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AEC Research and Development Report

PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
SEPTEMBER 1972

DIVISION OF PRODUCTION AND
HANFORD PLANT ASSISTANCE
PROGRAMS

OCTOBER 1972

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PACIFIC NORTHWEST LABORATORY

MONTHLY ACTIVITIES REPORT

SEPTEMBER 1972

Division of Production

and

Hanford Plant Assistance

Programs

by the

Staff of Battelle-Northwest

R. S. Paul, Director

October 1972

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PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
DIVISION OF PRODUCTION AND HANFORD PLANT ASSISTANCE PROGRAMS

W. D. Richmond

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Program II

The first complete Variable Thickness Transient (VTT) test simulation was run for the entire Hanford project, covering the period March 1968 through September 1971. The agreement between computed and measured water table elevations is excellent in areas where the aquifer parameters are well defined. Considerable deviations occur, however, in some areas, such as the Gable Butte and 200-W areas.

Sensitivity analyses were conducted to determine systematic ways of adjusting various aquifer parameters. An approximate storage coefficient distribution is being computed for the Hanford project to replace the constant value assumed now.

Neutron logging was performed on the lysimeter tubes on four days to determine soil moisture trends.

Laboratory soil column measurements are being prepared for input to the transport model.

A study was continued to obtain the capability of predicting the mean activity coefficients for high ionic strength mixed salt solutions.

Program IV

Salt cake volume reduction studies continued with experiments on nitrate reduction by CO and direct distillation of NaNO_3 . In one

experiment a weak explosion, sufficiently strong to blow material out the top of the reaction vessel, occurred during CO reduction. The cause has not been determined. Distillation of mixed NaNO_3 - CsNO_3 showed a separation factor of about 5 (Cs enrichment) in the condensed material.

Process Technology

Instrumentation studies in support of the PRF included canyon monitoring and in-tank assay, dispersed phase holdup in the CC column and design for an ELW stream analyzer.

Separations plant and waste management instrumentation studies were directed toward pH probe evaluation for the waste encapsulation process use, in-line Purex stream gamma analysis and flowmeter evaluation. The pH probe specified for encapsulation plant use appears to be satisfactory in high radiation fields if standardization during use is routine.

No beneficial effects were obtained in studies on the use of flocculants to improve solids settling rates during treatment of Purex plant stored alkaline waste sludge to produce feed (PAS) for B-plant solvent extraction processing.

Further modifications to the CsCl evaporation, melt, melt transfer system were made toward improved performance and reduced cooling water requirements.

The three station triaxial accelerometer system at 221-U plant was maintained in optimum continuous recording operation.

Work continued on documents covering the final two phases of the experimental program on atmospheric release of radioactive particles.

A stack filter and a high-volume cascade impactor were calibrated in the program to characterize radioactive particles in gaseous effluents from the 234-5Z Bldg.

In wind trajectory studies, computer data processing of one year of wind data obtained from 622-R, 200-E hill and Army Loop Road sites was completed.

Equipment is being assembled for measuring wind-caused particle resuspension from the U-pond area.

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Program II - Environmental and Regulatory Technology

Radionuclide Transport Model

(D. B. Cearlock, Water Resources Systems Section)

The Variable Thickness Transient (VTT) test simulation of the period starting in March 1968 through September 1971 was completed this month. For the 2000-foot grid, about 5 hours of computer time was required. The output from the simulation consisted of monthly time planes at the end of each of the 43 months calculated.

An evaluation of the simulation was made by comparison of the calculated with the measured groundwater contour or potential surfaces in May of 1970 and September of 1971. Hydrograph comparisons at specific wells were also made. In general, the model predicted elevations that were too low. The average difference was -1.4 ft and the root-mean-square difference was 33.7 ft on the south side of Gable Butte. The smallest difference was less than 0.01 ft on the north side of Gable Mountain near the river.

The following interpretations of the quality of the VTT simulation can be made: The agreement is excellent in the area southeast of the 200-East area. This is where the aquifer parameters are best known. The worst deviations occurred under the 200-West area, south of Gable Butte, and north of Gable Mountain. The mound under the 200-West area was too low in the model and water accumulated behind the barrier of Gable Butte. This indicates that aquifer parameters in the western part of the project are not accurate. A reduction in permeability or storage coefficient or an increase in aquifer bottom elevation or a combination of these is called for. Preliminary runs with the travel time program show seepage velocities that are much too fast in a channel extending from 200-West area to Gable Butte. This indicates a reduction in permeability or an increase in storage coefficient is necessary.

Near the project boundaries where permeability or transmissivity values had to be contoured in by hand, the quality of model performance is lower. Minor modification of the transmissivity calculation routine is necessary to enable it to compute right out to the boundaries of the region.

This first transient simulation of groundwater flow under the Hanford project indicates that some improvement in the model parameters is necessary, particularly in the western portion. Current plans are to rerun the simulation with a variable storage coefficient distribution. Perspective drawings are being made of the final VTT surface by the computer plotter.

Sensitivities were computed of the average flow rate imbalance to the various parameters at every fourth node of the VTT model 2000-ft grid. These sensitivities are functions of the period simulated and the values of the parameters at each node. A sensitivity factor was defined to provide a comparison between the various parameters. It contains subjective information about the confidence band for a given parameter. The confidence band determines the range through which the parameter probably needs to be adjusted.

An approximate storage coefficient distribution for the Hanford project is being computed, based on quantities derived from the sensitivity analysis. Adjustment of the storage coefficient is made so that the flow rate imbalance at a given node is minimized within physical constraints on the value of the storage coefficient.

Neutron logging of the lysimeter tubes was done on September 8 and 9 and again on September 25 and 27. The apparent trend in the data for a general moisture decrease is continuing. Data collection procedures are being examined to insure proper operation of the instruments and to determine the cause of discrepancies in the data, such as occurred on July 17, 1972, and August 1, 1972.

Data preparation is under way for model simulation of the laboratory measured soil columns. Results of the gamma attenuation measurements are being plotted in useful form.

Activity Coefficients in High Ion Strength Mixed Salt Solutions
(R. C. Routson, R. J. Serne, and D. A. Cochran, Water and Waste Management Section)

A study was continued to obtain the capability of predicting the mean activity coefficients for high ionic strength mixed salt solutions. Specific ion electrodes were used to experimentally determine the activity coefficients of binary salt mixtures. A calcium nitrate-sodium nitrate system was prepared at five different ionic strengths. At each ionic strength various percentages of sodium nitrate were made to cover the range 0-100% sodium. The mean activity coefficients for sodium nitrate in the mixed electrolyte were calculated using the following equation:

$$E-E_o = \frac{RT}{F} \ln \frac{(m_{Na^+}) (m_{NO_3^-}) (\gamma_{\pm NaNO_3})^2}{(m_{Na_o}) (m_{NO_{3_o}}) (\gamma_o)^2}$$

where the subscript (o) represents the pure sodium nitrate reference solution. The molalities of the reference solution m_{Na_o} and $m_{NO_{3_o}}$ and the molalities of the mixed solutions m_{Na^+} and $m_{NO_3^-}$ are known values. The activity coefficient of the pure sodium nitrate solution is γ_o . It is obtained from the table in "The Physical Chemistry of Electrolytic Solutions," by Harned and Owen. The experimental millivolt difference $E-E_o$ is measured with specific ion electrodes and $\gamma_{\pm NaNO_3}$ is calculated.

The mean activity coefficients for calcium nitrate in the mixed electrolyte were calculated using a similar equation

$$E-E_o = \frac{RT}{F} \ln \frac{(m_{Ca^{++}}) (m_{NO_3^-})^2 (\gamma_{\pm Ca NO_3})^2}{(m_{Ca_o}) (m_{NO_{3_o}})^2 (\gamma_o)^3}$$

Here the subscripts (o) refer to the pure calcium nitrate reference solution.

Experimental activity coefficients are compared to coefficients derived using the empirical equation described in the August, 1972, monthly report under the activity coefficient section.

A second salt mixture, sodium nitrate-potassium nitrate, was also studied. It was found that the potassium ion electrode was malfunctioning on all but the weakest ionic strength solutions. The manufacturers were contacted and it appears that a mislabeled ion exchange membrane or solution is the cause. New parts arrived September 25th and will be tested.

The experimental results fortify the belief that the empirical equation reported in the August report is applicable to the waste streams typical of the Hanford project. No experimental results were found in the literature for a predominantly nitrate system so the above experiments were used to verify the empirical equation. Conversely the close correlation of results also leads to confidence in the experimental technique which is the only source of activity coefficients for certain salts of interest such as calcium sulfate.

Program IV - Waste Management

Salt Cake Volume Reduction Studies

(J. L. Ryan and L. L. Burger, Applied Chemistry Section)

In the August monthly report, it was noted that the reduction of NaNO_3 by CO was negligible at 700, moderately slow at 775 and quite rapid at 850°C. Further results at 775°C indicate that the simple thermal decomposition at this temperature is very slow - less than 5% conversion to Na_2O in 2 hours. A sample of NaNO_3 held for two hours at 775°C contained 54 mole % NaNO_3 and 41 mole % NaNO_2 . After bubbling CO through the melt for 11 minutes, the melt contained 40 mole % NaNO_3 and 40 mole % NaNO_2 indicating either that the reduction of NaNO_3 to NaNO_2 occurred at about the same rate as the further reduction of NaNO_2 or that NaNO_3 was reduced directly to a non-nitrogen containing compound. In another experiment at 775°C, it was found that about 50% of the combined nitrogen was destroyed by CO in 55 minutes. Thus the reduction of NaNO_3 by carbon monoxide at 775°C is much faster than simple thermal decomposition, but it is still not rapid. A temperature of about 800°C or higher is necessary for rapid reduction by CO.

In one case at 775°C a weak explosion occurred in a NaNO_3 melt during CO reduction. The melt had been held at 775°C for two hours to reach equilibrium between NaNO_3 and NaNO_2 and CO had been passed into it at a fairly rapid rate for between 10 and 15 minutes. The explosion occurred when the gas addition tube (alumina) was moved slightly. The explosion had sufficient force to throw molten salt from the 6-inch-deep crucible to the hood ceiling about 4 feet directly above the crucible. The gas regulator pressure setting was much too low for this to have been caused by simple mechanical blowout of a plugged line. Air had been flushed out of the regulator and line before gas addition began. The reason for the explosion is not apparent. No evidence of carbon from the reaction $2\text{CO} \rightarrow \text{C} + \text{CO}_2$ was observed in the alumina gas addition tube. A possibility is that solid or liquid organic matter or possibly even water from the regulator or from the rubber tubing to the alumina tube entered the bottom of the melt. The rubber tubing appeared dry inside immediately after the explosion.

Several additional experiments on the direct distillation of sodium nitrate and nitrite were performed. A test with 80% NaNO_3 , 20% CsNO_3 showed a Cs enrichment in the deposited material (separation factor = 5). Under vacuum conditions nearly complete volatilization occurred but the product reacted with the outer silica containment tube. A carrier gas is required to remove the volatile component from the hot zone. As a carrier, NO_2 seemed to behave like argon.

Process Technology

Plutonium Reclamation Facility (PRF) Instrumentation Studies (O. H. Koski, Chemical Development Section)

Instrumentation studies in support of the PRF included analysis of the canyon monitoring design criteria, dispersed phase holdup instrumentation and preliminary ElW stream analyzer design.

Analysis of the PRF canyon monitoring system was conducted to establish the anticipated count rates for the various gamma energies characteristic of Pu-239. Attenuation in the aqueous system and through the

tank wall for Sch. 10, 40 and 80 300-series stainless steel pipe of 4, 5 and 6 inches diameter was considered. The effect of design criteria for 6-inch and 48-inch sensor-to-target distances was also considered. Preliminary results indicate that the 100, 124 and 384 keV Pu-239 gammas would be useful in in-tank analysis. Maximum useful detection distance is less than 48 inches due to the low count rates.

The instrumentation system for measuring dispersed phase holdup in the PRF CC column is 90% complete. Two transducers hydraulically attached to the column will provide information as to the region in which dispersed phase holdup is changing.

A preliminary design for a monitor for the ElW stream was made to determine the anticipated count rates for Am-241 and Pu-239 for alpha and gamma energies from 17 to 384 keV. Disregarding attenuation in the cell walls but not in the solution, the 60 keV Am-241 gamma appears to offer the most sensitive determination of Am-241 in the presence of Pu-239.

Separations Plant and Waste Management Instrumentation Studies
(O. H. Koski, Chemical Development Section)

Studies were directed toward evaluation of pH electrodes, in-line gamma analysis and flowmetering systems.

Continued irradiation of pH probes in the 3730 Bldg. cobalt-60 gamma source at exposure rates of 8×10^5 R/hr has showed that an off-set of about 1 pH unit occurred for the probe currently specified for use in the Waste Encapsulation Facility at an exposure of 2×10^8 R with the drift commencing at about 1×10^8 R. When the probe was removed from the source for a period of twenty-four hours, a drift toward the un-irradiated condition occurred. Two other combination pH probes were tested. One failed at about 2×10^8 R (glass electrode envelope burst). The other showed a markedly lower time constant as well as an offset of equal magnitude in the opposite direction to that of the specified probe.

It appears that, with the specified probe, standardization while

in service can be used to negate the offset acquired due to radiation. Erratic behavior observed during continuous monitoring was determined to be a result of the cabling used. Apparently cable leakage after irradiation became significant with respect to the high source impedance of the pH electrode.

Stream samples of Purex plant HAP, 1BP, 2BP and 2AW were used to derive gamma signatures for these streams in dilutions of 1, 5, 10 in all cases and 100 in the case of 2BP. Plutonium-239 contributions to the total at characteristic energies of 17, 39, 53, 100, 124 and 384 keV were useful in 2BP at all dilutions. For the other stream samples the higher energies appeared to be most useful at the higher dilutions.

A recently developed flowmeter which utilizes sound velocity across a pipe section is being considered for plant use. Salient advantages are an unobstructed flow, high radiation resistance and, for greater than one-inch pipe sizes, a clamp-on configuration. Disadvantages are high cost, sensitivity to gas bubbles and solids and a fixed tube size design.

Treatment of Purex Plant Stored Waste Sludge (L. A. Bray, Applied Chemistry Section)

Laboratory studies were continued toward improving procedures for treatment of Purex plant stored waste sludge to produce feed for B-plant solvent extraction processing. The base case flowsheet for this operation consists of suspending the sludge in its own waste supernatant liquid, allowing the slurry to settle, decanting the liquid, suspending the solids with added water, allowing the slurry to settle, decanting the liquid and leaching the residual solid with dilute HNO_3 . Slow settling of solids and incomplete removal of strontium during HNO_3 leaching are problems with the procedure. Laboratory tests on Tank 102A slurry were made to assess the effects of anionic and cationic flocculants on the settling rates and strontium recovery. No major improvement in settling rates was observed when the flocculants were used. A decrease in settling rates was observed when the flocculants were added during the water wash. Analytical data on the effects on strontium recovery are not yet available.

Cesium Chloride Melter Development

(L. K. Mudge, Chemical Engineering Section, and B. Norton,
Chemical Development Section)

Several modifications were made to the equipment being developed for concentration, melting and melt-casting of cesium chloride in the Waste Encapsulation Facility. These modifications are designed to adapt the equipment for in-plant installation, avoid line plugging during transfer of the melt to canisters, improve sealing the canisters to the fill head and minimize the flow of cooling water to the heaters. Total cooling water flow has been reduced from 40 to 6 GPM. A device to accept an impact wrench was attached to the capsule assembly. Vibrating the assembly with an impact wrench improved the canister-to-fill-head seals somewhat but a more positive aligning device is still being considered.

A melt transfer test run will be made as soon as the modifications are complete. Reduced pressure for the transfer will be produced by a steam jet. Cooling water discharge temperatures will be measured to determine if further flow reduction can be made.

Seismic Studies

(D. D. Tillson, Water Resources Systems Section)

The three station triaxial accelerometer system at 221-U plant was maintained in optimum continuous recording operation. There were no significant seismic events of sufficient magnitude within a close enough proximity to provide any new data. Releveling of the downhole accelerometer unit was completed. Additional electronic bias was added to the two horizontal accelerometer circuits for the downhole unit in order to eliminate any future need for physical removal of the probe should tilting occur. Two more electronic filters with a 34 Hertz cutoff were fabricated and installed in the 14-channel preamplifier stage between the accelerograph and magnetic tape recorder systems.

Repairs were completed on the two-channel Hathaway strong motion recording accelerograph and the unit restored to continuous standby-triggered record mode. All three accelerometers, downhole, surface,

and building are now fully operational with continuous record onto magnetic tape and strong motion record onto analog oscillographs. Recording range is 0.0001 g to 1.0 g at the very minimum over a frequency range of 17 Hertz down to 0.001 Hertz. Preparation of a system operation and maintenance manual continued.

The National Oceanic and Atmospheric Administration, Environmental Research Laboratory (NOAA/ERL) three-component, short period seismograph station at Gable Mountain (HAN) was maintained in continuous recording operation. In addition, satellite short period vertical seismometers were operated at Rattlesnake Mountain (RMT) and at the north end of the Yakima Firing Center (YFC) on the Saddle Mountain anticlinal structure. A three-component long period seismic system was operated continuously at 221-U plant, 200-W area. Attempts to install and operate a satellite short period vertical seismometer at Jumpoff Joe on the Horse Heaven Hills near Wallula Gap continue to be frustrated by electronic problems associated with the FM radio transmitter-receiver systems. The assistance of ARHCO/Radio Maintenance is being utilized to expedite a solution to the problem. Arrival time data from Station HAN was reported to the National Earthquake Information Center for 71 teleseismic and 43 regional seismic events that had been detected. Local seismic activity remained below normal for most of the report period. A small swarm of microearthquakes was detected on September 20 and reported by the U.S. Geological Survey to have occurred near Corfu along the Saddle Mountains. The largest microquake in the swarm appeared to be less than magnitude 2 on the USGS scale and would probably not have been felt on the surface.

Additional records of regional earthquakes for the period prior to 1961 have been added to the "Epicenter Location File." Under separate cover, a computer generated plot of the known historical macroearthquake epicenters that occurred prior to June 1972 between 42 and 52° N latitude and 110 and 126° W longitude has been forwarded to D. J. Brown, ARHCO. In addition, a revised computer printout of the earthquake epicenters listed according to chronology, distance from Hanford, and earthquake magnitude or intensity has been completed and forwarded to ARHCO.

Ten man-days of service were provided in maintaining the U.S. Geological Survey microearthquake network. Summary of the stations serviced and the corrective action taken is contained in a memo to D. J. Brown, ARHCO, September 26, 1972. No quarry explosions were monitored for origin time during the report period.

Characterization of Radioactive Particles in the 234-5Z Bldg. Gaseous Effluents

(L. C. Schwendiman and J. Mishima, Atmospheric Analysis Section)

An existing 8 in. x 10 in. filter holder was adapted for use in the stack sampling program. The flows of both the filter and a high-volume cascade impactor were calibrated. A work order was issued to modify the second sampling port on the stack.

Atmospheric Release of Radioactive Particles

(L. C. Schwendiman and J. Mishima, Atmospheric Analysis Section)

Work continued on documents covering final two phases of experimental program: "Some Experimental Measurements of Airborne Uranium (Representing Plutonium) in Transportation Accidents," and "Fractional Airborne Release of Uranium (Representing Plutonium) During the Burning of Contaminated Waste."

Gaseous Waste Assistance, Wind Trajectory Studies

(R. K. Woodruff and M. M. Orgill, Atmospheric Physics Section)

Computer data processing of one year of wind data obtained from 622-R, 200-E hill and Army Loop Road sites is completed. Analysis of the data is not complete but should be in the near future.

Particulate Resuspension Studies

(R. K. Woodruff and G. A. Sehmel, Atmospheric Physics Section)

Equipment is being assembled for measuring wind-caused particle resuspension from the U-pond area. Particle resuspension rates will be determined as a function of wind speed when the wind is blowing from an upwind sampling tower across the pond to a downwind sampling tower array.

High-volume air samplers and cascade impactors will be placed on downwind towers which are about 350 ft east of the east side of the U-pond. Five 20-ft towers and six 10-ft towers will be used at a tower spacing of 50 ft. Air samplers will be at the 1-, 3-, 6-, 10- and 20-ft elevations. The upwind samples will be obtained using a single tower west of the U-pond and west of the perimeter road. Tower erection has started.

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Environmental Evaluations

Tritium groundwater in N-Area exceeded the C.G. for off-site population drinking water. A high radioactive particle was detected near E-Area on the routine annual railroad track survey. The annual Environmental Surveillance Report for Hanford for 1971 was issued, showing calculated doses to all off-site population groups as less than 1% of appropriate guides.

Radiation Protection

A TLD reader has been relocated in the 747-A Bldg. for use in processing components of criticality dosimeters.

Radiation Standards and Engineering

Low energy dependence studies of CP-TP beta shields were completed. An emergency instrumentation inventory was completed.

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Evaluations

(J. P. Corley, Environmental Evaluations Section)

A groundwater concentration of 4×10^{-3} $\mu\text{Ci/ml}$ tritium was measured in August from a test well between the 100-N area crib and the river. Information received from DUN staff is that release of tritium in this crib is on the order of 10 to 20 Ci/day versus a release of 2 to 3 Ci/day in 1971; the increased release rate is expected to continue for the balance of the calendar year. Grab samples of river water at 100-D area have had tritium concentrations as high as 7.6×10^{-6} $\mu\text{Ci/ml}$ but continuous river samples at Richland have been consistently less than the analytical limit of 6×10^{-7} $\mu\text{Ci/ml}$. The Concentration Guide for tritium in uncontrolled areas is 3×10^{-3} $\mu\text{Ci/ml}$ of drinking water.

The annual instrument survey of the Hanford railroad track system was completed. The only contamination of significance detected was a piece of unidentified particulate material found approximately 1-1/2 miles west of the 200-E area railroad gate. Uncorrected Juno instrument readings of this particle were 7.5 Rad/hr, including 300 mR/hr at contact. Upon removal, radioanalysis showed a mixture of fission products, with Ce-Pr-144 predominating; Sr-90 analysis is not yet completed. No other material of this nature was found on the railroad track survey, in the immediate vicinity or on nearby roads.

The annual Environmental Surveillance for Hanford for 1971, BNWL-1683, with Addendum, was issued. Following the shutdown of KE Reactor, calculated radiation doses to the environmental population groups from plant operations were less than 1% of applicable guides.

Airborne particulate radioactivity showed general decreases during the month except for continued elevated beta concentration at 200-E area (with an average for the month of 8 pCi/m³) and a slight increase at the air sampling stations near Redox and the west side of 200-W area (maximum of 0.5 pCi/m³); the latter increase is attributed to the excavation and hauling of contaminated soil from the SX test farm.

Radiation Protection

(K. R. Heid, Personnel Dosimetry Section)

Efforts continue regarding development of a counter to determine plutonium lung burden by direct measurement of 17 keV x-rays. The phoswich counter still appears to be the best hope but is not yet a practical and usable system.

A TLD reader has been relocated to the 747-A Bldg. for use in processing components of criticality dosimeters.

Considerable effort has been expended to keep in line the Monroe high speed printer associated with in-vivo measurements. Correction of resin joints on crucial leads is expected to yield a functional tape deck.

At the request of AEC-Hq, through AEC-RL, information on the radiation exposure of reactor operation and maintenance personnel for the years 1968-1971 was provided.

A variety of problems were experienced with the reader for the multi-purpose thermoluminescent dosimeters. Most of these were associated with the heater and heating units. Installation of a new type light seal around the heater is in progress.

Radiation Standards and Engineering

(H. V. Larson, Radiation Standards and Engineering Section)

The energy dependence of the modified CP-TP beta shield was studied. The results indicated a poor match to the response of the TLD badge for energies below 40 keV. Two test shields have been fabricated which have an aluminum outer shell of the same thickness as the aluminum in the TLD badge. Energy dependence tests will be conducted to determine how well the experimental beta shield response matches the TLD badge response.

Pursuant to a request from the Commission for an inventory of emergency equipment, a complete inventory was taken of emergency supplies for which this Department is responsible, including materials in the Emergency Control Center and the Emergency Relocation Center.

Meteorological Services

(E. H. Phinney, Synoptic Meteorology Section)

Meteorological services, viz., weather forecasts and observations and climatological services, were provided to plant operations and management on a routine basis.

SEPTEMBER, 1972

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