a curve of beta counting efficiency vs. sample weight can be developed. Using a weightless standard solution deposited uniformly on glass fiber filters, a curve (Figure 5) of beta counting efficiency as a function of maximum beta energy can be plotted.

For large scale processing of samples, calibration data used in data conversion calculations must be selected based on assumptions made about the sample and the nuclide composition. Typically a filter sample averages less than 10 milligrams of total solids (less than 1 mg/cm^3) and therefore it is assumed that self-absorption is negligible. The maximum beta energy for mixed fission products, as a function of time after fission, averages approximately 1 MeV at any time after two days post fission.

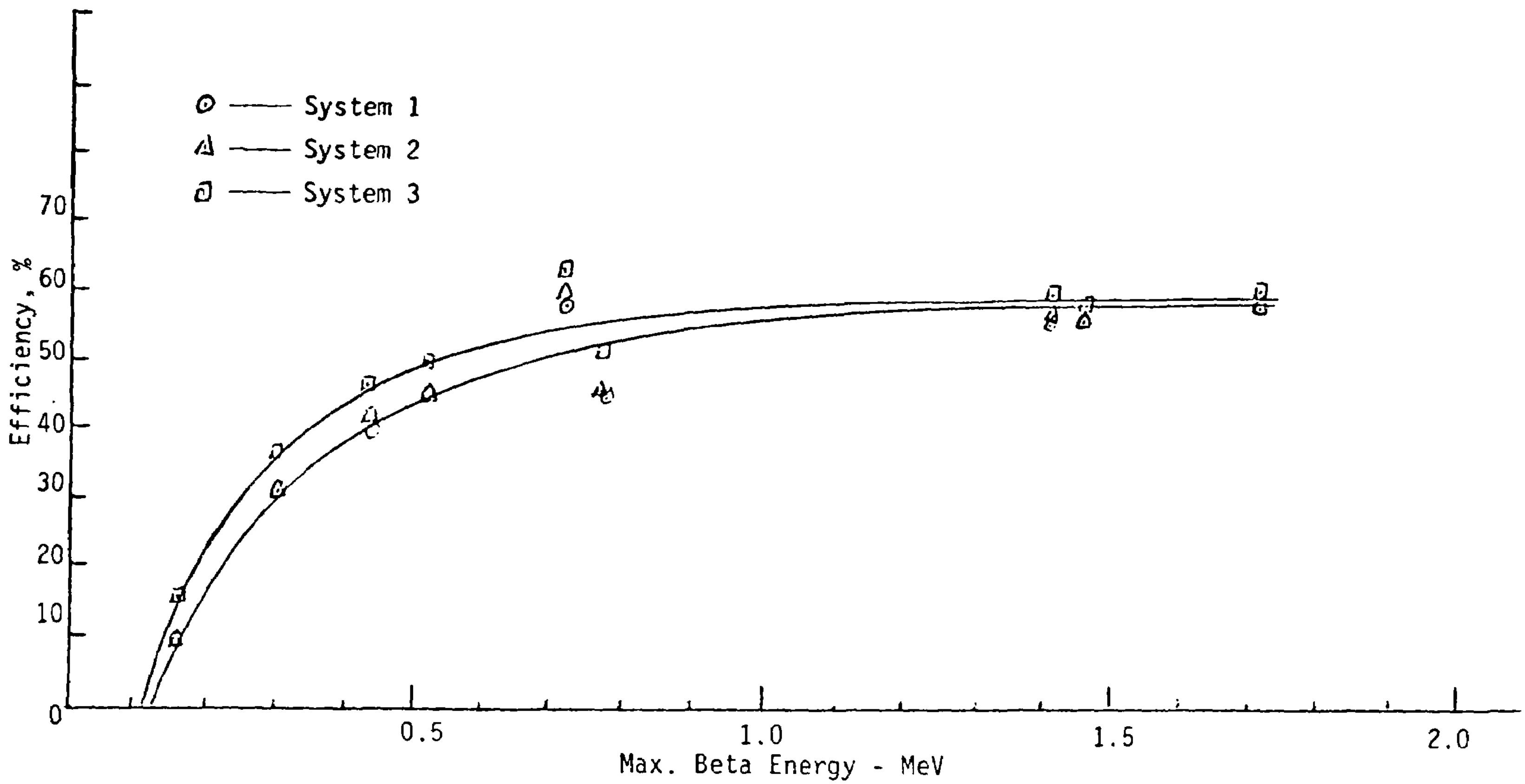


FIGURE 5
BETA CALIBRATION-COATED FILTER

A conservative efficiency value of 45% (corresponding to an average maximum beta energy of 0.5 MeV) is used for data conversion.

A daily instrumental quality control check is made on each system. This involves a 2-minute count of an alpha reference source (^{239}Pu) and a beta reference source (^{90}Sr-^{90}Y),as well as a 10-minute background count. Quality control charts are maintained on each system.

Sample Handling and Data Flow

The routine sample and data flow for air filters is shown in Figure 6. The filter is received by sample control along with its field data form. The filter is removed from the mailing envelope and put in a clean glassine envelope. Receipt of the sample is made on a posting form and obvious errors are corrected on the data form.

The collection information is keypunched in the first 24 columns of each of three-color-coded Hollerith cards. The sample along with its cards is submitted for the first beta count. The samples are stacked in the sample changer and the first count cards placed in the card puncher in the same order. The filters are counted for 2 minutes each and the count data are automatically punched onto designated fields on the card.

The first count card is checked and if the gross beta count is greater than 1000 counts, the filter is submitted for gamma scan (see Gamma Analysis System).

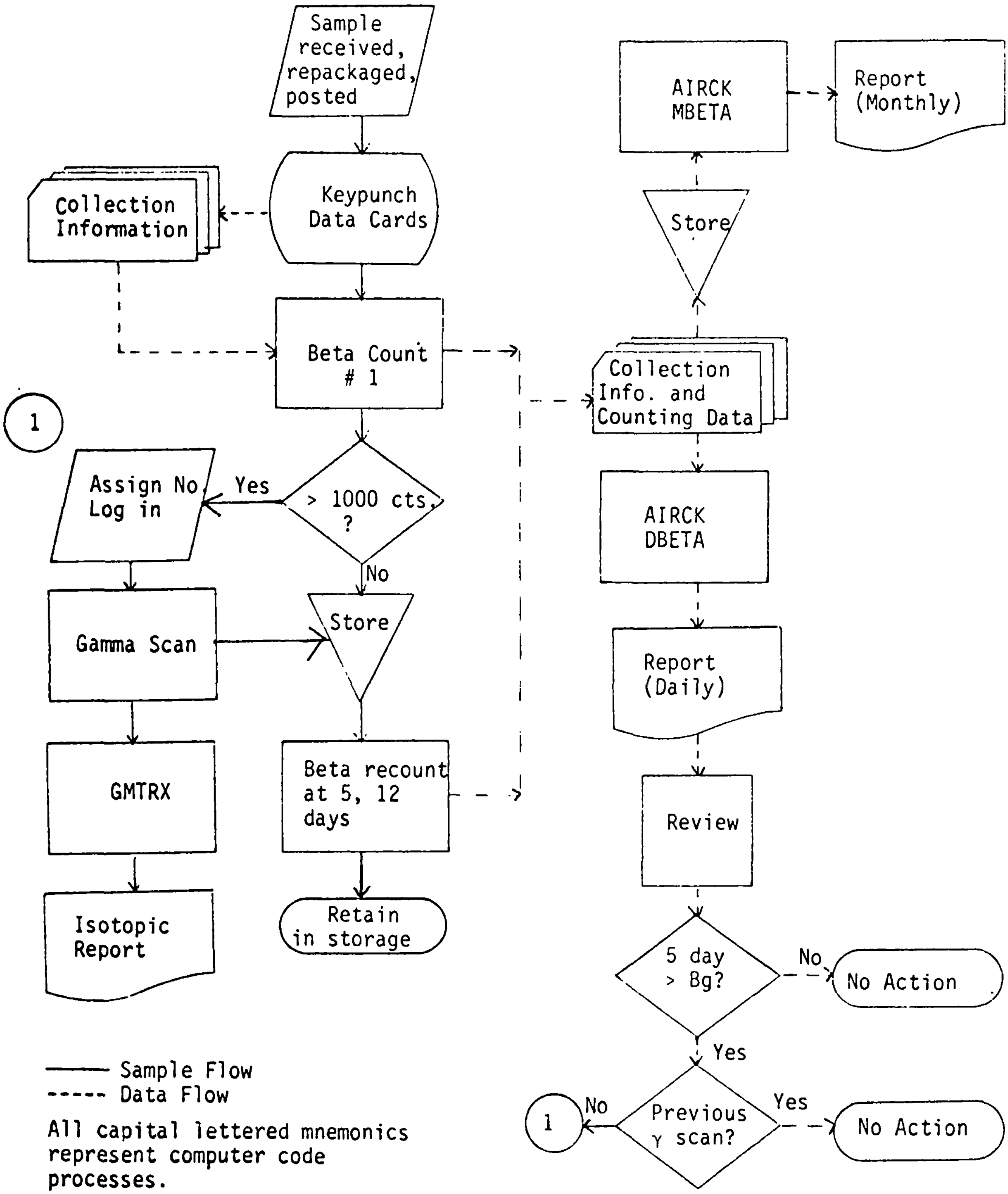


FIGURE 6
AIR FILTER, ROUTINE SAMPLE AND DATA FLOW

The filter is recounted five days after collection (after natural radon and thoron daughter products have decayed out) and at twelve days post collection. The filter may be submitted for gamma analysis if the 5-day count is unusually high.

Special (event-related) filters are handled in a similar manner with two significant changes. First, the filter is submitted for gamma analysis, and then for an initial beta count. If gamma analysis indicates that natural radioactivity is negligible, the second beta count is made at 24 hours after the first, and the third beta count at 5 days or less after collection. If natural radioactivity is prominent, the normal 5-day and 12-day beta counts are made. Second, a variety of algorithms are available for back extrapolation of beta count data to end of collection.

The individual count cards are submitted for data processing. The computer programs check for a variety of data errors, calculate the beta activity concentration at time of count, and produce a report of these values. At the end of each month the activity is back extrapolated to the end of collection and a report of extrapolated data is generated.

A variety of computer programs are utilized to analyze and report surveillance data.

System Performance

Routinely, 100 filters per day are received into the system, thus requiring 300 counts per day. As many as 300 filters per day could be handled. Turnaround from time of sample receipt to daily beta report (for first count) is routinely

24 hours. During event periods, a turnaround of 5 hours is possible for a batch of 60 samples.

Sensitivity is calculated for each individual filter. Minimum detectable activity is defined as that activity which produces a ±25% counting deviation at the 95% confidence level. For a typical routine sample, this is equal to a net activity of 50 cpm.

$$50 \text{ cpm} \times 1.00 \frac{\text{pCi}}{\text{cpm}} \times \frac{1}{350 \text{ m}^3} = 0.15 \text{ pCi/m}^3$$

Charcoal Cartridges

Routine charcoal cartridges are received by sample control and are held until 3 days post sampling before receiving a 10-minute gamma scan. The gamma spectra are processed through PONO. If the gross gamma count is equal to or greater than 300 cpm above background, the spectra are processed through GMTRX (see gamma analysis system). If the gross gamma is less than 300 cpm, a gross gamma result is produced on the card and results reported weekly. The spectra are reviewed in any case to confirm results.

Event-related cartridges are logged in, gamma scanned, and processed through the normal gamma system. Figure 7 shows the routine charcoal cartridge sample and data flow.

CHEMISTRY DATA ANALYSIS SYSTEM

General Description

Radiological counting data generated for samples that require radiochemical separation or preparation are processed by computer. Counting, calculations,

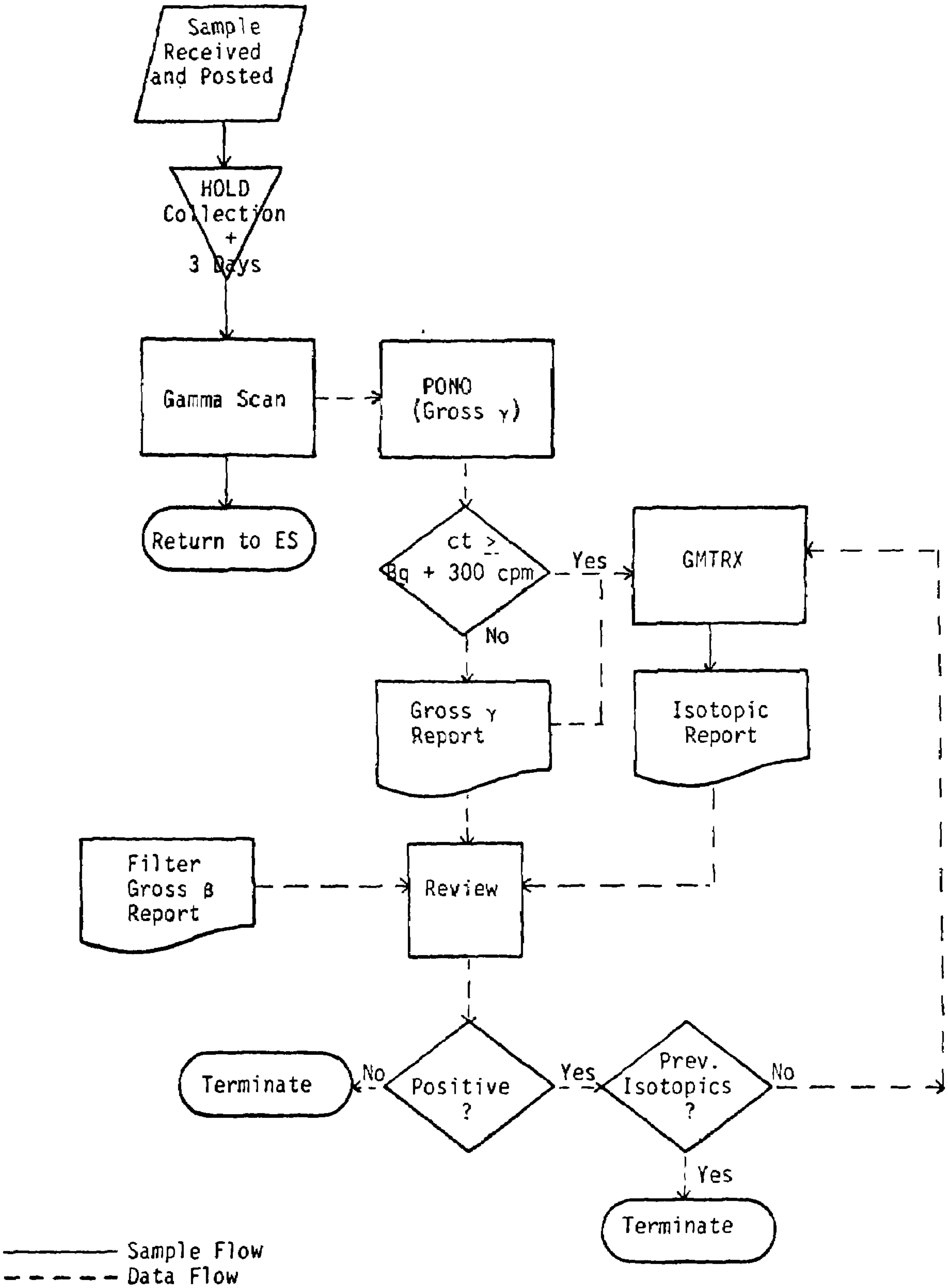


FIGURE 7
CHARCOAL CARTRIDGE ROUTINE SAMPLE AND DATA FLOW

reporting, and data storage and retrieval are described briefly in this section. Radiochemistry procedures and methods are fully described in Reference 2.

Instrumentation

Instrumentation described here is mainly for alpha and beta proportional counting of heat-dried samples and soft-beta spectroscopy of liquid scintillation solutions.

The proportional counter is a Beckman WIDEBETA II employing a 2 1/4-inch diameter detector with an 80 $\mu g/cm^2$ thin window. The gas flow system uses pure methane counting gas (99.99% pure). Background is reduced by guard detectors for cosmic radiation detection and 4-inch low-level lead shielding lined with OFAC copper. A random access automatic sample changer accommodates 100 sample planchets. Readout is by teletype printer. Three systems are in operation to accommodate heavy sample load periods.

Four Beckman LS-100 Liquid Scintillation Systems comprise the counting facility for soft beta spectroscopy. The systems operate at room temperature, accommodate 100 samples on a conveyor, and have a full three channel capacity. The systems have capability for automatic calibration (by the external standard-channels ratio method) with two separate and independent data channels for external standard counts, and with automatic subtraction of sample counts from standard counts. The output printer automatically displays data after each count including channel number and conveyor number, elapsed time, 2σ error and counts per minute.

Radon Gas analysis is described in the Southwestern Radiological Health Laboratory Handbook of Radiochemical Analytical Methods.[2] There are two separate systems utilizing the Lucas scintillation cell, an automatic and a manual system. The automatic sample changing system is basically a modified SHARP LOWBETA. The Lucas cell sets on a phototube which is coupled to a preamplifier and amplifier/discriminator for straight pulse height discrimination detection. There is no anti-coincidence circuitry. Readout is via a line printer which identifies the cell number, counting time, and the counts. The manual system is essentially the same with the exception of the sample changing mechanism. Four phototubes are incorporated in a light-tight box for simultaneous counting of four Lucas cells.

Plutonium and other alpha emitters amenable to electrodeposition are counted by alpha spectrometry. The alpha spectrometer systems consist of 8 solid state silicon surface barrier detectors, ($450 mm^2$ area and 300μ depletion depth) under vacuum, connected through suitable electronics to four multichannel analyzers. Two (2) detectors are operated in each vacuum chamber and two sets of electronics are routed to each multichannel analyzer. Voltage to the detector is provided from a bias power supply. The detector signal is routed through a preamplifier, linear amplifier and bias amplifier before being fed to the multichannel analyzer. The data are read out through a switching box to a parallel printer.

Sample counting time varies with the activity level of the sample, usually between 400 and 1400 minutes. Therefore, the weekly maximum output can vary from 40 to 100 samples.

Sample Handling and Data Flow

All samples pass through Sample Control. Those samples requiring radiochemical separation or preparation are identified accordingly. These are routed through the chemistry section after completion of non-radiochemical analysis. For liquid scintillation analysis an aliquot of the sample is removed so that processing of the sample for various analyses can occur simultaneously.

Counting data generated by these systems are merged with other information and submitted for computer processing.

Data Analysis

Several computer programs are used to process data, perform calculations, and generate various reports relating to radiochemical analysis. These report data are eventually merged with other radionuclide analysis data for generating other summaries and reports as well as for storage for future reference.

A sampling of computer programs in production is listed below.

- RCHEM: ^{89}Sr and ^{90}Sr analysis
- LIQSA: ^{3}H and ^{14}C from liquid scintillation counting
- RADON: ^{222}Rn in air analysis
- GROAB: Gross alpha and beta analysis
- PLUTON: Plutonium analysis

DATA MANAGEMENT SYSTEM

Although our complete data management system is still under development, a conceptual outline is shown in Figure 8. Analysis results are entered into the system by Hollerith card and placed in an in-process file. As analysis is completed a Tape File is created which in turn updates historical tape files. Data may be retrieved at any time, sorted according to sample type, location, time of collection, analysis, program, or a number of other more specific sample classifications. A typical retrieval time for a summary listing is two hours. A number of report generators can list data in a variety of formats. A complete description of the data management system is impossible in the context of this paper.

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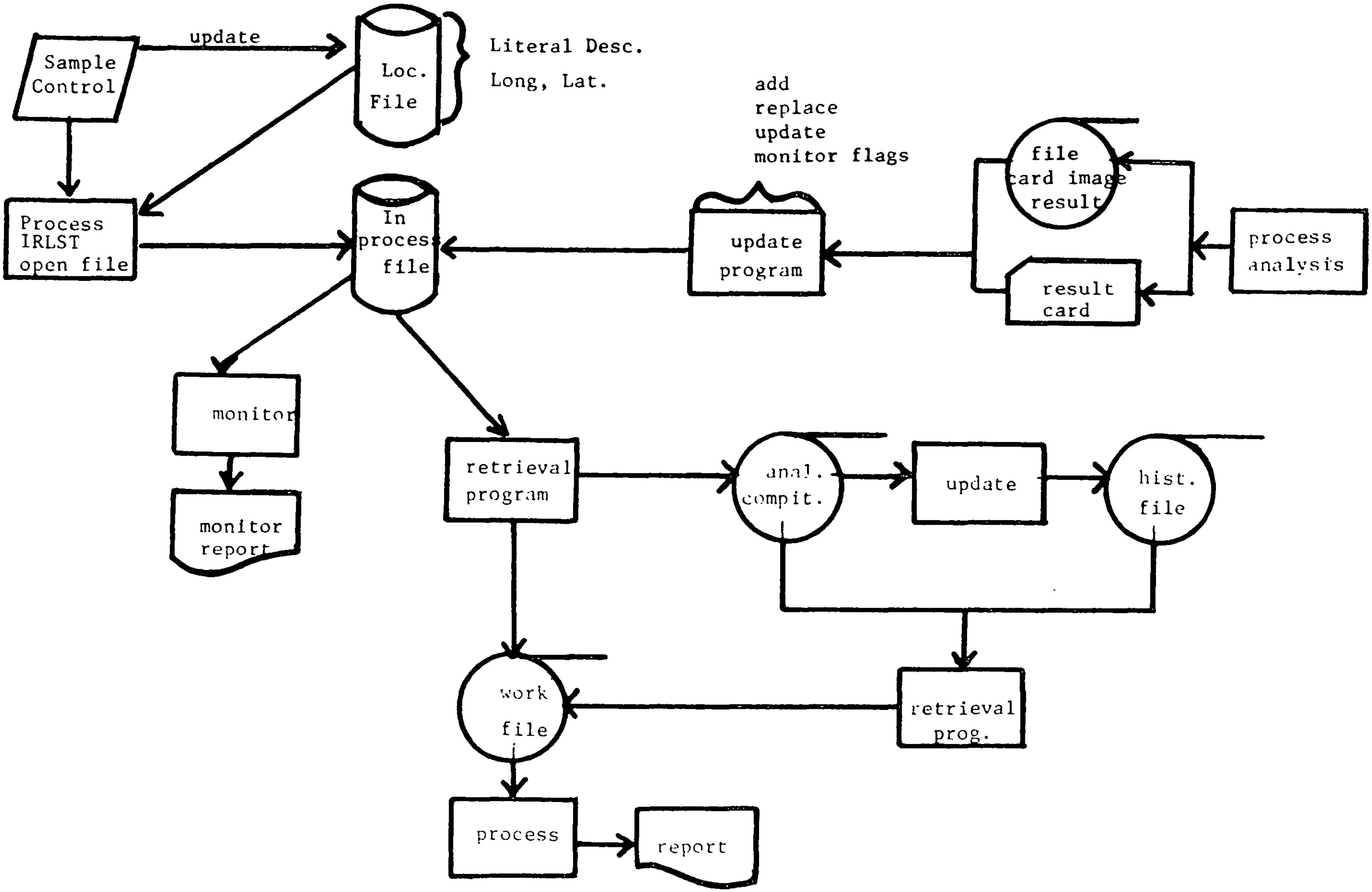
DATA MANAGEMENT SYSTEM



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