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PACIFIC NORTHWEST LABORATORY
MONTHLY ACTIVITIES REPORT
NOVEMBER 1973

Division of Production and Materials
Management and Hanford Plant
Assistance Programs

Battelle

Pacific Northwest Laboratories
Richland, Washington 99352

DECEMBER 1973

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MONTHLY ACTIVITIES REPORT

NOVEMBER 1973

Division of Production and Materials Management
and
Hanford Plant Assistance
Programs

by the

Staff of Battelle-Northwest
E. L. Alpen, Director

December 1973

BATTELLE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

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

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Summary

Process Technology, Development Engineering Department

Waste Concentration and Encapsulation Support

Further waste management flowsheet development studies were aimed at combining the sulfate precipitation step for Sr-Ba separation with the hydroxide precipitation step for Sr recovery.

Process Technology, Research Department

Plutonium Trench Soil Characteristics

Plutonium trench soil chemistry studies involved the rate at which Pu(IV), added to NaNO_3 - $\text{Al}(\text{NO}_3)_3$ - $\text{Mg}(\text{NO}_3)_2$ solutions, converted to a form absorbed by soil.

Most of the plutonium particles in 216-Z-9 enclosed trench sludge were identified by x-ray diffraction as crystalline PuO_2 . An occasional noncrystalline plutonium compound also was indicated.

Tank Farm Support

Work was continued to measure soil chemical and sorption parameters for a transport model determination of the movement of radionuclides from a high-level waste tank leak.

Problems of soil interface calculation have been solved in the tank leak problem.

Prevention of Accidental Release

- Surficial Contamination

The first substantial analytical results for airborne radioactive particles in the U-Pond area and the B-C Crib showed that airborne concentrations of Cs-137 ranged from 2×10^{-15} to 3×10^{-14} $\mu\text{Ci/cc}$ downwind of the B-C Crib. At the U-Pond Cs-137 concentration ranged from 2.1×10^{-14} to 5.8×10^{-13} $\mu\text{Ci/cc}$. The Cs-137 per gram of airborne soil was from 5 to 24×10^{-5} $\mu\text{Ci/gram}$.

Characterized, contaminated soil from the B-C Controlled Area was dropped into air drawn through the 242-B Wind Tunnel at 20 mph to evaluate the potential airborne release of surface activity during planned decontamination operations. About 5 percent of the soil remained airborne during the period when the wind tunnel blower was operated.

The mass fraction of soil less than 10 μm (using sieves) in the ten 1 m^2 B-C Controlled Area plots sampled ranged from 0.003 to 0.094 percent. These low fractions are being reconciled with size distributions measured in soil dispersed in the wind tunnel.

No change in soil size distribution or the distribution of UO_2 particles in the various fractions was detected when UO_2 - soil mixture samples were aged approximately two months.

A preliminary survey was made of the routine and alert air monitoring systems and procedures for the 291-Z stack. Findings which will assist in providing an improved system were transmitted to ARHCO personnel.

The program to produce wind field plots is now operational for the CALCOMP plotter. The drain-induced shear observed on the Hanford tower has been confirmed from climatological records.

Hardware was designed and constructed for control of upwind and downwind samplers to be used in monitoring waste pond and crib areas.

- Groundwater Management Studies

The program to produce a temporal rate of change map of the Hanford groundwater system is being implemented. The data has been accumulated over the past three months.

Error and sensitivity analyses have been run on the routines used to develop a transmissivity map. Significant error reductions have been achieved by implementing new techniques.

Temperature logging of the Hanford wells has begun.

New calculations in the PST program were made to handle soil interfaces in a more accurate way.

- Biological Transport Studies

Studies were continued on the effects of plant age, chemical form and depth of placement on plant uptake of transuranium elements from soil. A transverse ditch was excavated across the decontaminated dry waste trench 216-T13 to observe rooting patterns of field grown tumbleweeds.

The experimental goldfish populations enclosed over areas of relatively high and low radioactivity in Gable Mountain sediments and plants have both essentially attained equilibrium in activity uptake.

ASSISTANCE TO ATLANTIC RICHFIELD HANFORD COMPANY

Process Technology, Development Engineering Department

Waste Concentration and Encapsulation Support

Strontium - Barium Separation Flowsheet Development
(L. A. Bray, Applied Chemistry Section)

In Hanford waste management studies, a Sr-Ba separation process was developed previously which uses small amounts of sulfate to selectively precipitate Ba while leaving Sr in solution. Additional tests have now been completed aimed at combining Ba removal with the caustic strike Sr process.

Barium removal and Sr recovery were determined for several runs in which various amounts of Na_2SO_4 were added to the Sr feed at various pH stages during the NaOH precipitation step. Similar Na_2SO_4 addition tests were run on feed which had been previously adjusted with NaOH to a pH of 10. With initial feed containing 0.01M Ba and 0.25M Sr, the results indicate that, (1) a sulfate concentration of 0.01 to 0.02M will provide a Ba DF of from 3 to 10; (2) the best results were obtained when the sulfate was added to the acidic solution before it was adjusted to pH 10 or during the first hour of digestion after it was adjusted to pH 10; and, (3) that the Ba DF depends on the solution pH at the time of sulfate addition (DF_{Ba} at pH 0 greater than at pH 10). The results also indicated a 95 percent and an 88 percent Sr recovery for 0.01 and 0.02M sulfate, respectively. Two-thirds of this strontium loss is associated with the hydroxide precipitation; subsequent washing of the precipitate will recover at least half of the loss. Future laboratory tests will attempt to develop a method of recovering most of the remaining Sr from the washed solids for recycle to the solvent extraction facility.

Process Technology, Research DepartmentPlutonium Trench Soil CharacteristicsPlutonium Trench Soil Chemistry and Mineralogy

(J. L. Swanson, Chemical Development Section and
L. L. Ames, Land Resources Systems Section)

In high-salt solutions typical of those charged to the 216-Z-9 enclosed trench, the formation of Pu species which are readily sorbed by soil (presumably Pu polymer) was found to vary with pH in a somewhat surprising manner. In $2.4M$ $NaNO_3$ - $0.6M$ $Al(NO_3)_3$ - $0.3M$ $Mg(NO_3)_2$ solutions, addition of 0.06 mg/ml Pu(IV) to the solution adjusted to pH 1.6 resulted in slow conversion of the Pu to a form that was sorbed by soil; about one week (at $23^\circ C$) was required for half of the Pu to convert to a sorbable form. In a similar experiment at pH 1.9 the conversion to a sorbable form was more rapid; only about one day was required for half of the Pu to convert to a sorbable form. At pH 2.5, however, little if any of the Pu was converted to a sorbable form in one day. The sorption contacts employed 0.1 g soil per ml of solution and a five minute contact time.

The rate of formation of the sorbable Pu is highly dependent on the concentration on non-sorbable Pu. In the experiments described above, at a given pH the rate of formation of sorbable Pu varied with at least the third power of the concentration of non-sorbable Pu remaining.

Five plutonium particles were located and removed from the sludge overlying core 4-11 taken from the 216-Z-9 trench. The particles, ranging in diameter from 10 to 100 micrometers, were mounted on fine glass rods for x-ray examination. X-Ray diffraction patterns of four of the five particles were of PuO_2 , varying in quality from good to excellent. The fifth particle, contained 84 wt% Pu, but gave no x-ray diffraction pattern, indicating noncrystallinity. Thus, a portion of the plutonium particles could be present as plutonium polymer or hydroxide rather than crystalline PuO_2 . The x-ray examination of more particles is underway to confirm the presence of noncrystalline plutonium compounds and to determine the crystalline to noncrystalline ratio, if possible.

the soil was dropped into a 20 mph airstream in the wind tunnel. Further work will be undertaken to define the fraction of particles in the respirable size range which may become airborne.

Aliquots of the uranium dioxide powder-soil mixture were allowed to age several months in shallow trays and deep cylindrical containers. One of the aliquots in the shallow tray was periodically sprinkled with water and dried on a hot plate to simulate crudely the effects of precipitation and drying. No significant displacement of the uranium appears to occur due to this aging process--neither the distribution of activity in the soil fractions, nor the activity at various depths appears affected.

Characterization of Radioactive Particles and Assessment of Sampling Systems 234-5Z Plant

(J. Mishima and L. C. Schwendiman, Particulate and Gaseous Waste Research Section)

An initial survey of the sampling system, emission calculations, and procedures for the routine and the off-standard release alert air monitoring system for the 291-Z stack was performed. Improvements which could be made to the system were identified.

Particulate Resuspension Studies

(G. A. Sehmel and L. C. Schwendiman, Particulate and Gaseous Waste Research Section)

Preliminary generalizations from resuspension experiments at B-C and U Areas can now be made even though additional analytical results are still awaited. Airborne Cs-137 concentrations at U-Area are greater than at B-C Area. This increase could be caused by shorter concentration averaging times, higher wind speeds, or more readily resuspended source. All calculated concentrations have been orders of magnitude lower than an MPC of 2×10^{-8} $\mu\text{Ci Cs-137}/\text{cm}^3$ for continuous exposure.

Airborne measurements from August 30, 1973 to September 14, 1973 at B-C Area show that: (1) Ru-103 concentrations were from 1.8 to 2.4×10^{-15} $\mu\text{Ci}/\text{cm}^3$ in air and 3.6 to 5.6×10^{-5} $\mu\text{Ci}/\text{g}$ airborne soil, and (2) Cs-137 concentrations were from 0.19 to 3.0×10^{-14} $\mu\text{Ci}/\text{cm}^3$ and 0.45 to 2.4×10^{-4} $\mu\text{Ci}/\text{g}$ airborne soil. During the early part of that time period, a road had been constructed across B-C area which passed between sampling towers 5 and 6. The highest

observed Cs-137 concentration was at tower 6 which indicates increased Cs-137 resuspension caused by the road construction. In contrast to the Cs-137, Ru-103 concentration did not show any increased airborne concentration at tower 6.

Daily airborne Cs-137 concentrations at U Area from July through October (same two filter locations changed each day) ranged from 0.15 to 55×10^{-14} $\mu\text{Ci}/\text{cm}^3$. A few samples showed no detectable activity. In the first U Area resuspension experiment from February 6 to April 6, 1973 for 123 sampling hours during predicted higher wind speed conditions, the airborne Cs-137 concentrations ranged from 2.1 to 58×10^{-14} $\mu\text{Ci}/\text{cm}^3$. Airborne Cs-137 concentrations appear to be somewhat uniform as a function of sampling height up to 20 feet. Additional samplers at greater heights are needed to determine the upper heights of the airborne Cs-137 plume.

Equipment installation is continuing for simultaneously automatic sampling with both upwind and downwind towers.

Wind Trajectory Studies

(R. K. Woodruff, L. L. Wendell, W. F. Sandusky and M. M. Orgill,
Atmospheric Sciences Department)

The first computer plot of measured wind data and the interpolated wind field for the grid covering the Hanford site was produced during this reporting period. The plot was generated on a CALCOMP plotter. A section has been designed for the program which allows convenient scaling and placement of the plots on a page. This will allow eight three-hourly (one day's data) wind field plots to be presented on two pages, which allows more rapid visual assimilation of the data than one per page. The program should be fairly easily converted to generate microfilm plots on the new equipment acquired by CSC. The trajectory plot program is in the checkout phase and should soon be producing plots of wind-field derived transport over the Hanford site.

No constant volume balloon tracking flights were made in November because of the unusual amount of poor flying weather during the month. However, the ballasting equation was rederived to eliminate the need for pressure measurements. Now the amount of ballast needed for the balloon can be determined with temperature measurements only at the launch point and flight level.

The indication of a conflict in the drainage flows at the Hanford tower was confirmed through an examination of a previous climatological summary comparing the 50 and 400 foot wind direction. There is a detectable correlation between the southerly wind at 50 feet and the 500 foot west to northwest wind. Since this is the case when all wind observations are considered, it should be enhanced if the same comparison was made for the early morning hours only.

In order to model the drainage potential quantitatively, the terrain heights must be read onto a grid for numerical analysis. During this reporting period a plastic overlay containing an appropriate grid was produced. The grid and a topographic map may now be used to read the heights.

Inquiries have been made to Thiokol Chemical Corporation about telemetry systems they have developed. An attempt is being made to determine the most appropriate updating scheme for the existing telemetry system which has become expensive to maintain and is proving unreliable.

Instrumentation Development for Meteorological and Environmental Surveillance
(O. H. Koski, Chemical Development Section)

Design and construction of hardware adaptable to existing equipment to be used in surveillance of waste ponds and crib areas was pursued. Simultaneous control of upwind and downwind samplers used in crib and pond surveillance was desired. The controls are to be adaptable to present equipment and to provide manual or automatic selection of from one to four upstream samplers. Isolation of the control signals from the supply power and the use of low voltage control signals were employed to avoid problems associated with the long run of signal cable required (about one-half mile) and the use of separate power sources on the transmitting and receiving end.

Preliminary tests of a zinc sulfide-coated Maylar sheet sensor demonstrated potential use as an alpha detector in the assay of air sampler filters.

- Groundwater Management Studies

Generation of Improved Transmissivity Distribution

(K. L. Kipp and D. R. Friedrichs, Land Resources Systems Section)

The five sets of water level measurements were entered into the data file. A program to do the temporal rate of change calculation is being implemented. Platforms are being constructed by the shop for the water level recorders. These will be installed on selected wells to assist in defining transient changes in the water table.

Error and sensitivity analyses were run on the transmissivity Iterative Routine to examine the accuracy of the flow tube calculations. The parameters that were shown to affect the accuracy were the gradients, stepsize, width adjustments, angle of deviation, and computer floating point calculation inaccuracy. Analyses of these parameters were made using steady state and transient surfaces. Applying new techniques significantly reduced the errors.

Applying new techniques and more accurate potential storage, the new Iterative Sequence when applied to a surface having characteristics similar to the Hanford groundwater system has an error range of 2 to 20 percent. The Hanford system contains few areas where 20 percent error would be experienced. Formerly errors ranged up to 170 percent in the gradients.

Field Measurement and Monitoring Assistance

(K. L. Kipp, Land Resources Systems Section)

Temperature logging of the project wells has been started as the result of a reported temperature anomaly in one of the WPPSS #2 site wells.

Moisture content of the soil in the lysimeter was monitored with the neutron probe on November 9 and again during the week of November 26 to 30. This may be the wettest fall on record and may add significant moisture to the vadose zone.

Model Development and Application

(S. W. Ahlstrom, A. E. Reisenauer and D. R. Friedrichs,
Land and Resources Systems Section)

Significant improvements in the computational method of Macro-Micro Ion Transport Program method of calculations have been recognized and are being

implemented. Significant time saving should be realized when the improvements are complete.

Tracer Dye Test 216-U-Pond

(H. P. Foote and S. B. Ailes, Land Resources Systems Section)

The dye tracer study on the 216-U-Pond was delayed by weather and recent changes in operations.

● Biological Transport Studies

Plant-Soil Interaction

(K. R. Price, Environmental Chemistry Section)

Studies investigating the effects of plant age, chemical form, and depth of placement on plant uptake of transuranium elements from soil are continuing. Thirteen week old tumbleweed and cheatgrass plants had about the same concentrations of Pu-239 and Am-241 per gram of tissue as ten week old plants, but the total amount removed from the soil increased in proportion to increased dry matter production. Americium uptake continued to be much greater than Pu. Sixteen week old plants are being analyzed which will complete the series.

Tumbleweed and cheatgrass plants were grown on soil treated with Pu-239 polymer and harvested after two months. Analyses are in progress. Additional plants grown for two months in large pots treated at various depths with organic acid complexes of Np-237 and Cm-244 were harvested and are being analyzed.

The decontaminated dry waste trench 216-T13 was partially excavated by digging a transverse ditch in order to observe the rooting pattern of field grown tumbleweeds. The trench originally was about 2 m deep. Roots from medium sized tumbleweeds were noted to extend to a maximum of 2.8 m. More roots occurred in the backfilled material than in adjacent undisturbed soil. Additional excavations are planned.

Aquatic Studies at Gable Mountain Pond

(D. G. Watson and C. E. Cushing, Freshwater Ecology Section)

The experimental goldfish populations enclosed over areas of relatively high and low radioactivity in the sediments and plants have both essentially attained equilibrium. The fish placed over the area with higher radioactivity levels attained about two times as much Cs-137 as those over areas of lower radioactivity; Zr-95 and Co-60 levels were similar at both sites.

Radionuclide concentrations in ducks restricted to Gable Mountain Pond exhibited increasing concentrations of Cs-137 in the muscle and carcass, whereas levels of Zr-95 and Co-60 remained unchanged.

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Summary

Environmental Evaluations

Increased tritium and nitrate concentrations were noted in the groundwater pumped at the WPN-2 site in the month of November.

Radiation Protection

Phase two of the computer program to automate the processing of the in-vivo measurements is now complete and the program is being used regularly to process the magnetic tape of in-vivo count spectra.

Radiation Standards and Engineering

Problems associated with one Hanford contractor's stack monitors have been identified and necessary repair undertaken.

TECHNICAL ASSISTANCE TO THE HANFORD PLANT

Environmental Evaluations

(J. P. Corley, Environmental Evaluations Section)

A temperature increase was reported by construction personnel in water being pumped from the unconfined aquifer at the WPN-2 site. Although an increased temperature could not be confirmed, special sampling of all the wells at the WPN-2 site at the request of WPPSS showed further increases in both tritium and nitrate ion concentrations since the last sampling in May 1973. The maximum tritium concentration measured was 1.6×10^{-4} $\mu\text{Ci/ml}$, or 5 percent of the Concentration Guide for uncontrolled areas, and the maximum nitrate ion concentration was 13 ppm, compared to the PHS Drinking Water Standard of 45 ppm. Consideration of data over the past year from these and nearby test wells indicates that a lateral extension of the plume of groundwater contamination to the south has occurred. Concentrations are not expected to increase in the near future and may decrease with decreased groundwater usage.

Two barely detectable increases in river radioactivity, each lasting for several hours, were detected during the month by the Automatic Columbia River Monitor at the 300 Area. The observed increases of approximately 1-2 $\mu\text{R/hr}$ above the water surface would not be a significant contribution to annual environmental doses. Radioanalysis of river water samples at Richland showed no unusual radioactivity, and questions of contractor staff failed to reveal a source.

All other monitoring measurements were within normal ranges, with the road monitoring indicating no detectable contamination on Hanford highways.

Radiation Protection

(H. V. Larson, Personnel Dosimetry Section)

Phase two of the computer program to automate the processing of the in-vivo measurements is now complete and a program is being used regularly to process the magnetic tape of in-vivo count spectra. Personnel at the

in-vivo counter facility are entering in-vivo identification and examination data for the spectra on the magnetic tape on a regular basis now using the phase I program.

Consideration is being given to building a new lung phantom to more nearly duplicate conditions than with the present lung phantom. We have had splendid cooperation with Dave Waite and Larry Anderson of Livermore and Phil Dean of Los Alamos to date in this effort. Improvements considered are efforts to simulate the source detector geometry, the chest wall thickness, and the transmission through intercostal cartilage as well as bone marrow.

Radiation Standards and Engineering

(J. M. Selby, Radiation Standards and Engineering Section)

Problems associated with all of the stack monitors (four) used by one Hanford contractor are being investigated. The primary problems have been traced to defective photomultiplier tubes and preamplifiers. Performance of four preamplifiers plus three spares was examined in detail. All but one were found to be defective. All of the preamplifiers have been repaired and one instrument is completely operational.

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.

November 1973

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