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17

**Part 2: Environmental Sciences
April 1989**

**Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830**

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Staff Members
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Pacific Northwest Laboratory
Richland, Washington 99352

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PREFACE

This 1988 Annual Report from Pacific Northwest Laboratory (PNL) to the U.S. Department of Energy (DOE) describes research in environment, safety, and health conducted during fiscal year 1988. The report again consists of five parts, each in a separate volume.

The five parts of the report are oriented to particular segments of the PNL program. Parts 1 through 4 report on research performed for the DOE Office of Health and Environmental Research in the Office of Energy Research. Part 5 reports progress on all research performed for the Assistant Secretary for Environment, Safety and Health. In some instances, the volumes report on research funded by other DOE components or by other governmental entities under interagency agreements. Each part consists of project reports authored by scientists from several PNL research departments, reflecting the multidisciplinary nature of the research effort.

The parts of the 1988 Annual Report are:

Part 1: Biomedical Sciences	
Program Manager: J. F. Park	D. L. Felton, Report Coordinator and Editor
Part 2: Environmental Sciences	
Program Manager: R. E. Wildung	S. G. Weiss, Report Coordinator and Editor
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Program Managers: L. G. Faust
W. T. Pennell
J. M. Selby	L. G. Faust and W. T. Pennell, Report Coordinators
S. K. Ennor, Editor

Activities of the scientists whose work is described in this annual report are broader in scope than the articles indicate. PNL staff have responded to numerous requests from DOE during the year for planning, for service on various task groups, and for special assistance.

Credit for this Annual Report goes to the many scientists who performed the research and wrote the individual project reports, to the program managers who directed the research and coordinated the technical progress reports, to the editors who edited the individual project reports and assembled the five parts, and to Ray Baalman, editor in chief, who directed the total effort.

Members of the Scientific Advisory Committee, established in 1985, are:

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FOREWORD

This report summarizes progress in environmental sciences research conducted by Pacific Northwest Laboratory (PNL) for the Office of Health and Environmental Research in FY 1988. Research is directed toward developing a fundamental understanding of processes controlling the long-term fate and biological effects of fugitive chemicals and other stressors resulting from energy development.

The PNL research program continues to make important contributions to the resolution of important national environmental problems. The research, focused principally on subsurface contaminant transport and detection and management of human-induced changes in biological systems, forms the basis for defining and quantifying processes that affect humans and the environment at the regional and global levels. Each research project forms a component in an integrated laboratory-intermediate scale field approach designed to examine multiple phenomena at increasing levels of complexity. This approach is providing system-level insights into critical environmental processes. Strong university liaisons now in existence are being markedly expanded so that PNL resources and the specialized technical capabilities in the university community can be more efficiently integrated.

Building on PNL technical strengths in geochemistry, environmental microbiology, hydrodynamics, and statistical ecology, research in the environmental sciences is in an exciting phase, and new investments have been made in molecular sciences, chemistry, biotechnology, use of remote imagery, and theoretical ecology. The section on exploratory research provides unique insight into the value of these investments and into the future of PNL environmental sciences programs.

Raymond E. Wildung
Program Manager
Environmental Sciences

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Detecting and Managing Change in Terrestrial Ecosystems

Detecting and Managing Change in Terrestrial Ecosystems

More accurate predictions of terrestrial system responses to natural and energy-related disturbances require a better understanding of how ecological, hydrologic, biogeochemical, and atmospheric processes interact to influence long-term changes and how spatial scales influence environmental responses. Only recently has technology become available to make simultaneous measurements of these processes. In addition, numerical models and knowledge-based systems are being developed to interface newly acquired data sets. Such techniques and instruments are significantly increasing the ability to predict the response of terrestrial systems in ways and over time and space scales not previously possible, and are the focus of this research.

During FY 1988, a high priority was placed on initiating research that integrates basic knowledge about major land processes with ecological theory and advanced computational and analytical capabilities. These studies are being conducted on several scales (laboratory, microcosm, field, and landscape) and are designed to provide a new approach for detecting and evaluating environmental changes.

Hanford's National Environmental Research Park (NERP) provides the long-term protection, continuity, and organizational structure necessary for the successful completion of these studies; it also provides the necessary institutional structure for faculty and student participation from the academic community.

Understanding Terrestrial Ecosystems

The research described here focuses on understanding and predicting the response of terrestrial systems to environmental stress. The field studies are primarily focused on landscapes in the NERP, which has served as a field laboratory for long-term environmental research since its establishment in 1975.

Since the research park was formed, substantial gains have been made in developing computer analytical capabilities, remote sensing technologies, and environmental sampling methodologies. However, these technological advancements have not been well integrated with ecological theory in terms of providing advanced interpretation and prediction of changes within the environment.

Within the next 5 years it should be possible to continuously monitor and display the environmental status of the NERPs and their surrounding lands, and thus, by extrapolation, monitor the worldwide environmental status of large-scale environmental changes.

Figure 1 illustrates the scope of the research park program. Part A, Basic Environmental Studies, focuses on interrelated research in biotic, hydrological, biogeochemical, and atmospheric processes. These studies are concerned with abiotic/biotic linkages, control mechanics, site-specific data sets, and process models. Part B,

Ecological Theory, concerns developing and refining the necessary theoretical base for understanding ecological efficiency, scaling (remotely based sensors), spatial temporal relationships, system steady states, and system linkages and controls. Analysis and Predictions, Part C of the program, focuses on sampling designs, integrating data, developing geographical information system (GIS) models, knowledge-based systems, and quantitative change detection systems.

These three parts of the program are being integrated to provide Part D, a Landscape Response Surface (Figure 2), which is based on energy, water, and mineral efficiency relationships. The theory is that the shape of the response surface will change as a result of environmental stress or mitigative activities in productivity, air and water quality, and contaminant and/or nutrient and sediment transport.

When fully functional, this response surface system will be valuable to the U.S. Department of Energy (DOE) and other government agencies by providing advanced testing methods for contaminant cleanup scenarios and predicting responses to all forms of natural and human-related stress (e.g., CO₂ increase, drought, wildfire, waste storage, soil disturbance, pest outbreaks). All these scenarios can be related to energy, water, and mineral use efficiency. The status of these studies is described in the following subsections.

FIGURE 1. Program Overview for the National Environmental Research Park, Showing the Integration of Research, Theory, Interpretation, and Prediction

FIGURE 2. A Postulated Landscape Response Surface for National Environmental Research Parks, Based on Energy, Water, and Mineral Efficiency Relationships

Arid Lands Water Balance

G. W. Gee, J. S. Depner, and S. O. Link

Arid lands water balance has been the subject of several recent reviews (e.g., Gee and Hillel 1988; Gee et al. 1988), initiated in part by a growing interest in selecting waste disposal sites in areas that have low rainfall and deep unsaturated soils. There is a need to understand the processes that

govern the water balance in these areas and to obtain data that can be used to test models for predicting recharge and quantifying the transport of contaminants into underlying groundwater reserves. Plants largely control the fate of water that falls on arid lands through their contribution to evapotranspiration (combined water loss to the atmosphere from soil and plant surfaces). Because evapotranspiration depends upon plant characteristics, disturbances that alter plant communities can affect evapotranspiration, and thus alter the water balance and change (increase or decrease) rates of recharge at a given site. Little work has been done to evaluate the effects of changes in native plant communities on water balance of arid or semiarid sites.

Studies have been conducted at the Watershed Research Facility, located on the Arid Lands Ecology (ALE) Reserve, to evaluate the water balance of distinctly different plant communities growing on similar soils under similar climatic conditions to better understand how water is cycled by plants in arid environments. Three sites, two dominated by grass cover (bunchgrass, *Agropyron spicatum*) and one covered with a mixed grass and shrub (sagebrush, *Artemisia tridentata*) plant community growing on fine-textured silt loams (Xerollic Camborthids), have been monitored for soil water storage (by neutron probe logging) for the last 4 years. Plant biomass and leaf area data from these sites have also been collected annually (Link and

Thiede 1987). In late 1986, pairs of precision weighing lysimeters were installed at the mixed grass and shrub site and one of the grass-dominated sites (Gee 1987). The lysimeters are used for detailed measurement of the daily water balance (i.e., precipitation, evapotranspiration, water storage, and drainage) of the two plant communities. The annual precipitation at the test sites has been above and below the average of 22 cm/yr (Thorp and Hinds 1977) and has varied by more than 50% during the last 4 years. Data collected from the neutron probes and the lysimeters have shown that water available for plants was used as well by the bunchgrass community as by the mixed grass and shrub (bunchgrass and sagebrush) community. The water extracted has changed little for each plant community throughout the year over the 4 years of testing, with the water storage in the top 1.25 m of soil being reduced to similar values (11 ± 2 cm) for both the grass and the mixed grass and shrub community. Anderson et al. (1987) have observed similar water balance controls by monocultures of grasses and shrubs in fine soils under semiarid climate conditions in Idaho.

A water balance model was tested on data collected from the bunchgrass lysimeters at the Watershed Research Facility. The UNSAT-H (Fayer et al. 1986) model, an unsaturated water

flow model that includes plant transpiration to simulate daily water balance (i.e., infiltration, evapotranspiration, redistribution, drainage), was used for a 1-year period (July 1, 1987 - June 30, 1988); model results were compared with data collected from the bunchgrass-covered lysimeters for the same period. Simulations were run to test the effect of precipitation variation on soil water balance and also to test the adequacy of UNSAT-H to predict annual transpiration losses from grass cover at the ALE site.

Figure 1 shows the results of model simulations of daily soil water storage changes for bunchgrass under two precipitation regimes (16 cm/yr and 33 cm/yr) compared with lysimeter measurements. The model, based on transpiration data collected at ALE for cheatgrass, *Bromus tectorum* (Hinds 1975), was modified to simulate transpiration for a 250-day period (September through May), after which it was assumed for the month of June that soil evaporation was the only process controlling water loss from the vegetated surface (Fayer 1987). The model also extended roots to depths as great as 150 cm to optimize water extraction. The water extraction function of Feddes et al. (1978) was used to simulate time-dependent water extraction from the soil profile.

FIGURE 1. Daily Water Storage for Bunchgrass with Simulated (16 cm/yr and 33 cm/yr) and Actual (Lysimeter) Precipitation

The model reasonably predicted the late fall water losses for 1987, but significantly underpredicted bunchgrass evapotranspiration occurring in the spring and early summer (April through June) of 1988. The model predicted that there would be annual water storage increases ranging from 1 to 8 cm of water for the two simulated climates, while the data show that there was an actual loss of storage of about 4 cm of water, and that the bunchgrass was effective in recycling the annual precipitation (27 cm) to the atmosphere. The data indicate that bunchgrass evapotranspiration continued actively into July. Clearly there is a need to modify the transpiration algorithm to more accurately account for the apparent difference in plant parameters between cheatgrass (as modified in the model) and bunchgrass (as measured in the lysimeters). Efforts have been undertaken to study these differences and thus incorporate the observations into an improved transpiration algorithm for bunchgrass.

Ecophysiological observations (i.e., leaf area index, plant xylem pressure, stomatal conductance, leaf transpiration) were made on lysimeters and adjacent areas in July 1988. Preliminary testing in July 1988 showed that xylem pressures were similar in plants on and off the lysimeters, but that stomatal conductances were lower on plants on the lysimeter than on adjacent plants. Ecophysiological measurements taken periodically throughout the year on lysimeters will provide additional data with which to test model performance. Such efforts are planned for the coming year. In addition, the water balance of the sagebrush lysimeters will be simulated.

In summary, both bunchgrass and sagebrush/bunchgrass plant communities were found to effectively exploit the annual precipitation at the ALE watershed test sites by removing the available soil water annually for the last 4 years. This evapotranspiration occurred under conditions where the annual precipitation has varied by more than 50% above and below the normal (22 cm/yr). The lysimeters appeared to adequately measure the daily water balance during the last year and continue to provide data that can be used to calibrate unsaturated water flow and water balance models under semiarid site conditions. Detailed ecophysiological measurements on and off the lysimeters throughout the year will aid in assessing the use of the lysimeter data for predicting water balance of grass and shrub plant communities.

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The Effect of Water and Nitrogen Perturbations on Gas Exchange in Arid Lands Ecosystems

S. O. Link, P. A. Beedlow, M. E. Thiede, and J. L. Downs

The availability of limited nitrogen and water resources in arid terrestrial ecosystems strongly influences nutrient dynamics, biogeochemical cycling processes, and productivity. Human activities that result in the release of nitrogen-containing compounds to the atmosphere or that modify climate have the potential for causing large-scale changes in plant productivity. Because such perturbations are likely to occur over large areas, and because arid landscapes in the western United States are vast, it is important that the consequences of these perturbations be assessed at the scale of identifiable landscapes. To this end, the changes in important plant processes resulting from ecosystem alteration must be known at the plant scale to have accurate data to scale up to landscape and global levels.

Field studies were initiated in the spring of 1987. Two study sites were chosen on the ALE Reserve to represent an undisturbed sagebrush-, *Artemisia tridentata*, bunchgrass, *Agropyron spicatum*, community and a disturbed old field (Lower Snively) community dominated by cheatgrass, *Bromus tectorum*. The undisturbed community covers large expanses of the Columbia Plateau. The old field, dominated by cheatgrass, represents a community type that has become established over wide regions of the arid West where humans have disturbed the soil.

At each site, 20-m² study plots were randomly chosen with replication for untreated controls, water, nitrogen, and water plus nitrogen treatments. Nitrogen was added at the rate of 100 kg/ha. In fall 1987 and spring 1988, water was added to equal the 30-year maximal precipitation record. All gas exchange observations were obtained under constant chamber conditions of temperature, light, relative humidity, and CO₂. This report presents results for the gas exchange [net photosynthesis (a measure of metabolic energy balance), transpiration, and water use efficiency] and xylem pressure potential (water stress) observations (see Figures 1 through 3).

FIGURE 1. The Effect of Additional Water and Nitrogen on a) Xylem Pressure Potential, b) Transpiration, c) Net Photosynthesis, and d) Water Use Efficiency of *Bromus tectorum*. Error bars are one standard error of the mean (n = 5). The same letter above two or more error bars for each block indicates no significant differences at the 95% confidence level by Duncan's Multiple Range Test.

It was hypothesized that additional water would result in lower water stress (xylem pressure potential values closer to zero), greater transpiration, greater net photosynthesis, and lower water use efficiency. Water stress was lower for all three species, as would be expected when water is added to this arid ecosystem (all comparisons are relative to control levels). Transpiration, however, while greater for bunchgrass and sagebrush than controls, was unaffected for cheatgrass. It was expected that all three species

FIGURE 2. The same as Figure 1, but for *Agropyron spicatum*

would exhibit greater transpiration given that water stress was less in all three. However, because stomatal conductance in cheatgrass was unaffected by additional water, the species showed no change in transpiration. Stomatal conductance was relatively greater for the other two species.

Additional water was also expected to result in greater net photosynthesis as a consequence of lower water stress. This was the case only for bunchgrass. Cheatgrass had lower net photosynthesis, which may be related to the relatively

FIGURE 3. The same as Figure 1, but for *Artemisia tridentata*

greater growth in the leaves resulting from reduced water stress. If growth is high, then net photosynthesis is low because respiration, which is required for increased growth, is high. No effect was found for sagebrush. Water use efficiency is generally expected to decrease when water is added to arid systems because transpiration increases more than net photosynthesis. This occurred for cheatgrass and bunchgrass, while no significant effect was found for sagebrush.

Additional nitrogen was hypothesized to cause higher water stress, lower transpiration, greater net photosynthesis, and higher water use efficiency. Water stress was higher for bunchgrass and sagebrush, while no effect was found for cheatgrass. With additional nitrogen, growth increased, which reduced the reserve of soil water, thereby causing greater water stress. Transpiration was lower only for sagebrush, which implies that the soil reserve of water was lowered because of increased growth. Transpiration showed no changes for the grasses, even though water stress was greater, suggesting that the additional water stress was not biologically significant. Additional nitrogen had been expected to cause higher net photosynthesis; however, the grasses showed no such effect suggesting that the increased water stress had more of an effect on net photosynthesis than did the additional nitrogen. Sagebrush had lower rates of net photosynthesis suggesting that the additional water stress was more limiting, or that higher growth rates (higher respiration) reduced net photosynthesis. Water use efficiency was higher for cheatgrass and bunchgrass while water use efficiency was unaffected for sagebrush.

Additional nitrogen plus water was expected to result in lower water stress, higher transpiration, greater net photosynthesis, and no difference in water use efficiency, because neither water nor nitrogen would be limiting. However, water stress was lower only for cheatgrass with no effect for bunchgrass and sagebrush. The only effects for transpiration, net photosynthesis, or water use efficiency for these species was higher transpiration in cheatgrass and lower net photosynthesis in sagebrush. Again, higher transpiration may be the result of lower water stress, and the lower net photosynthesis may be the result of higher growth rates (sagebrush had significantly greater growth in this treatment). In the other cases, the additional water may have been used to support the additional growth caused by the additional nitrogen in a compensating manner, yielding rates for gas exchange and water stress similar to the control plants.

These results are a step toward understanding how additional nitrogen and water induced by human alterations of the atmosphere will influence plant productivity and interact with ecosystem-level dynamics of nitrogen and water. Then, process-based models can be developed to predict landscape-level response to stress.

The results of this research will be linked with similar work conducted in cooperation with scientists at the Idaho National Engineering Laboratory (INEL) to better understand the general nature of plant response to perturbation in arid landscapes.

Spatial Variability in Ecological Factors Influencing Surface Water Distribution in the Shrub-Steppe

R. L. Skaggs and L. L. Cadwell

It is recognized that reliable physics-based hydrologic and transport models require detailed characterization of surface and subsurface hydrogeologic properties, vegetation, and surface roughness. Studies are being conducted at the Watershed Research Facility to determine the significance of spatial variability in vegetation and soil properties on hydrologic response in the shrub-steppe and to evaluate the importance of measurement scale on parameter estimation. Current work encompasses measurements of physical properties and hydrologic response across two scale gradients: point ($\sim 4 \text{ cm}^2$) to subplot (1 m^2) and subplot to plot (30 m^2). At the point scale, field and laboratory measurements of hydraulic and physical properties will be (geo)statistically analyzed to determine the relationships between soil and plant spatial variability. Subplot studies facilitate measurement of hydrologic response to isolated landscape features such as vegetation type or soil condition, while the plots provide measurements of integrated response to spatially varying plant cover, surface and subsurface soil properties, and micro-relief. Continuous lysimeter measurements of evapotranspiration, soil moisture storage, and drainage are used with the plots to fully characterize the study site water balance.

The primary objective of the multiple-scale investigations is to answer the question: When scaling up hydrologic model parameter estimates from point scale to subplot, plot, hillslope, landscape, and larger scales, what uncertainties are introduced, and to what extent can they be quantified?

Other objectives include:

- determining the optimum measurement scale for estimating key hydrologic parameters (e.g., hydraulic conductivity)

- determining the areal extent of the influence that selected vegetation types have on soil properties
- computing representative mean values of soil properties in the vicinity of selected vegetation types
- determining correlations between mean values of saturated hydraulic conductivity and results of plot and subplot experiments
- identifying cross correlations between soil properties in the vicinity of selected vegetation types.

During FY 1988, the initial series of infiltrometer studies was conducted. Simulated rainfall was applied to 25 subplots containing five replicates each of five cover types: sagebrush, *Artemisia tridentata*; bunchgrass, *Agropyron spicatum*; Sandberg bluegrass, *Poa sandbergii*; cryptogam; and bare soil. Approximately 6.0 in. of precipitation was applied over 25 hr at intensities of 2.55 and 5.0 in./hr at varying antecedent moisture conditions, including dry, wet, very wet, and near saturated. Data collected include rainfall amount, antecedent soil moisture, and surface runoff. Preliminary analysis of the data provides measures of time-varying infiltration rate as a function of vegetation cover type, antecedent moisture, slope, and rainfall intensity.

The ratio of runoff to rainfall varied from a low of 0.23 in./hr for sagebrush to a high of 0.42 in./hr for the bare soil (Figure 1). Several trends are evident in the results. The low ratio for the sagebrush plots reflects the effect of plant roots and organic matter in the soil on effective permeability, soil structure, bulk density, porosity, depression storage, and surface roughness. At the other extreme, the bare soil and cryptogam plots had similarly high runoff to rain ratios. These treatments had little impact on porosity, soil organic matter, or microrelief. Sandberg bluegrass and bunchgrass demonstrated the incremental effect of more shallow-rooted plants on infiltration capacity.

The influence of plant cover on the hydrological balance is further illustrated in Figure 2, which shows a comparison of infiltration curves for a sagebrush plot and a bare soil plot for dry antecedent conditions. For both plots, the initial infiltration rate matched the rainfall application rate of 2.55 in./hr. After about 5 min, however, the infiltration capacity of the bare soil plot dropped

FIGURE 1. Average Ratios of Runoff to Simulated Rainfall for Five Cover Type Treatments. The lower values for plants reflect the positive influence of roots and organic matter in rainfall infiltration.

FIGURE 2. Water Infiltration Curves for Sagebrush and Bare Soil

below the rainfall rate, and runoff began. The infiltration rate continued to decrease throughout the event, with a final value of about 1.3 in./hr. The sagebrush plot did not begin to generate runoff until more than 7 min into the event, and the infiltration rate did not drop below 2.4 in./hr.

These results are examples of how the mechanisms of surface and subsurface flow in arid ecosystems interact with vegetation. The current work is being conducted at relatively small scales to isolate and study at the process level selected system units with relatively homogeneous soils, terrain, and plants. The understanding gained

from these studies will help account for the effects of spatial and temporal variability, thus allowing quantitative extrapolation from plot studies to hillslope and watershed scales.

Measuring Landscape-Level Processes

P. A. Beedlow and L. E. Rogers

Landscape-level research at PNL is focused on scaling-up measures of controlling processes from plot to landscape scale using remote sensing technology. The purpose of this research is to determine which landscape processes can be quantitatively measured remotely. Initial work has concentrated on surface energy balance and evapotranspiration; however, other processes will be addressed as findings from this and other research projects become available.

The approach taken to remotely measure landscape processes is quantitative. Quantitative remote sensing can be contrasted to qualitative remote sensing by approach and application (Table 1). Qualitative interpretation is based on correlating remote and ground data empirically using multivariate analysis. With satellite data, spectral band ratios (used in registering the energy reflected and emitted from plants and soil) are correlated with ground measures such as leaf area or biomass. Quantitative interpretation, however, is based on the measurement of absolute radiance of the land cover. Absolute radiance values are used for direct measurement of land surface features, such as soil properties or vegetation. Quantitative measurements require calibration for atmospheric absorbance, emissivity of land-cover features (for thermal data), and reflectance angles.

TABLE 1. Quantitative Versus Qualitative Remote Sensing

	Qualitative	Quantitative
Definition	An interpretation of data based on correlation between satellite and ground truth	An interpretation of data based on absolute radiance
Approach	<ul style="list-style-type: none"> • Band ratios • Ground truth • Multivariate analysis • Reflectance angle 	Calibration for: <ul style="list-style-type: none"> - Atmosphere - Emissivity
Application	<ul style="list-style-type: none"> • Restricted in time and space to ground truth data and space • Traceability 	<ul style="list-style-type: none"> • Extrapolation beyond ground truth • Consistency in time

Qualitative data application is restricted in time and space by empirically derived relations to ground data. Ground data must be used to calibrate the remote data for each new site and each new date. Once quantitative remote data are calibrated for atmospheric absorbance, emissivity, and reflectance angle, data interpretation is not limited in time or space because the information is in terms of absolute radiance. Quantitative measures, then, allow for extrapolation beyond ground truth data and provide for consistency in time and space. Additionally, quantitative measures permit comparisons through time, especially difficult qualitatively because atmospheric conditions are rarely the same from one time period to another.

Thus, to further develop quantitative remote sensing technology to understand landscape-level processes, the objectives of this research include:

- obtaining direct measures of parameters controlling landscape processes, such as surface temperature, reflectance, net radiation, soil moisture, aboveground organic material, and leaf area. The measurement of absolute biophysical-hydrological quantities using remote techniques would allow quantification of key landscape processes (e.g., net primary production, evapotranspiration, runoff, infiltration) across time and space. These principal factors would also provide for determinations of water quality and quantity, and ecosystem state or condition.
- providing measures of uncertainties (variability) for risk assessment
- identifying and quantifying landscape units. Measuring structural features of landscapes using remote techniques is necessary for resource inventories and for modeling ecosystem and landscape processes not amenable to direct measurement from airborne platforms (e.g., decomposition).
- providing remote measurements to aid in establishing policy and making management decisions. Critical information for management and policy decisions would include watershed dynamics and ecosystem health.

Over the last 3 years, the research has addressed critical aspects of quantitative remote sensing of landscape processes to determine if such measures were possible from satellites and aircraft. Research issues included: 1) atmospheric

correction of imagery to obtain absolute radiance values, 2) emissivity of land cover types, 3) the effect of reflectance angles on remote land cover measures, 4) development of ground truth techniques, and 5) adaption of landscape process models to operate on remotely collected data.

The OHER research at PNL has used information from research being conducted for the National Aeronautics and Space Administration (NASA) and the U.S. Department of Defense (DOD) to gain the techniques and expertise necessary to remotely measure landscape processes.

A NASA on-orbit calibration of the Landsat Thematic Mapper (TM) Band-6 and reflectance bands, combined with remote sensing (REFLEX) research, addressed the atmospheric and land-cover calibrations for landscape processes. Absolute surface and atmospheric temperature and reflectance measures made on the ground at the time of Landsat overpasses were compared with TM-determined values. Atmospheric corrections were made using the LOWTRAN 6 code, developed at the U.S. Air Force Geophysical Laboratory, based on local and nonlocal radiosonde data and standard atmospheric corrections. The accuracy of TM-determined measures of surface temperatures was demonstrated to within 1°C when atmospheric corrections were made. Local atmospheric measures were necessary to attain maximum accuracy. However, U.S. standard values produced relatively good results, especially for regional measurements. It was necessary to use local emissivity calibrations for terrestrial land-cover measurements, such as for green vegetation, dry vegetation, and soil types, to achieve accuracies within 1°C.

The effects of reflectance angles on TM data were determined using ground measurements from a portable spectroradiometer taken during satellite overpasses. These measurements were used to test the accuracy of TM Bands 1 through 4. Bidirectional reflectance measures of major land-cover types were made seasonally to determine reflectance spectra. Results of this research are summarized in Table 2. Reflectance angles can have a significant effect on the accuracy of land-cover measurements taken from satellite data. This is because surface features, especially vegetation, reflect solar radiation differently in different directions. This phenomenon can result in measurement errors when band ratios are used (e.g., to determine leaf area or biomass). Different types of land cover, such as grassland

TABLE 2. Summary of Landsat TM Calibration Results

Parameter	Accuracy/Error
Thermal	
• Water	
uncorrected	~1°C ± 4%
corrected with local radiosonde	~0.6°C ± 2%
• Vegetation	
uncorrected	~5°C
corrected; emissivity = 1	~3°C
corrected; emissivity ≠ 1	<1°C
Visible	
• Nadir reflectance	~2%
• Bidirectional reflectance needed for on-nadir adjustments and for land-cover characterization	

and shrubland, have distinctive bidirectional reflectance spectra. This implies the ability to differentiate vegetation types using bidirectional data. The MODIS sensor planned for NASA Eos (satellite) platforms will be able to make bidirectional measurements.

The accuracy of TM visible Bands 1 through 4 was determined by comparing TM values, corrected for atmospheric effects, with spectroradiometer measures taken at the time of overpass. (The TM scans only at nadir solar angles, which is when the sun is at zenith.) June and July data were taken to compare green (June) versus dry (July) vegetation. The overall error between TM and ground truth values averaged 1.5%. These results show relatively little reflectance error during the summer when the sun angle approaches nadir. However, the error at greater solar angles can be expected to be larger.

Portable micrometeorological stations are being used in PNL's research to provide ground validation for satellite data and for developing energy balance models of landscape processes. During ground validation, micrometeorological data are collected during satellite overpasses. For energy balance modeling, micrometeorological data are used in conjunction with data from precision weighing lysimeters located at PNL's Watershed Research Facility.

Ground validation research has focused on instrumentation and determining energy balance differences for major landscape units at Hanford (Figure 1). Data collected during Spring 1988 and analyzed using the Bowen ratio technique demonstrated that major landscape units at Hanford

FIGURE 1. Comparison of Evapotranspiration (ET) for Sagebrush and Grass, Calculated from Energy Balance Data and Measured by Lysimeter Weight Changes

have significant differences in the energy balance parameters that drive evapotranspiration (ET), such as surface temperature, soil heat flux, and air temperature. Differences in micrometeorological measures are supported by thermal and visible band differences in TM data. Subsequent research will compare ET estimates from TM data with ground measures from the micrometeorological stations.

Floristics and Climate in the Deserts of the Western United States: Anticipating Global Change

W. H. Rickard

One of the most serious environmental concerns of today's society is the greenhouse effect, a change in global climate induced by the warming effects of human-induced releases of carbon dioxide into the earth's atmosphere during fossil fuel combustion. Under a scenario of warming air temperatures and decreasing precipitation, it is expected that primary productivity on arid lands would decrease. Some plant populations may migrate along latitudinal or altitudinal gradients, and others may become extinct. In the absence of field data and permanent plots, changes in

floristics could go undetected, especially if dependent upon relatively short-term field studies. Thus, the taxonomic composition and canopy cover for two desert sites have been established and compared to provide baseline information for indication of change.

Floristic comparisons of landscapes in different portions of the deserts of the western United States have seldom been made. This study floristically compares two broad, flat, intermountain valleys, each supporting natural stands of vegetation in widely separated locations. Jackass Flats is on the Nevada Test Site in Nevada, and the Cold Creek Valley is on the Hanford Site in Washington. The study areas are similar in size and have similar land-use histories, i.e., long-term absence of grazing livestock. Cold Creek Valley is located 10° north of Jackass Flats and is 2190 feet lower in altitude (Table 1). It is also climatically characterized by slightly greater annual precipitation, lower maximum air temperature, and higher minimum air temperature (Table 1) than Jackass Flats. The same methods of field survey were used in both locations.

Five broad categories of plants, other than cacti, were present during this study (Table 2). Thirty

TABLE 1. Comparison of Selected Geographic and Climatic Features of the Cold Creek Valley, Washington, and Jackass Flats, Nevada

Location	Longitude	Latitude	Altitude (ft)
Washington	119° 36'W	46° 34'N	733
Nevada	116° 03'W	36° 57'N	3924
	Precipitation (in.)	Maximum Temperature (°F)	Minimum Temperature (°F)
Washington	6.25	64.8	41.2
Nevada	5.73	72.5	37.4

TABLE 2. Number of Taxa and Canopy Cover Provided by Five Plant Categories in the Cold Creek Valley, Washington, and Jackass Flats, Nevada

Location	Shrubs	Perennial Grass	Annual Grass	Annual Forbs	Perennial Forbs	Total
Number of Taxa						
Washington	4	5	2	12	7	30
Nevada	10	2	2	36	0	50
Canopy Cover (%)						
Washington	20	10	12	1	<1	43
Nevada	12	1	2	9	0	24

plant species (taxa) were found in the Cold Creek Valley compared with 50 at Jackass Flats (Table 2). More shrub and annual forb taxa occurred in Jackass Flats, but there were more perennial grass and perennial forb taxa in the Cold Creek Valley. The dominant shrub in the Cold Creek Valley was big sagebrush, *Artemisia tridentata*, and the dominant shrub in Jackass Flats was creosote bush, *Larrea tridentata*. Creosote bush is restricted to the Mojave and Sonoran deserts of California, Arizona, Nevada, and Utah, while sagebrush is widely distributed in the intermountain valleys of Oregon, Idaho, northern Nevada, northern Utah, and western Wyoming. Sagebrush flowers in early autumn (September through October), while creosote bush flowers in April and May. Both plants have the C_3 photosynthetic pathway.

The genus *Artemisia* is represented by several taxa in the western United States, but *Larrea* is represented by a single species. Both shrubs tolerate extended periods of summer soil drought. Sagebrush is a relatively short-lived plant in the Cold Creek Valley, less than 100 years old, but creosote bush lives to be much older (Rickard 1988). Sagebrush is easily killed by burning; it does not sprout, and re-establishment after

burning is by seeding. Creosote bush stands seldom burn under natural conditions because of the low fuel potential created by wide spacing of shrubs and a sparse herbaceous understory.

Creosote growing at the northern extremity of its range in Utah has been reported to die back by abnormally cold winter weather; however, it can survive occasional diebacks (Rickard 1963; Rickard and Beatley 1965). Sagebrush is extremely tolerant to cold temperatures, and its southern distribution may be limited by hot summer weather in conjunction with low precipitation.

Primary production of herb taxa as crudely measured by canopy cover is greater in the Cold Creek Valley than in Jackass Flats, indicating that the Cold Creek Valley has more potential for livestock grazing than Jackass Flats but, in either case, grazing potential is limited (Table 2). The most abundant livestock-palatable herb in the Cold Creek Valley is the alien annual grass, cheatgrass, *Bromus tectorum*, which is palatable when green to all classes of livestock as well as some wildlife.

Only five plant taxa out of 75 were shared between the study locations. These were spiny hopsage, *Grayia spinosa*, a shrub; Indian ricegrass, *Oryzopsis hymenoides*, a perennial bunchgrass; six week's fescue, *Vulpia octoflora*, an annual grass; and *Cryptantha circumscissa*, *C. pterocarya*, and *Mentzelia albicaulis*, three annual forbs. These plants provided little canopy cover, and they are regarded as minor constituents of the flora. Spiny hopsage dominates shrub cover at only two locations in the Cold Creek Valley, but it is an important constituent of the vegetation elsewhere on the Nevada Test Site.

One of the basic tenets of ecological studies is that the global distribution of terrestrial plants is determined by climatic variables, especially precipitation and temperature. Local plant distributions are segregated by differences in rooting substrates and/or by contrasting topography. Also, the periodicity of burning determines the local distribution and abundance of plants, at least during periods of less than 100 years.

This research shows that conventional nondestructive measurements of species composition, in conjunction with measurements of canopy cover, provide a satisfactory method of detecting and evaluating differences in natural plant communities

in arid and semiarid regions. Thus, the distribution of representative plants can be used as a natural indicator of regional or global climatic changes resulting from energy-related impacts. Changes over time can also be detected and evaluated, but these are most cost-effectively and accurately determined by establishing a series of permanent plots in representative plant communities. Permanent plots are most desirable in large areas that are free from livestock grazing, herbicide drift, off-road vehicles, and other extraneous plant-damaging human-induced interferences. Such places occur at the DOE NERP sites in southeastern Washington, southeastern Idaho, and southern Nevada. These sites also have data on historical weather patterns, previous land uses, and soil properties, which influence plant species composition and/or canopy cover. The NERP sites are particularly desirable in detecting and evaluating wide-ranging, slowly paced changes, such as those anticipated by a changing global climate. Under these conditions, conventional field plot experimental designs, for example treatment versus nearby control, are not appropriate.

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Future Research

PNL research over the next 2 years will focus on computing net radiation and surface temperature from TM data. Successful computation of these parameters and ground validation will permit construction of energy balance maps (including ET) for the shrub-steppe vegetation on the Columbia Plateau. This information will be assessed in terms of its applicability to other regions. Procedures will be made final for calibrating satellite imagery for emissivity and reflectance angles. Spectral indicators of

vegetation stress will be investigated. Ground truth technology will be improved and alternate methods, such as eddy correlation, will be tested. An ET model based on energy balance parameters will be refined for arid landscapes.

Longer-term research will focus on direct quantitative measurement of surface energy balance parameters combined with advanced spatial modeling to estimate and predict rates and changes in landscape processes. This research will use planned NASA space platforms such as Eos. The intent is to increase the number of variables that can be measured directly from space while minimizing the amount of ground data that must be collected. For example, net radiation and surface temperature measures currently can be obtained from satellites; computation of ET requires air temperature data from ground stations. With the instrumentation slated to fly on Eos, air temperature measures will be attainable from space. Other parameters, such as total solar and diffuse radiation, will not be directly measured from satellite and will require atmospheric modeling.

Although ET was the first major landscape process addressed in this research, it is recognized that a holistic view of landscape dynamics will require measurement of additional processes. A major process controlling the health of ecosystems and the process dynamics of watersheds is net primary production. Net primary production can be estimated from energy balance data at small scales. Future research will address large-scale (landscape and regional) estimations of net primary production.

Advanced air- and spaceborne spectrometers will enable measurement of plant stress, and the chemical composition of plant material over large areas. This capability will greatly enhance the ability to detect ecosystem stress in advance of irreversible damage. The ability to estimate the chemical composition of vegetative material over large areas will allow the construction of regional biogeochemical models. The successful development and use of spatial models depicting the response of landscapes and regions to stress relies on the ability to integrate and synthesize large amounts of information from a wide variety of sources. To support this effort, PNL is developing knowledge-based approaches to analyze and model biosphere processes. Initial research is concentrating on distinguishing and quantifying landscape units for inclusion in watershed models. Future research will construct

knowledge-based systems to evaluate ecosystem health and predict responses to stress factors.

The resolution of issues surrounding the effects of global change on the biosphere will require new paradigms in ecology. Our current view of ecosystems and how they respond to stress has grown from structure-function research conducted at laboratory and plot scales. The result has been an unmanageably complex view of

ecosystems and how they respond to change. Future ecological research must build on physical theories to develop a thermodynamic perspective of the biosphere that can be measured on a range of scales from global to individual watersheds. This perspective must recognize physical models of the atmosphere, oceans, and land surface, and use data gathered from spaceborne instruments.

Biogeochemical
Phenomena

Biogeochemical Phenomena

Biological processes associated with rhizospheres often have strong human health and environmental implications on a global scale. These processes are dynamic and can rapidly influence contaminant behavior and fate. PNL has pioneered research on the metabolic processes that influence the transfer of energy-related radionuclides, inorganic ions, and organic compounds in the food chain. These investigations provided a unique background for probing fundamental biological controls on elemental cycling in these systems.

This program helped elucidate and quantify the biogeochemical processes controlling the behavior of inorganic elements and organic compounds important in global cycling and transport pathways. The research established basic concepts in the laboratory and validated these concepts under controlled field conditions. The information developed added a metabolic and kinetic dimension to elemental cycling, organic transformation, and food ingestion models, which have lacked long-term predictive capability.

Soil and Plant Processes Controlling Cation Behavior in Terrestrial Systems

H. Bolton, D. A. Cataldo, R. E. Wildung, and T. R. Garland

Soil Microbial Processes

The ability to understand and predict the long-term fate and behavior of a cationic element in the environment requires fundamental knowledge of biotic, chemical, and physical processes. Soil microorganisms can influence both nutrient and contaminant cation mobility by altering the physicochemical environment and by producing organic complexing ligands. Laboratory experiments were designed to investigate rhizosphere-microbial processes that influence cation mobility in soil. Ongoing field research at the Arid Lands Ecology (ALE) Reserve provides field validation of these basic laboratory experiments.

Cheatgrass, *Bromus tectorum*, and bunchgrass, *Agropyron spicatum*, were selected for laboratory studies because these grasses are major components of the arid shrub-steppe at the Hanford Site, and field validation of observed laboratory phenomena could be conducted in the future. *B. tectorum* is an annual grass with shallow roots, while *A. spicatum* is a perennial with deep roots. Both grasses support a large microbial population in field soil.

Seedlings of *B. tectorum* and *A. spicatum* were grown from pre-germinated disinfected seed in a vermiculite/sand mixture with a nutrient solution supplied for plant growth. The growth tubes were amended with trace levels of nickel-63, and 100 ppm nickel plus nickel-63. One treatment was inoculated with a mixture of field soil from

underneath *B. tectorum* and *A. spicatum*. The rhizosphere soil solutions were collected by centrifugation after seedling growth for 10 days. These solutions were leached through soil columns to determine the soil mobility of the nickel-63 present in the rhizosphere solutions. These soil columns bound any free and weakly complexed nickel, while only strongly complexed nickel was soil soluble. *B. tectorum* and *A. spicatum* seedlings increased the soil-soluble nickel concentrations in the rhizosphere solution by 4 and 6 fold, respectively, when compared with the controls (no plant growth). Thus, plant growth and rhizosphere processes can increase the mobility of contaminant cations.

Rhizosphere microorganisms can also influence cation mobility. *Cylindrocarpus* sp. strain 458, a rhizosphere fungus isolated earlier by PNL scientists, can increase the mobility of nickel through soil by producing an organic ligand that complexes nickel. Strain 458 was reisolated after long-term storage and was still able to complex nickel as determined by thin-layer electrophoresis. Initial findings of this in vitro study indicate the nickel complex forms very fast and is stable under sterile conditions over time. The fungus is able to use glucose as a sole carbon source and readily produce the complex without induction, indicating the fungus may be able to grow well in the rhizosphere and produce the complex. Later, the growth of strain 458 was stimulated in the rhizosphere, but the organism grew slowly, indicating extended in vivo plant growth studies may be necessary to fully understand the influence of strain 458 on cation mobility in the rhizosphere. Separate growth studies show the fungus is able to grow well from 15 to 37°C, indicating it is well suited to the arid environment. In separate studies, preliminary results indicate strain 458 was

able to influence nickel mobility in the rhizosphere of *B. tectorum* when compared with heat-killed cells.

Plant Complexation Potential for Cations

Terrestrial plants, via food webs, are an important link between their environment and humans. As such, plants can be a significant source of potentially toxic elements. Although a substantial effort has been expended over the last 40 years to evaluate and quantify the transfer of toxic elements from soils to plants, comparatively little attention has been given to the physiological controls influencing element uptake, transport, and most importantly, chemical form in plants. The chemical form of an element will not only affect its chemical and physiological behavior once accumulated by plants, but also its solubility, chemical stability, and ability to be bioconcentrated along food chains.

Once an essential element is absorbed into the root, some mechanism must be present to prevent sorption, hydrolysis, or non-specific chemical reaction between the myriad of trace and macroions being stored, transported, or metabolized. A similar process must affect pollutant elements as well. In plants, this problem is most likely avoided by organic complexation of cations. A substantial body of qualitative data indicates that the nutrient ions calcium, cobalt, iron, manganese, and zinc exist in organically complexed form in xylem exudates, as do manganese and zinc in phloem exudates.

Recently, xylem exudates have been receiving increased attention in studies attempting to understand the role of chemical complexation in the chemical stabilization and subsequent availability to animals of potentially toxic elements. Although the importance of plant complexation as a transport process began with the work of Tiffin (1977) on iron complexes in exudates, more recent studies have addressed the behavior of pollutant elements such as neptunium, plutonium, nickel, and cadmium in plant exudates, and the composition and role of xylem exudate constituents on cation complexation and chemical solubilization. These studies have shown that a wide range of cations, even the most hydrolytic polyvalent species such as plutonium, are mobile and soluble in transport fluids such as xylem exudates, and exist primarily as organically complexed species. These studies, coupled with the fact that many nutrient and non-nutrient cations extracted from plant tissues (leaves, roots, and seeds) are

soluble and associated with organic ligands of varying molecular weight, suggest that plant-produced organometal complexes may be important in the transfer of potentially toxic elements from plants to animals.

The objectives of these studies were to characterize the major organic components of soybean xylem exudates; evaluate the complexation capacity of xylem exudates for plutonium, iron, nickel and cadmium; and assess and compare the overall physiological and metabolic behavior of neptunium.

Natural Transport Ligands in Plant Fluids. The xylem exudates of soybean, *Glycine max* cv. Williams, were characterized as to their organic constituents and in vivo and in vitro complexation patterns for plutonium, iron, cadmium, and nickel. Ion exchange fractionation of whole exudates into their compound classes, followed by thin-layer electrophoresis, permitted evaluation of the types of ligands stabilizing these elements. The polyvalent elements plutonium-238 and iron-59 were found primarily as organic acid complexes, while the divalent elements nickel-63 and cadmium-109 were associated primarily with components of the amino acid/peptide fraction. Nickel was complexed by a tripeptide, which was only partially characterized. It was not possible to fully duplicate complexes formed in vivo by back reaction with whole exudates or individual class fractions, indicating the possible importance of plant induction processes, reaction kinetics, and/or the formation of mixed-ligand complexes.

Soil Plant Behavior of Neptunium. Soil-plant concentration ratios (CR) for neptunium in bush bean, *Phaseolus vulgaris* var. Tendergreen, measured over the range of 5.2×10^{-7} to 4.1 mg Np/g soil, are approximately 2 at soil concentrations below 4×10^{-4} $\mu\text{g/g}$, and increase to 12 at higher soil levels. The CR values determined for soybean, bush bean, barley, and alfalfa range from 0.5 to 4 at a soil concentration of 2.6×10^{-6} mg/g. Root absorption by soybean seedlings of neptunium from solutions containing 7×10^{-7} to 473 $\mu\text{g Np(V)}/\text{mL}$ is generally proportional to concentration, but exhibits some saturation in root absorption at higher concentrations. Seed concentrations in bush bean and wheat are a factor of 10 lower than vegetative tissues. Neptunium is transported within the plant in organic complexes containing one or more organic acid residues. Fractionation of plant tissues indicates that neptunium is substantially more soluble than plutonium, especially in seeds,

with approximately 50% of the soluble neptunium in roots and leaves associated with plant ligands of less than 5000 molecular weight. The relative behavior of neptunium versus plutonium can be postulated to result from the aqueous speciation dynamics of these two actinide elements.

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Environmental Behavior of Inorganic Anions

T. R. Garland, D. A. Cataldo, H. Bolton, R. J. Fellows, and R. E. Wildung

Nitrogen Cycling

Anions, including those derived from nuclear and fossil fuel development and use, are generally mobile in the environment and exhibit high bioavailability. These traits can result in relatively rapid movement to surface/groundwater systems and produce adverse impacts in food chains. Unlike the dynamics of cation mobilization, the processes controlling contaminant anion behavior appear to be directly related to, and controlled by, the endogenous enzyme systems responsible for the reduction and incorporation of nutrient anions, particularly nitrogen and sulfur. Resolving the behavior and fate of potentially hazardous anions requires that we address those processes responsible for anion speciation at the molecular and biochemical levels.

The study of nitrogen transformations in soil will aid our understanding of the basic principles and processes that govern anion mobility, speciation, and bioavailability. The soil nitrogen cycle is a useful model for investigating anion behavior in soils, because the nitrogen cycle has anionic constituents, is subject to biological reduction and incorporation, and contains both organic and inorganic volatile components. By understanding the physical, chemical, and biotic parameters controlling anionic nitrogen transformations in soil, other anions will be better understood. In addition, nitrogen is an essential element for growth of all ecosystems. The influence pollutant anions have on the dynamic process on nitrogen

transformation must be known to predict anion mobility, speciation, and behavior in natural ecosystems.

Field plots have been established on the ALE Reserve. Treatments include additional water, additional nitrogen as ammonium nitrate, additional water and nitrogen, and a control. These treatments are being used as perturbations of ecosystem nutrient cycling to influence nitrogen pool sizes and rates of transfer between pools. The field plots are located at an annual grassland site and a perennial shrub-steppe site. Nitrogen-15 as ammonium sulfate was added to microplots containing *Bromus tectorum*, a dominant annual grass at the annual grassland site, and Sandberg bluegrass, *Poa sandbergii* (Vasey), a dominant perennial grass at the shrub-steppe site. Nitrogen was applied uniformly to a depth of 10 cm at 3 $\mu\text{g/g}$ soil to pulse label the mineral nitrogen fraction of the soil. The soil was sampled at 0- to 5- and 5- to 10-cm depths at 1, 3, 7, 10, and 14 days after application of the nitrogen-15. The nitrogen pool sizes were determined, but the rate of transfer between pools is still to be determined through nitrogen-15 analysis of the various pools.

A major source of plant and microbial available nitrogen is the organic nitrogen fraction mineralized by the soil microbial biomass. Organic nitrogen mineralization is a dynamic process that results in the cycling of nitrogen to various pools in the ecosystem. An understanding of controlling factors and rates of nitrogen mineralization allows for predictions of contaminant anion influences on this essential biotic process. Soils were collected from ALE underneath various arid ecosystem plant communities. These soils were incubated in the laboratory and subjected to sequential leaching over time to quantify microbially mediated organic nitrogen mineralization. Differences between the various soils sampled were only apparent at the surface (0- to 5-cm depth). As soil depth increased to 5 to 10 and 10 to 15 cm, differences between the various plant communities decreased, indicating the surface soil contains the majority of mineralizable organic nitrogen. Soils samples underneath grasses (*Bromus tectorum*, *Agropyron spicatum*, *P. sandbergii*) had higher rates and amounts of organic nitrogen mineralized than soil underneath sagebrush, *Artemisia tridentata*, and cryptogamic soil lichen crusts. Future studies are planned, adding various oxyanions to study their influence on organic nitrogen mineralization.

Soil/Plant Behavior of Radiochloride

The plant availability of soil-amended radiochloride and endogenous soil chloride was determined for alfalfa, *Medicago sativa*, and barley, *Hordeum vulgare*, grown on Burbank surface soil and subsoil. Plant/soil concentration ratios (CRs) were as high as 200 and 130 for first-harvest alfalfa leaf and stem tissue, respectively. For barley, CR values were approximately 15 for leaves and stem, and 30 for seed heads. Little difference in CR was apparent for surface soils and subsoils. Similarly, the presence or absence of amended carrier chloride had little effect on CR value. In multiple harvest studies, a rapid depletion of soil-amended tracer was evident. Evaluation of the relative plant availability of endogenous soil chloride pools showed a similar level of bioavailability, indicating a saturation of the plant uptake process for chloride at normal soil chloride levels.

Unlike many other radionuclides or inorganic contaminants entering soil systems, chloride is a normal soluble constituent of many soils. As such, the presence of endogenous soluble chloride pools in soil may influence and control the apparent plant availability of radiochloride. Results presented above, namely similarities in CR for carrier-free and added carrier treatments, indicated that plant uptake was saturated and near maximal at chloride levels $<3.2 \mu\text{g/g}$ soil. This would suggest that CR values for stable chloride should be comparable to those for radiochloride.

The purpose of the present study is to determine the relative plant availability of $^{36}\text{Cl}^-$ amended to Burbank surface soils and subsoils. This soil is

characteristic of the Hanford 200-West Area, and is adjacent to proposed burial locations for decommissioned reactor components. The availability of soil-amended $^{36}\text{Cl}^-$ was determined for several croppings of alfalfa, and for up to 1-year soil equilibration before planting barley.

Table 1 compares the plant availability of endogenous soil chloride with the availability of amended radiochloride. The CR values presented for the endogenous chloride are based on both the hot water and acid extract values for surface soil of 3.7 and $7.9 \mu\text{g/g}$. For alfalfa, CR values for leaves and stem tissue are 244 and 261, respectively. Leaves, stem, and seed head of barley have CR values of 33, 45, and 91, respectively. Use of the acid extract value of $7.9 \mu\text{g/g}$ decreases the calculated CR values by a factor of approximately 2. Tissue accumulation patterns for the endogenous chloride are similar to those determined for the radiochloride, with the seed head showing elevated accumulation over vegetative tissues.

In general, CR values for the endogenous chloride are 2 to 3 times higher than for the radiochloride treatments when the water extractable soil chloride values are employed for calculation of CR. If the higher soluble soil concentrations determined by acid extract were employed for the endogenous calculation, the endogenous and amended CR values would be more comparable. This increasing similarity indicates that the acid-extractable soil chloride pool better reflects the plant-available fraction of soil chloride. The overall similarity between CR values for radio-traced amendments and endogenous soil chloride pools indicates that any contaminant chloride

TABLE 1. Comparative Bioavailability of Endogenous and Amended Soil Chloride

Plant Species/ Tissue	Endogenous Chloride ^(a)			Radiochloride ^(b)	
	Tissue	CR HWE ^(c)	CR AE ^(c) ($\mu\text{g/g}$ tissue)	CR Carrier Free	CR With Carrier
Alfalfa					
Leaves	905 $\pm$ 150	244 $\pm$ 40	114 $\pm$ 19	170 $\pm$ 7	139 $\pm$ 24
Stem	967 $\pm$ 189	261 $\pm$ 51	122 $\pm$ 24	144 $\pm$ 23	91 $\pm$ 15
Barley					
Leaves	123 $\pm$ 35	33 $\pm$ 9	15 $\pm$ 4	17 $\pm$ 3	16 $\pm$ 6
Stem	165 $\pm$ 37	45 $\pm$ 10	21 $\pm$ 5	18 $\pm$ 5	15 $\pm$ 1
Seed head	335 $\pm$ 57	91 $\pm$ 1	42 $\pm$ 7	35 $\pm$ 12	25 $\pm$ 7

(a) $n = 4$

(b) $n = 3$

(c) HWE and AE denote hot water extract and acid extract values for soluble soil chloride used to compute concentration ratio (CR).

entering these soil systems will mix with and trace the endogenous pools. The results also reinforce the hypothesis that chloride uptake from these soils is operating at maximum rates, because rates appear to be independent of carrier addition.

examination of the combined effects of chemical and microbial phenomena on the behavior of DOE mixed-waste components (e.g., radionuclide-synthetic organic liquid complexes).

Future Research

This research will be redirected in FY89 toward biogeochemical aspects of subsurface contaminant transport, providing a unique basis for

Subsurface Science

The contamination of subsurface water supplies by chemicals has become a major national concern, and DOE sites have come under increased scrutiny because of the presence of mixed (organic-radionuclide) waste. Because scientific understanding of contaminant behavior in subsurface environments is very limited, PNL is investigating basic processes that control the reactivity, migration, and persistence of organic chemicals and inorganic ions in these environments as a basis for prediction and remediation. Microbiological research is investigating the nature of microorganisms in different deep hydrogeological environments, their ability to degrade aromatic organic chemicals and complexing ligands, and the relationship of these processes to sorption phenomena. Subsurface chemistry studies are investigating multisolute sorption phenomena, cosolvent effects and complexation, and abiotic surface-enhanced degradation on subsoil and aquifer materials. Hydrological investigations are defining the flow physics of organic liquid/water multiphase mixtures in saturated and unsaturated porous media, and systematically examining the effects of subsurface heterogeneities on transport. Multidisciplinary transport studies are being performed in columns and in a unique intermediate-scale experimental facility where the combined effects of microbiology, chemistry, and hydrology on co-contaminant transport can be assessed. The research is driving the development of improved contaminant migration models and effective new strategies to reduce migration rates and remove contaminants from groundwater.

Subsurface Chemistry of Organic Chemical Mixtures

J. M. Zachara, C. C. Ainsworth, E. M. Murphy (PNL), and J. C. Westall (Oregon State University)

Organic contaminants in the subsurface often exist as compound mixtures with complex geochemical behavior. The transport rates of these mixtures cannot be readily predicted with current knowledge because the chemical reactions that control the sorptivity and abiotic degradation of these contaminant mixtures on groundwater solids are not well understood. This project was initiated to investigate the interfacial geochemistry that controls the aqueous concentrations of organic chemical mixtures in subsoils and groundwater. The overriding goal is to provide a basis for improved contaminant reaction models to predict contaminant migration in the subsurface.

The objectives of this project are to quantify and understand the surface reactions that are most important in regulating organic contaminant concentrations, and then to determine the environmental factors controlling the manifestation of these reactions in the subsurface. As a general approach, the project has been investigating phase transfer, sorption, and abiotic degradation reactions in "simple" surrogate experimental systems containing reactive phases (clays, oxides, organic substances, humic and fulvic substances, etc.) common in groundwater. Reaction-based descriptors from these idealized systems are then applied to more complicated, but more realistic, experimental systems that contain multiple

contaminants, cosolvents, or heterogeneous subsurface materials to identify the complex effects of sorbent heterogeneity, solute/surface solvation, and multispecies surface equilibria. Experiments with microorganisms are also performed in a companion PNL project to identify the joint effects of sorption and biodegradation on organic contaminant concentrations in groundwater.

This report summarizes the results of selected phase transfer, sorption, and sorption-degradation experiments performed in FY 1988. The research has relied heavily on the use of aromatic amines and neutral polyaromatic and N-aromatic compounds. These model compounds pose concern as environmental contaminants and exhibit carcinogenic and mutagenic behavior. Important new research hypotheses and questions have been resolved; these results are currently being prepared for publication. The research has been performed at PNL and in graduate studies at Oregon State University (OSU). Research funding on this project was also redirected this past year to support the GEMHEX (Geochemical/Microbiological/Hydrological Experiment) and age-dating components of the OHER Deep Microbiology Program. The results of this effort will be briefly described.

Cosolvent Effects

An important focus of the research program has been the sorption and abiotic transformation phenomena of organic contaminants in complex organic chemical mixtures. Mixtures of contaminants at dilute concentration in water and in the

presence of organic cosolvents have been investigated and published in past years of the program. In FY 1987, the program demonstrated the complex effects of organic cosolvents on the sorption of a hydrophobic ionizable compound by subsurface materials and clays. As part of that research, a thermodynamic approach was applied that successfully accounted for the cosolvent effect on the sorption phenomena. What was not resolved, however, was how the cosolvent affected the solution-phase ionization of the sorbate, which was an N-heterocycle compound and an organic base. The solution-phase ionization of the sorbate in a cosolvent must be accurately known to rigorously assess the cosolvent effect on the sorption of ionizable organic compounds. Additionally, measurement of the change pK_a in a cosolvent provides important information on the solvation status of the neutral and ionized organic species that is needed to interpret the environmental chemistry of the compounds.

As part of its subcontract, OSU measured the conditional acid dissociation (K_a) for a series of methylanilines in methanol-water mixtures (Table 1). The acid dissociation constant was observed to increase with methanol content, indicating an increased preference of the system for the deprotonated aniline and hydronium over the anilinium and water. This result may be rationalized by considering the competition between hydrophobic and ionic effects in the solvation of the species B, BH^+ , and H^+ in the following reaction:

The trend in the K_a suggests that the increase in the hydrophobic solvation of the un-ionized aniline in combination with the decreased ionic solvation of the anilinium ion drive the above reaction to the right, increasing K_a with increasing methanol content. Consistent with this hypothesis, it is noted that K_a -cosolvent changes (Table 1) increase with increasing hydrophobicity of the compounds (K_{ow}).

Effects of Mineral-Bound Humic Substances on Hydrophobic Organic Compound Sorption

Previous published research on this project has shown that carbazole sorption on subsurface materials is controlled by the organic carbon content of the natural sorbent. It has also been found, however, that the organic carbon fraction of different low-carbon subsurface materials exhibits a differing affinity (on a mass C basis) for carbazole. As a result, the partition coefficient (K_p) normalized to the organic carbon content (K_{oc} , where $K_{oc} = K_p/f_{oc}$ and f_{oc} = the mass fraction organic carbon in the sorbent) is not found to be constant on different subsurface materials, nor is it quantitatively predictable from the octanol-water partition coefficient (K_{ow}). A significant fraction of the organic carbon in low-carbon subsurface materials is believed to exist in the adsorbed state on the surface of mineral particles. Further, the variation in K_{oc} that occurs between different subsurface materials is hypothesized to be due to differences in the types and chemical properties of the adsorbed natural humic substances and differences in the mineral surfaces to which they are bound. Research was initiated over this last year to evaluate the above hypotheses.

Research in this topical area is expected to continue for approximately 2 years and involve collaborative work with Dr. J. McCarthy of Oak Ridge National Laboratory. It is designed to determine 1) the extent of adsorption of humic substances on mineral solids common to groundwater and 2) the effects of these adsorbed humic substances on the co-adsorption of hydrophobic organic contaminants. In FY 1988, the adsorption of reference humic substances (humic and fulvic acids, obtained from the International Humic Substances Society) was investigated on kaolinite and hematite. The humic substances were chosen to bracket properties commonly observed in groundwater-dissolved organic carbon. The reference minerals (kaolinite and hematite) exhibit surface chemical properties typical of subsurface sorbents. Carbazole was selected as the probe

TABLE 1. The Acid Dissociations Constants ($\log K_a$)

% MeOH (w/w)	$\log K_a$			
	AN	4-MA	3, 4-DMA	2, 4, 5-TMA
0	-4.565 $\pm$ 0.007	-5.050 $\pm$ 0.001	-5.153 $\pm$ 0.022	-5.012 $\pm$ 0.006
10	-4.572 $\pm$ 0.008	-5.059 $\pm$ 0.002	-5.172 $\pm$ 0.003	-4.989 $\pm$ 0.008
20	-4.511 $\pm$ 0.006	-5.014 $\pm$ 0.005	-5.140 $\pm$ 0.005	-4.967 $\pm$ 0.003
40	-4.329 $\pm$ 0.034	-4.898 $\pm$ 0.026	-4.978 $\pm$ 0.018	-4.846 $\pm$ 0.005
60	-4.101 $\pm$ 0.028	-4.645 $\pm$ 0.011	-4.841 $\pm$ 0.011	-4.737 $\pm$ 0.014

selected as the probe molecule to determine the effect of adsorbed humic substances on hydrophobic contaminant adsorption. Plans call for similar investigations using a broader range of humic substances (humic, fulvic acids), mineral solids (illite, calcite), and contaminant molecules (anthracene, dibenzothiophene).

Humic substances were found to adsorb significantly on subsurface mineral surfaces (Figure 1). Adsorption of both humic and fulvic acids on hematite and kaolinite increased with decreasing pH as expected for an acidic, anionic sorbate adsorbed by ligand exchange. The ligand exchange reaction may be generalized as follows:

where SOH is a hydroxyl surface site on the solid. The increased development of positive charge on the iron oxide and kaolinite at lower pH also assists in the sorption of the humic substances. The humic substances were adsorbed more strongly on the iron oxide, which contains more surface hydroxyl sites than the kaolinite. The relative affinity of FeOH sites on iron oxide, versus AlOH sites on kaolinite, for the humic substances cannot be determined from these data alone. The higher molecular weight, more hydrophobic humic acid was adsorbed more strongly (on a per C basis) than was the humic acid. These results suggest that these and comparably behaved

mineral surfaces may be partially or fully covered with adsorbed natural organic substances in groundwater systems.

The adsorption of humic acid on both hematite and kaolinite increased the co-adsorption of the hydrophobic compound carbazole (Figure 2a). In the absence of adsorbed humic substances there was virtually no adsorption on either mineral sorbent. Percentage carbazole sorption increased with increased adsorption of the humic acid. The humic acid exhibited a different affinity for carbazole when it was adsorbed on hematite or kaolinite. The humic acid appeared to be more accessible to or more selective for carbazole when adsorbed on hematite. These results suggest that the physical structure of adsorbed humic acid may vary on different adsorbents, and that this structure affects the subsequent sorption of hydrophobic contaminants.

The binding coefficient of the adsorbed organic carbon phase (K_{oc} , where K_{oc} equals the macroscopic coefficient, K_p , divided by the mass fraction of organic carbon in the humic coated sorbent) for carbazole differed between sorbents and was higher for humic acid adsorbed on the hematite (Figure 2b). The K_{oc} on both sorbents was relatively insensitive to the degree of surface coverage by the adsorbed humic acid. The K_{oc} estimated from the octanol-water partition coefficient (K_{ow}) has been shown to adequately

FIGURE 1. Adsorption of Humic Acid (HA) and Fulvic Acid (FA) on Subsurface Mineral Surfaces

FIGURE 2. Co-adsorption of Carbazole with Humic Acid (HA) on Hematite and Kaolinite

the K_{ow} estimation procedure significantly underestimates the selectivity of adsorbed humic substances for hydrophobic contaminants, especially when they are adsorbed as thin layers on mineral surfaces. Use of the K_{ow} relationship in predictive modeling for low-carbon groundwaters would significantly overpredict the mobility of the hydrophobic contaminant if the organic carbon phase behaved as shown in Figure 2b. We propose that sorptivity of different humic substances adsorbed to different mineral surfaces must be understood

before accurate predictive models of subsurface adsorption can be developed for hydrophobic compounds.

Sorption-Degradation Interactions

The sorption of organic compounds on surface and subsurface materials influences the transport of the organic contaminants. It is becoming increasingly clear, however, that sorption may also influence the persistence and form of organic

contaminants. Sorption may adversely affect microbial degradation rates if compounds are unavailable as a carbon source in the sorbed state, or sorption processes may enhance or cause abiotic degradation of organic contaminants through surface catalysis. Alternatively, the two processes, biotic and abiotic degradation, may interact in a synergistic or antagonistic manner. Regardless of the process, sorption-degradation interactions are chemically complex. Work, initiated previously, continued in FY 1988 to better understand the interactions of sorption with 1) abiotic degradation and 2) microbial degradation.

Abiotic degradation. Previously, aminonaphthalene was shown to undergo a surface-catalyzed transformation at the surface of subsurface materials; their clay fraction isolates and pure montmorillonite culminating in the formation of unidentified higher molecular weight compounds. It was hypothesized that aminonaphthalene ($pK_a = 3.92$, solubility ~ 1700 ppm) sorbed to clay surfaces through a cation exchange mechanism

exchange mechanism and that a surface-catalyzed oxidation of the sorbed aminonaphthalene, and concomitant reduction of clay lattice ferric iron, resulted in a surface-stabilized radical cation. Subsequent interactions between the oxidized surface species with aqueous aminonaphthalene or other natural organic compounds produced higher molecular weight compounds. The rate of transformation was shown to depend on the pH and the aminonaphthalene and clay concentrations. FY 1988 research continued the investigation of aminonaphthalene surface-catalyzed degradation by clay minerals to isolate and understand the individual reactions that compose this complex interaction and its importance in the fate of organic compounds in subsurface environments.

Sorption of aminonaphthalene in a aqueous suspension of sodium-hectorite, a pure trioctahedral smectite with no ferric iron substitution, reaches equilibrium rapidly and without any evidence of transformation (Figure 3). Additionally, as a

FIGURE 3. Solution and Clay Surface Concentration of Naphthylamine in Selected Clay Suspensions with Time at pH 4.5 and an Initial Naphthylamine Concentration of $1 \times 10^{-4} M$

function of pH, sorption parallels the ionization fraction (I.F.) of aminonaphthalene:

$$\text{NA} + \text{H}^+ = \text{NAH}^+; \text{pK}_a = -\log[(\text{NA})(\text{H}^+)/(\text{NAH}^+)] \quad (1)$$

$$\text{I.F.} = [\text{NAH}^+]/[\text{NA} + \text{NAH}^+] = 1/\{1 + K_a/[\text{H}^+]\} \quad (2)$$

where NA and NAH^+ are the protonated and neutral aminonaphthalene species, and () denotes activities. Additionally, the initial sorption (5 min) of aminonaphthalene on montmorillonite and its subsequent rate of transformation decreases with increasing pH. These data strongly support the hypothesis that the sorption process is a cation exchange mechanism, with the protonated aminonaphthalene replacing Na^+ on the surface:

$$K_c = [(\text{Na}^+) \text{NXNAH}/(\text{NAH}^+) \text{NXNa}] \quad (4)$$

where K_c is a conditional exchange equilibrium constant and N is surface mole fractions. The K_c for aminonaphthalene exchange on hectorite (at pH 4.5) was calculated to be approximately 94, which means the aminonaphthalene cation is preferred by a factor of 94 over Na^+ . If, however, ferric iron is placed on the surface of hectorite as an exchangeable cation, aminonaphthalene is rapidly taken up and transformation is observed in a similar fashion as that seen in the Na-montmorillonite suspension (Figure 3). Ferric iron, either as an exchangeable cation (as with the hectorite) or as a part of the clay lattice (as with montmorillonite) is a necessary component for the transformation of aminonaphthalene; Fe^{3+} probably acts as an oxidant to form a surface-stabilized aminonaphthalene radical cation. The clay mineral surface is also necessary because the reaction is not observed in aqueous solution.

The major transformation products appear to consist of four compounds that have the same molecular ion mass to charge ratio (m/z) of 284 (as determined by gas chromatography/mass spectrometry [GC/MS]), suggesting that they are structural isomers of one another. These products are observed in a montmorillonite suspension regardless of pH, and in the ferrated hectorite suspension. In an attempt to determine a possible structure for the transformation products, a deuterium oxide (D_2O) exchange study was carried out. Because protons are more or less labile depending on their bonding environments, aminonaphthalene (Figure 4a) and diphenylamine (Figure 4b) were mixed with D_2O at 50°C until the protons associated with the nitrogens

exchanged with deuterium; deuterium is two times the mass of hydrogen. Exchange was determined by observing the mass change of the molecular ion by mass spectrometry. Complete exchange took 7 and 30 days for aminonaphthalene and diphenylamine, respectively (Figures 4a and b). Transformation products, removed from the clay surface, showed a mass increase of 2 units after about 10 days in D_2O and evidence of a 3-unit increase after 30 days. This increase suggests that the products have a primary amine (similar to aminonaphthalene) and a secondary amine (similar to diphenylamine); therefore, the structure (284 mass units, Figure 4c) is probably one of the transformation products; the other three products are probably structural isomers of this compound.

A simplified model of aminonaphthalene transformation based on the above data is 1) sorption through cation exchange, 2) production at the surface of a radical cation by reduction of surface-sorbed or clay lattice ferric iron, 3) interaction between the surface-stabilized radical cation and a neutral aminonaphthalene, and 4) appearance of the reaction product on the clay surface. The importance of this sorption-degradation interaction is not the products hypothesized from these simple clay systems, but 1) that their formation is probably through a radical-cation mechanism that is capable of reacting with both natural and anthropogenic organic compounds present in the subsurface and thus able to form a variety of products, and 2) the sorption-degradation process removes aminonaphthalene, a mobile and carcinogenic compound, from the environment.

Microbial Degradation. Quinoline, a two-ring ionizable N-heterocyclic compound ($\text{pK}_a = 4.92$), is rapidly degraded by a bacterium isolated from the deep subsurface. However, quinoline is strongly sorbed by cation exchange to subsurface materials and pure clay minerals such as montmorillonite; the conditional exchange constant (K_c) for quinoline exchange with Na-montmorillonite varies from about 200 to 1200 depending on the surface concentration ($K_c = [(\text{Na}^+) \text{NXQH}/(\text{QH}^+) \text{NXNa}]$, QH is the ionized quinoline cation, N is surface mole fractions, and () denotes activities). This strong interaction between quinoline and clay suggests that if an organism can mineralize only the quinoline in solution, then use of the compound (as measured by CO_2 evolution) could be considerably reduced compared with degradation when clay is not

FIGURE 4. Deuterium Exchange of Amine Hydrogens on a) Aminonaphthalene, b) Diphenylamine, and c) Proposed Structure of Transformation Product

present. This work is undertaken in conjunction with Dr. R. Hicks, a microbiologist in PNL's Terrestrial Sciences Section.

The degradation rate of quinoline was measured in a sterilized mineral nutrient solution and an identical sterilized solution with montmorillonite added; quinoline, at a total concentration of 1×10^{-6} M, was added, and after 24 hr, each was inoculated with approximately the same number of organisms (108 cells/mL), and CO_2 monitored over a 48-hr period. The degradation of quinoline, in pure culture, evolved CO_2 rapidly over the first 16 hr, indicating almost total removal and mineralization of the added quinoline (Figure 5). The clay system sorbed about 60% of the quinoline before inoculation, and the rate of quinoline mineralization in this suspension was much reduced compared with the nonclay system (Figure 5). Previous work by Hicks had demonstrated that the rate of mineralization does not depend on the concentration of quinoline when quinoline is present between 10^{-4} and 10^{-9} M.

Clearly, the presence of the clay is causing a reduction in the rate of mineralization of quinoline. However, the original hypothesis was that the clay would affect the rate of mineralization only after the solution concentration of quinoline was exhausted; beyond this point it was felt that the desorption of quinoline from the clay would limit the mineralization if desorption was slower than the organism's ability to use solution quinoline. If the above simple hypothesis were correct, then coincident rates of mineralization should have been observed until the solution concentration of the clay system became dependent on the desorption of quinoline from the clay surface; at this point, the two rates should diverge and the clay system's mineralization should slow. This is clearly not the case; the clay system's mineralization is slowed from the outset compared with the nonclay system.

Although desorption of quinoline is apparently not the mechanism involved in the shift in mineralization rate, it cannot be ruled out as a contributing factor. Indeed, the change in rate observed

FIGURE 5. Quinoline Mineralization Rate as Influenced by the Presence of Na-Montmorillonite

after about 18 hr may be due to desorption (Figure 5), but there is another mechanism involved that is also probably related to the presence of the clay. A tentative hypothesis, now being tested, is that the clay is binding to the organism (the clay particles are approximately 10 times smaller than the microbial organism), and affecting its ability to take up quinoline from solution. Importantly, both mechanisms could have significant impact on the degradation of organic contaminants in the subsurface environment because clay colloids moving with the water could bind to organisms. This work is continuing in FY 1989 with both static experiments, as described above, and dynamic studies.

Geochemical Research Supporting OHER's Deep Microbiology Program

Research funds were redirected from this program to support OHER's Deep Microbiology Program. Two activities were performed: 1) development of a plan for groundwater dating at Savannah River Project (SRP) to assist in age determinations of the microorganisms and 2) design and initiation of GEMHEX on the fourth deep borehole at SRP.

PNL developed a sampling plan and established university contacts to allow ¹⁴C and ¹³C isotopic analysis of dissolved inorganic carbon, dissolved organic carbon, and specific chemical fractions of the dissolved organic carbon fraction in SRP groundwaters for determination of groundwater age and source location of the dissolved organic

carbon phase. This information is needed to understand the origin of microbes in deep groundwaters at SRP and their role in carbon cycling. A series of wells will be sampled in the Middendorf Formation, including the previous microbiology boreholes and others both up- and downgradient. This sampling will be done cooperatively with SRP and state personnel from South Carolina in November. PNL will prepare the samples and interpret the data; the accelerator mass spectrometry for the isotopic analyses will be performed at the University of Arizona.

The GEMHEX experiment developed out of a perceived scientific need on the first three boreholes to establish the relationship between fine-scale variation of physical and geochemical properties within a given aquifer unit and the number and distribution of microorganisms. To this end, a statistical design was developed for an experiment on the fourth deep microbiology borehole. This design consisted of obtaining and characterizing three distinct continuous 15-ft core intervals located near group sample locations in the Middendorf and Cape Fear formations. More elaborate geochemical and hydrological measurements will be performed on the core samples than were performed on the first three boreholes. These measurements in conjunction with extensive measurements of pore water chemistry will help answer important questions concerning possible contamination by drilling muds and hydrological and geochemical influences on microorganism

distributions. The cores were obtained in September 1988, and the characterization activities, beginning with pore water displacement and microbiological enumeration, began immediately. Further analyses and data evaluation and interpretation will be completed in FY 1989.

Subsurface Behavior of Organic Fluids

C. S. Simmons, J. W. Cary, and J. F. McBride (PNL) and J. C. Parker (Virginia Polytechnic Institute)

Research Directions

The key to controlling and remediating subsurface contamination by immiscible organic liquids is the ability to predict the physical behavior of these liquids in porous media for vadose zone conditions. Thus, this project is providing essential information for development of predictive models of organic liquid flow behavior in the subsurface environment. Intermediate-scale laboratory experiments, using a selected set of representative organic liquids spanning a diverse range of physical properties, are examining hypotheses about the physical mechanisms that control immiscible and multiphase flow behavior in subsurface media. An improved multiphase theory is needed to quantify the complex simultaneous transport of water, air, and organic liquids in subsurface media.

FY 1987 research focused on developing an improved mathematical model for describing the three-phase conduction of water, organic liquids, and air in granular porous media. The model is based on a pore-scale capillary conduction theory, which uses only fundamental physical parameters and the measured soil-water retention relation for the particular medium. A basic hypothesis being examined is that the fluid-media scaling principle is adequate to predict the conduction of each immiscible liquid from measured interfacial surface tension and viscosity parameters. This hypothesis and laboratory experiments to test it have been explained in three research papers accepted for publication in professional journals. A synopsis of the work was given in a previous report (Simmons et al. 1988). The theory proved successful when predicting the entry of an organic liquid into previously water-wetted porous media (sand, loamy sand, and silt loam). However, it could not account for how rapidly water infiltrated into an oil-wetted porous medium. Additional theoretical and experimental efforts were undertaken to explain this rapid water

infiltration phenomenon, which may have considerable influence on how quickly soluble organic components are carried away by water passing through an organic contamination source.

Past research covered the study of immiscible flow behavior only in one-dimensional columns containing the example porous media; the actual flow behavior in a three-dimensional porous system will be examined during FY 1989. To study three-phase flow in a more realistic three-dimensional domain, specially instrumented flow cells will be used (Figure 1). In FY 1988, the emphasis was on developing and testing the special instruments needed to measure the physical variables of fluid content and pressure governing the flow processes that would be observed in a flow cell. Cooperative research with Virginia Polytechnic Institute (VPI) has also produced a two-dimensional finite element three-phase flow code that will be used to test the fluid conduction theory by comparison with experimental data obtained in the flow cells. The code is formulated in radial coordinates to make a three-dimensional comparison possible, while using only a two-dimensional code. Infiltration experiments performed with radial symmetry make the comparison possible.

Special Instruments

Three instrumentation systems are being developed for measuring liquid pressures in each phase and liquid contents in situ. The first system consists of hydrophobic and hydrophilic tensiometers connected to precision pressure transducers. Pressure readings are taken automatically by a computer data acquisition system, which continuously records pressure changes. The novelty of the hydrophobic-type tensiometer is that it uses a porous polyethylene probe that excludes water and allows liquid connection to only the organic phase. A standard hydrophilic probe, on the other hand, connects to only the water phase. Together the two kinds of tensiometer probes make possible the measurement of capillary pressure between the water and organic liquid interfaces.

A second system that applies a newly invented concept uses electro-optical liquid detectors to sense the change in either water or organic liquid content in situ. This new detector, submitted for patent, operates on the degree of light transmission through a translucent porous material containing an imbibed liquid. As the amount of imbibed liquid increases, the transmission of light

FIGURE 1. Design of Experimental Flow Cell for the Study of Three-Dimensional Multiphase Flow. An oil infiltration experiment is shown.

through the translucent material also increases. The electro-optical device is made specifically sensitive to water or organic liquid by using either a ceramic or porous polyethylene material, respectively. It is constructed with a simple LED light source that is detected by a solid-state photoresistor placed opposite the LED with the translucent material sandwiched between. This inexpensive and simple to construct device is expected to revolutionize the in situ detection of organic liquids. In conjunction with inventing the electro-optical probe, experiments demonstrated the sensitivity of the detector to varying amounts of liquid in a surrounding porous medium. This probe uses a totally new approach to liquid pressure measuring. A manuscript on the functional aspects of the electro-optical probe was submitted to the *Soil Science Society of America Journal* for publication.

The third instrument system being assembled is a standard gamma radiation attenuation scanner

for determining fluid contents in situ. By using two distinct radiation energy beams, it is possible to assess the amounts of water and oil present in a porous material, provided the material has a sufficiently thin cross section (limited to a maximum of 15 cm). Technology for such a device is well known and in common use. However, these flow cell experiments call for using the radiation attenuation method in a novel fashion. A single radiation beam source mounted on an automatic vertical-plane scanner table will be used to map only the total liquid content distribution during infiltration of an organic liquid plume. This approach allows the nondestructive sampling of a cross section greater than 15 cm. A conceptual drawing of such an experiment is also shown in Figure 1. The radiation scanner and electro-optic sensors will be used together to map a spreading oil plume over time. Location data derived from such an experiment are adequate to test the predictive capability of the

three-phase flow code that embodies the hypothesized conduction theory based on Darcy's law.

Experiments

The key to making multiphase flow theory a predictive tool is being able to obtain two constitutive properties that characterize the physical processes of fluid conduction in porous media: fluid retention relations (capillary pressure saturation curves) and permeability saturation functions. This project emphasizes developing and proving capillary theories for obtaining the constitutive relations required to describe the simultaneous flow of water, organic liquid, and vapor in subsurface media; such systems are called three-phase flows. The instruments and measurements previously described are essential in performing the research.

The hydrophobic/hydrophilic tensiometer system is used to measure the capillary pressures (forces) that cause absorption of liquids in porous media. This year the tensiometer system was used in two basic experiments that helped clarify three-phase flow mechanisms. First, a hydrophobic tensiometer was used to make novel measurements of oil [Soltrol-220 (Phillips Petroleum Company, Bartlesville, Oklahoma) and diesel fuel] spreading pressure on a free water surface. This spreading pressure was found to be as much as 100 times larger than theoretical calculations suggest. This pressure is caused by interfacial-tension-related forces that cause certain oils to spread on water surfaces. A hypothesis is that spreading pressure contributes substantially to in situ pressure measurements in unsaturated media. The pressure gradient produced by surface spreading may be responsible for more rapid imbibition of oils than is predicted by prevailing multiphase constitutive theory. A more rapid imbibition was observed in the previous year's column infiltration experiments; however, the discrepancy remains unexplained.

A second set of experiments on water and oil infiltration into a long column used the tensiometer system for additional studies of the rapid, unstable entry of water into a porous sand medium, which was previously infiltrated by Soltrol oil. Previous infiltration experiments reported last year showed that water entered an oil-wetted porous medium much more rapidly than theoretical equations for permeability predicted. The hypothesis being tested is that the wetting order assumption, which is implicitly built into the fluid-media scaling theory for constitutive relations,

breaks down temporarily while water flow is rapid. These experiments are being used to revise capillary flow theory for more general circumstances. Cary et al. (1989) provide comparisons of data and computer modeling results that confirm the inadequacy of prevailing constitutive relations.

Figure 2 demonstrates the response in time of a hydrophobic tensiometer at a 4-cm distance from the input end (top) of a column filled with the sand. Four hours after a pulse of Soltrol oil was added, water was applied. The peak shows a sharp increase in oil pressure, becoming more negative (decreasing pressure) as the water passed by. A negative pressure indicates how strongly oil is held in the sand. The increase in oil pressure reflects its dependence on the total liquid content of both water and oil together, in conformity with theory. Another plot, not provided, showed that the water pressure 2 cm from the column's top did not increase as it should have if the liquid wetting order within pores held true to theory. After the water pulse passed, the return of oil flow to a drainage pattern was also evident as the pressure again decreased. The measurements in Figure 2 show the operation of a hydrophobic probe.

Another device used to measure two-phase retention and conduction of immiscible fluids in porous media is the permeameter cell. A system of these experimental cells was previously assembled through cooperation with VPI researchers. By performing displacement outflow experiments using these cells, the predictive accuracy of multiphase flow codes for the simplest one-dimensional case can be tested. Experiments this year have led to the idea that the concept of the residual saturation should be modified in capillary conductivity equations. It was found that the displacement of water from the sand by Soltrol could not be reasonably matched in terms of the outflow quantity by code predictions unless the residual saturation value was allowed to differ for the retention curve and the two-phase relative permeability functions. This use of different values of residual saturation is referred to as "uncoupling the residual saturation parameter in capillary models of conductivity."

Three-Phase Conduction Experiments

Direct measurements were made of three-phase conductivity for water and oil with air present to test the scaling of conductivity with the viscosity ratio. Previously, the hypothesized capillary model for three-phase conductivity could only be

FIGURE 2. Response of a Hydrophobic Tensiometer Measuring Soltrol Pressure During Infiltration of Water into the Sand Column at 4.1 cm

verified indirectly by comparing computer model simulations with observed infiltration behavior. Only the two-phase flow situation has been tested in the permeameter cells. The purpose of this work is to determine which of two theoretical capillary conductivity models, derived respectively by VPI and PNL researchers, is more accurate for a variety of porous media (i.e., silt loam, loamy sand, and sand). For ideal porous media, it should be possible to define an intrinsic permeability and a relative permeability function that applies regardless of what immiscible fluids are involved. However, these conduction experiments confirmed that soil swelling effects and the gas-slippage effect (Klinkenberg effect) could not be neglected in theoretical conductivity models. In particular, porous media other than the sand are subject to substantial swelling, which is manifested in such a way that the saturated conductivity is less for water than for oil or air on an equal viscosity basis. So far, this has been proven true only when scaling is used to compare fluids on an equal viscosity basis. The saturated conductivity determines how rapidly a fluid can flow through a porous medium when the pores are entirely filled. On the other hand, air flows more rapidly than oil as an immiscible fluid through equal pore volumes in an unsaturated porous medium, provided the fluids are scaled to have equal viscosity. The augmented flow rate of air is caused by the Klinkenberg effect, which

must be corrected for to use air for determining permeability.

This work shows that the same relative conductivity function is not obtained irrespective of the particular fluid involved. Based on the three-phase conductivity data for many combinations, a correction factor was found to account for swelling and gas slippage. Using a correction factor, it is possible to achieve a good correlation of observed and predicted fluid conductivity over all immiscible fluids and test porous media, as shown in Figure 3. In Figure 3 the predicted conductivity is based on the theory put forth by PNL researchers. Low conductivity is generally associated with mineral oil, whereas high values are for air. The data range over seven orders of magnitude and indicate the variability that must be accommodated in a multiphase flow model.

Two-Dimensional Simulations

Cooperative research with VPI under subcontract with PNL to develop a two-dimensional three-phase flow code was completed. The work fostered the inclusion of a hysteresis capability in a finite element code previously developed and published by VPI. Hysteresis was included in the code to simulate the first two-dimensional flow cell experiment performed by H. O. Schiegg (Schiegg and McBride 1987). The experimental set-up is

FIGURE 3. Correlation of Observed and Predicted Fluid Conductivities for Air and Oils in Three Soil Types. Predicted conductivity for immiscible fluids is based on the PNL model, which is corrected for soil swelling and air-slippage effects.

summarized in the above reference. A radially symmetric version of the code was also produced and used to obtain preliminary simulations of infiltration/redistribution experiments (Figure 1) to be performed by PNL in FY 1989.

Figures 4a and b show water saturation distribution with profile depth and total liquid saturation at two locations in Schiegg's experimental system after 1 day. The area between the curves is the infiltrated amount of oil. Dashed curves show the corresponding simulation results. The considerable discrepancy between observed and predicted results indicates that in the simulation oil reached bottom even 60 cm to the side of the flow cell's center, whereas the data show that no such movement occurred (Figure 4b). A hypothesis is that the retention relation does not accurately describe the capillary fringe, which has apparently kept the oil up near the cell's top. However, the data and corresponding pictures of the plume developed in the cell suggest that the particular sand profile was not as homogeneous as believed. Perhaps unintended layers were produced

in the cell's sand filling process. These layers may have caused uneven lateral flow (Schiegg and McBride 1987). For instance, unexplained repeated bulges appear in the observed center water-saturation profile. To obtain the smooth simulation results shown in Figures 4a and b, a constraint on the fluid-media scale factors was necessary. Otherwise, the simulated saturation profiles near the sides of the cell would have displayed unusual and most likely false sharp bends. In summary, the required constraint on scale factors for oil-water and air-oil fluid interfaces does not currently account for contamination of water interfaces by oil. This contamination process changes the scale factors locally from the assumed constant input values. In any case, the simulation shows that the effect of change in oil-water interfacial tension caused by oil contamination needs to be better accounted for in the computer model. The lack of conformity between the simulation and experiment is expected to motivate model improvements in future studies.

FIGURES 4a and b. Comparison of Schiegg's Results (Experiment No. 1 at Day 1) With Simulations Performed by VPI; 4a is at the Center of the Container, 4b is on the Side of the Container, 60 cm from Center. Shown are distributions of water and total liquid saturation, 1 day following infiltration of an oil into an unsaturated sand profile.

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Radon Transport Modeling in Soils

P. C. Owczarski, H. D. Freeman, D. J. Holford, and G. W. Gee

This new project is focused on understanding the dynamics of the major source of indoor radon: soils around buildings. Thus, the objectives are to construct and apply a comprehensive

computer-based model of radon transport within and from soils into the atmosphere and structures.

The model will solve all pertinent unsteady-state differential equations governing the transport of radon, soil gas, liquid and vapor water, and energy. The model will be three dimensional, unsteady state, and multiphase. The principal outputs of the model will be unsteady-state fluxes and concentrations of radon gas at the soil boundaries. The model will be validated as far as existing and near-future data allow.

The applications of the model will include 1) systematically studying the relative importance of radon transport mechanisms, the sensitivity of controlling parameters, and the uncertainties in the model outputs caused by uncertainties in the parameters and the presence of soil heterogeneities; 2) developing criteria for soil measurements related to radon transport; 3) linking up to a building transport and exposure model for estimating population exposures; and 4) providing a model to correlate soil properties with aeroradiometric surveys and to study the effects of changes in meteorology on radon transport.

Radon Transport Properties Database

A database of information pertaining to radon transport through and from soils is being

developed to help validate the comprehensive radon transport model being developed. The database will include information on soils, meteorological conditions, and structure (building) properties obtained from a literature review. Pertinent data will also be gleaned from ongoing radon programs, including other OHER-sponsored programs, as the information becomes available. The information will be used to develop credible scenarios to define the input parameters for modeling exercises. The model output will be compared with available data to determine the validity of the modeling approach. A review of the information in the database will identify deficiencies, which is necessary to fully validate a comprehensive radon transport model.

The radon transport data were broken up into several records to allow the data to be entered and retrieved in logically related blocks. The record types identified as important are

Record	Description
REF	Reference information
SOIL	Soil properties
GSOIL	Generic soil properties
PORE	Soil pore space properties
SURFACE	Soil/air interface properties
HETER	Heterogeneities in soil (e.g., cracks)
STRUCT	Structure properties
MET	Meteorological conditions
PROJECT	Information on ongoing radon projects

A literature search was conducted on the Dialog Information Retrieval Service to identify references relating to radon transport in and from soils. The literature search revealed several hundred references with potential application; a further review of abstracts narrowed the list to approximately 250 references. To date, approximately 200 have been received.

FY 1989 activities will focus on entering data from the references and ongoing projects into the database. If necessary, reference data that are presented only in graphs will be digitized. Report formats compatible with the model input files will be developed.

A Finite Element Model of Radon Advection and Diffusion in Unsaturated Cracked Soils

D. J. Holford, G. W. Gee, P. C. Owczarski, and H. D. Freeman

Model Development

A three-dimensional finite element code has been developed to simulate the effect of changes in surface air pressure on radon flux from unsaturated soil with parallel, partially penetrating cracks. The governing air flow equation assumes transient, compressible, ideal gas flow. The radon transport equation includes advection through the air in unblocked pores, diffusion in the air-filled and water-filled pore space (including Knudsen diffusion in extremely small air-filled spaces), radioactive decay, and radon emanation from the soil. A single macroscopic radon diffusion coefficient and effective air-filled porosity are estimated from the moisture content and the pore-size distribution of the soil by considering the soil pores to be composed of all possible serial combinations of the pore-size increments (Nielson et al. 1984). The pore-size distribution can be estimated from measured soil-water retention characteristic curves or from sieve analyses and bulk densities (Arya and Paris 1981). Soil permeabilities at different moisture contents are predicted from the soil-water retention curve by a functional relationship (van Genuchten 1980). Radon emanation is assumed to increase linearly with moisture content from some rate at dryness until a plateau value is reached, at which point it remains constant until the material is saturated (Nielson et al. 1982).

Sensitivity Analysis

A sensitivity analysis has been used to investigate the effect of changes in crack geometry, soil parameters, and surface pressure changes on radon flux to the atmosphere from an unsaturated, cracked soil (unpublished data). All simulations were run for a period of 6 hr, and the base case parameters are shown in Table 1.

TABLE 1. Base Case Parameters for Sensitivity Analysis

Parameter	Value
Air dynamic viscosity	1.8×10^{-4} g/cm/s
Radon diffusion coefficient in air	0.1 cm ² /s
Radon decay constant	2.1×10^{-6} /s
Radon production rate	5.25×10^{-2} atoms/cm ³ /s
Depth to water table	10 m
Crack depth	400 cm
Crack spacing	800 cm
Crack width	0.06 cm
Soil porosity	0.35
Soil intrinsic permeability	2.7×10^{-8} cm ²
Tortuosity	0.26
Radon diffusion coefficient in soil	2.6×10^{-2} cm ² /s
Initial atmospheric pressure	85 kPa
Change in atmospheric pressure per unit time ($\Delta P_{\text{atm}}/\Delta t$)	-0.09 kPa/hr
Time increment (Δt)	900 s

Radon flux generally increases with increasing crack width, length, and depth. However, radon flux reaches a plateau value with increasing crack depth because radon concentrations approach a maximum value with depth (Figure 1). Radon flux can actually decrease if cracks are spaced too closely (Figure 2).

Radon flux also increases with 1) increasing median pore diameter, 2) decreasing soil moisture content, and 3) decreasing standard deviation of the soil pore-size distribution. Increasing the median pore diameter and decreasing the soil moisture content both increase the air-filled porosity of the soil. Increasing the air-filled porosity of the soil causes an increase in the diffusive flux from the soil simply because a larger proportion of soil volume is filled with the same concentration of radon. Increasing the air-filled porosity of the soil also causes an increase in the advective flux from the cracks because that increases pressure gradients in the soil toward cracks, driving more radon into the cracks. Pressure gradients in the soil are directly proportional to the porosity, because increasing porosity increases the gas storage capacity of the soil, and pressure changes do not move as far down into the soil. Decreasing the standard deviation of the soil pore-size distribution decreases the tortuosity of the soil. Decreasing the tortuosity of the soil results in a higher diffusion coefficient for the soil and hence a higher radon flux.

Radon flux increases with increasing rates of falling atmospheric pressure (Figure 3). A net

FIGURE 1. Effect of Crack Depth on Percentage Increase in Radon Flux with Respect to a Homogeneous Soil

FIGURE 2. Effect of Crack Spacing and Width on Percentage Increase in Radon Flux with Respect to a Homogeneous Soil. Crack depth of 400 cm.

enhancement of radon flux occurs during sinusoidal variations in atmospheric pressure, such as those attributable to wind. However, the enhancement is greater for a soil with cracks than for an homogeneous soil (Figure 4).

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FIGURE 3. Effect of Different Rates of Increasing (+) and Decreasing (-) Atmospheric Pressure on Radon Flux from Cracked Soil

FIGURE 4. Effect of Sinusoidally Varying Atmospheric Pressure on Radon Flux from Cracked and Homogeneous (No Cracks) Soil

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Subsurface Microbial Phenomena

R. J. Hicks and J. K. Fredrickson

Previous investigations during this project have demonstrated that indigenous microbial populations have an important role in governing the persistence and transport of organic contaminants within subsurface environments. In addition, they may represent one of the only feasible methods to remediate organically contaminated wastes at depth. However, scientific understanding of the identity and function of microorganisms inhabiting depths from fifty to several thousand meters is incomplete. Therefore, fundamental investigations are being conducted at PNL to examine the microbial metabolic processes governing the behavior of aromatic compounds within deep subsurface environments and to examine the geochemical and hydrological properties that control microbial activity and diversity.

The mechanisms for biodegradation of contaminants and, therefore, contaminant persistence, depend on the type of electron acceptors available. Experimental studies in FY 1988 focused on microbial degradation reactions using O_2 (aerobic) and NO_3^- (facultative anaerobic) as the terminal electron acceptors. Aerobic and anaerobic biodegradation of ^{14}C -labeled aniline, aminonaphthalene, nitrobenzene, quinoline, and pyridine in deep (176 to 262 m) subsurface sediments underlying the coastal plain of South Carolina at Savannah River Project (SRP) were investigated. Results are shown in Figures 1 and 2. The extent of aniline and nitrobenzene

FIGURE 1. Mineralization of Nitrogen-Containing Homo- and Heterocyclic Aromatic Compounds in SRP Samples Incubated Under Aerobic Conditions

FIGURE 2. Biodegradation of Nitrogen-Containing Homo- and Heterocyclic Aromatic Compounds in SRP Samples Incubated Under Anaerobic Conditions

degradation was greater under anaerobic conditions (74% for aniline, 13% for nitrobenzene) than under aerobic conditions (53% for aniline, <1% for nitrobenzene). Quinoline was extensively degraded under both conditions. Pyridine was degraded mainly under aerobic conditions. Less than 5% of the added aminonaphthalene was degraded either aerobically or anaerobically. Biodegradation was not related with pH, Eh, clay content, pore water organic carbon, NO_3^- , or PO_4^{3-} ; however, there appeared to be an association between the extent of aerobic biodegradation of the compounds and populations of denitrifiers. Significant biodegradation occurred in only three of the six sediment samples examined, indicating spatial variability of microorganisms able to metabolize these compounds. The data indicate that deep subsurface environments support diverse microbial populations with the potential to degrade a variety of energy-derived aromatic compounds.

Specific biodegrading subsurface bacteria have also been isolated and examined to further define the metabolic capabilities of organisms indigenous to the deep subsurface. Two Gram-negative, motile bacteria, isolated from deep subsurface sediments, have been shown to mineralize quinoline under aerobic conditions and transform

quinoline to soluble intermediates under anaerobic conditions. In addition, these bacteria are capable of degrading other aromatic compounds, including naphthalene, biphenyl, benzene, 2-hydroxyquinoline, quinaldic acid, kynereenic acid, benzoic acid, salicylic acid, catechol, parahydroxybenzoic acid, protocatechuic acid, gentisic acid, and alpha and gamma resorcylic acid under aerobic conditions. These bacteria were also able to use quinoline at concentrations as low as $0.002 \mu\text{g mL}^{-1}$. Rapid aerobic mineralization of quinoline at concentrations as low as $0.002 \mu\text{g mL}^{-1}$ indicates these organisms possess a high-affinity uptake and use system, which would help them survive in oligotrophic environments such as the deep subsurface. Both bacteria harbored four plasmids of identical size, ranging from 50 to 440 kb, the consequences of which are hypothesized below.

Bacteria were isolated from the deep subsurface sediments and examined for plasmid content, antibiotic resistance, and minimum inhibitory concentrations (MIC) for Cu^{2+} , Cr^{3+} , and Hg^{2+} to study the physiology and genetics of subsurface microorganisms. The overall frequency of plasmids in bacteria isolated from the subsurface was 33% (Figure 3), which is considerably greater than the plasmid frequency observed in more

FIGURE 3. Overall Frequency of Plasmids in Bacteria Isolated from the Deep Subsurface Underlying SRP

shallow aquifers (usually <8%). This higher level suggests fundamental differences in the compositions of the bacterial communities of these two systems. Antibiotic resistance was most frequent to penicillin (70% of all isolates), ampicillin (49%), and carbenicillin (32%). The frequency of resistance to penicillin and ampicillin was significantly greater for isolates bearing plasmids than for those without plasmids. However, no relationship was obvious between the presence of plasmids in subsurface bacteria and MICs for Cu^{2+} , Cr^{3+} , and Hg^{2+} .

A high proportion (53%) of the plasmids associated with subsurface bacteria were larger than 150 kb. In addition, several of the subsurface bacterial isolates contain plasmids similar to plasmid DNA encoding the degradation of toluene (i.e., TOL plasmid).

One hypothesis to explain the high frequency of large plasmids and the homology of TOL plasmid DNA with the DNA extracted from subsurface isolates is that subsurface bacteria carry plasmids encoding for the catabolism of aromatic compounds. Organic compounds identified in groundwaters consist mainly (88%) of recalcitrant humic and fulvic acids, tannins, and lignins, all of which are condensed polyaromatic structures. As groundwater and associated dissolved organic carbon is separated in time and distance from the point of infiltration, compounds with more readily available carbon are metabolized, leaving the more recalcitrant humic and fulvic materials. It would therefore be advantageous for bacteria in deeper or more isolated sediments to harbor catabolic genes for aromatic organic fractions. The residence of these genes on plasmids would require that the genes be needed by the bacteria only occasionally and that the positive contribution from these genes outweigh their maintenance cost. Further work to test this hypothesis is being performed.

Integrated experiments, in collaboration with the geochemistry and hydrology components of the PNL Subsurface Transport Program, are being conducted using one-dimensional columns and a two-dimensional Subsurface Experimental

Research Facility (SERF) to describe better the role of microbiological activities on the transformation and persistence of organic compounds at a scale relevant to field conditions. Experiments in FY 1988 have focused on the relationship between sorption-desorption processes and biodegradation (see the section "Subsurface Chemistry of Organic Compounds" this volume), the construction of the columns and introduction of bacteria capable of degrading specific aromatic contaminants.

A model synthetic soil (composed of a chemically standardized montmorillonite clay bonded to alumina particles) with a particle size distribution and hydraulic conductivity of an actual soil is being used to rigorously define contaminant behavior in aquifer sediments. Initial range-finding experiments are being conducted with synthetic soil colonized with a quinoline-degrading bacterium in glass chromatography columns. Methods of introducing bacteria into the synthetic soil to obtain population densities, distribution, and a metabolic state similar to conditions existing in the subsurface environment are currently being investigated. Quinoline will then be supplied at a concentration and flux representative of a subsurface environment to investigate the chemical and microbial behavior of the system. These experiments will define conditions and parameters governing the transport and fate of organic contaminants in subsurface environments.

Future Research

Several new activities are planned, representing an evolution of concepts and past research. In particular, PNL will collaborate with the university community to further define the relationships between deep subsurface hydrogeochemical conditions and microbial distribution and function. This collaboration will also be used to examine the interactions between sorption and microbial degradation of organic compounds, and between radionuclides and complexing ligands, as these phenomena influence contaminant form and mobility. New emphasis will also be placed on a systematic evaluation of the effects of heterogeneity on key subsurface transport processes.

Marine Sciences

The goal of this research is to elucidate the molecular, biochemical, and cellular bases for marine invertebrate adaptation to environmental stresses that result from energy production. In particular, physical and chemical stresses can act synergistically with naturally present pathogenic microorganisms, or with latent cancer in the organisms to cause significant infectious diseases in marine animals. To understand the complex chain of stress interactions and adaptive responses at the organism level, it is necessary to study and evaluate the subcellular components of the responses. Thus, to facilitate this goal, the most applicable model among marine invertebrates to study these fundamental biological processes was selected in FY 1987. This model is hemic neoplasia of the bay mussel, *Mytilus edulis*, a proliferative or cancer-like process with similarities to leukemia in humans and other animals.

Environmental Stress and Disease in Marine Invertebrates

R. A. Elston

Research on Marine Bay Mussel Hemic Neoplasia

Hemic neoplasia disease of the marine bay mussel was discovered in Washington State in 1984. The condition is characterized by the replacement of normal blood cells or hemocytes by abnormal cells, resulting in the eventual death of the mussel. Similar diseases occur globally in a wide variety of bivalve mollusks. The possible induction and promotion of the diseases by environmental contaminants had been studied without definitive results for at least 10 years. PNL studies are the first to address the subcellular mechanistic processes in the disease. Once these processes are understood, it will be possible to experiment on the chemical induction and promotion of the disease. In FY 1987, a polyclonal antibody to the neoplastic or cancerous cells was produced as a preliminary step toward developing a means to study the transformation process of cells from the normal to neoplastic state.

During FY 1988, monoclonal antibodies to the neoplastic mussel cells were produced. The characterization of the antibodies is not yet complete, but at least two distinctive clones that react only with neoplastic cells and not with normal cells have been developed. By using a fluoresceinated second antibody, the abnormal cells were demonstrated to exhibit surface protein markers that are not present on normal cells. These markers suggest the expression of a non-host protein by the cells, related to the neoplastic transformation, that could result from a retroviral infection of the cells. The underlying cause of the cancer is, however, unknown at this point. The monoclonal antibody probes will now be used to

study the differentiation and transformation of the mussel's hemocytes.

Also this year, certain individual mussels were determined to be capable of causing the disease to go into remission, at least for 200 days, while others in the populations succumb to a progressive disease without any sign of recovery or remission. The remission and progression processes were studied by flow cytometric methods and revealed a repeatable pattern for both courses of the disease (Figure 1). In the progressive course, the DNA complement of hemocytes shifts to a hyperdiploid state. Subsequently, a large population of abnormal cells is

FIGURE 1. Composite Profile of DNA Content of Hemocytes of the Marine Bay Mussel Affected with a Leukemia-Like Disease, Hemic Neoplasia. S represents the haploid standard of 1; N represents the normal diploid DNA complement of normal blood cells. In represents the initiating event in the disease process or the hyperdiploid state; Si represents a signalling event that indicates the early progression of the disease; Ir represents the production of large numbers of neoplastic blood cells, an irreversible event; and T represents the last stage of the disease, in which blood cells with a DNA complement of about 10.4 times normal appear in the mussel's blood.

generated from the doubling of this hyperdiploid state; these cells have an abnormal DNA complement of about 5.1 times haploid (Peak 1r in Figure 1). Remarkably, these cells form the basis of another cycle for a population of cells containing a DNA complement of about 10.4. In the case of individuals that go into remission, the initial hyperdiploid state may occur, along with a small increase in peak 1r, but a large increase in this peak never occurs.

These results show that the development of this cancer in the progressive, fatal disease results from an ordered and repeatable replication pattern of abnormal cells with high aneuploid complements of DNA. The results also show that a state of remission occurs when this ordered abnormal pattern is interrupted at a relatively early stage. This interruption may occur through the recognition of the cancer cell antigens (determined in the monoclonal antibody studies) by certain immunocompetent individuals. It is also possible that the expression of surface antigens may not be uniform in the case of all infected cells, and thus some individual animals may not have the opportunity to recognize the foreign antigens.

Future Research

This continuing research will focus on hemic neoplasia in the mussel. Specifically, the

differential capacity of blood cells from certain individuals to recognize cancer cells will be compared with blood cells from other individuals that cannot recognize and dispose of these blood cells. These studies will be conducted in control test-tube experiments. Thus, considerable effort will be devoted to the maintenance of blood cell cultures in the laboratory, and tests will be conducted to compare the function of cells derived from mussels with the progressive disease and with the remissive disease. Monoclonal antibodies will be used to determine if any observed recognition processes can be blocked with, and thus identify, the important cell surface antigens in the recognition process. In addition, the flow cytometric methods used to characterize the cell cycling in whole animals will be used to determine the ability of the normal and cancerous cells to replicate in a test tube environment.

The longer-term goal of these studies is to use the mechanistic information gained in subcellular studies to understand the physiological organismic, and ultimately, population-level consequences of environmental stress and adaptation on marine invertebrates.

Theoretical (Quantitative) Ecology

Issues such as long-term monitoring, global change, and the environmental effects of hazardous waste sites illustrate the need for solid research progress in theoretical and quantitative ecology. Standard experimental designs, demographic theory, and methods of statistical estimation are generally not applicable for estimating the influence of natural and human-induced stresses on wildlife populations and plants. It is clear from the scope of the problems that technology must be developed to assess options at the landscape level. In addition, severe limitations exist in terms of lack of theory and suitable designs for sampling contaminants and populations in time and space. This program is directed at establishing a theoretical basis for landscape-level sampling as well as field designs, applying new paradigms, conducting simulation studies based on real data, and adapting concepts from other fields, such as sampling for modeling.

Several major new findings resulted from research conducted in the theoretical ecology program during FY 1988. These include:

- demonstrating that our method of determining ecological scales for studying ecological phenomena is unique, and unlike most other methods, is statistically sound
- developing a quantitative methodology to assess the effects of interventions (e.g., power-plant effluents or herbicide sprays) on the survival of birds or small mammals
- discovering that commonly accepted methods of predicting future large mammal populations will result in confidence limits that are far too narrow.

Designs for Environmental Field Studies

J. M. Thomas, L. L. Eberhardt, V. I. Cullinan, and M. A. Simmons

During FY 1988, research was conducted on ecological scale (sampling for pattern) and sampling to detect hazardous wastes (analytical sampling, sampling for modeling, and inventory). Ecological scale is defined as a given physical area in which some ecological process or characteristic has a statistically meaningful influence. Sampling for ecological scale, to detect global changes or for long-term ecological monitoring, will involve, at the least, landscape-level measurements. The optimal location, size, and spatial distribution of plots and transects where these measurements are obtained are critical to the success of such programs. Thus, as part of this ongoing PNL research on sampling for pattern, a major effort was initiated to derive optimal transect lengths and their landscape-level dispersion. The cover type examined for these studies of scale, bunchgrass, *Agropyron spicatum*, was measured both at the ALE Reserve and at the Idaho National Engineering Laboratory (INEL); the cover of a shrub, winterfat, *Eurotia lanata*, was also measured at INEL. These data were used to test statistical models of the strength of interrelationships between ecological processes. These models

define the most appropriate level of scale and thus can determine optimal transect size and dispersion over the landscape.

A literature review of quantitative methods currently used by landscape ecologists to assess environmental change was conducted. This review indicated that the PNL method of determining scales for studying ecological phenomena is unique. Most landscape ecologists use quantitative methods that give little attention to the size of transects or study plots relative to their ability to efficiently and accurately measure landscape characteristics or change. When attention is given to questions of scale, however, methods used to determine plot size are usually either not designed for such a purpose or have neither valid statistical nor scientific reasoning behind them.

For instance, based on observed plant cover, the optimal transect size for bunchgrass and winterfat was found to be about 100 m long. Such a transect is much longer than normally used or mentioned in the ecological literature. Most references cite cover estimates taken over a 1-m² area; rarely are they over 10 m². To estimate the optimal dispersion of these 100-m-long transects over the landscape, a unique correlation model was used. At INEL the processes that influence cover were different beyond 300 m for both

bunchgrass and winterfat. At ALE, this distance was more than 400 m (Figure 1). These distances define the environmental scale of influence, beyond which two points in the landscape no longer experience substantially similar habitat conditions.

This work on arid grassland species was extended to studies of the scale of mussel, *Mytilus californianus*, and subalpine fir, *Abies lasiocarpa*, distributions. Patterns observed in the estimated spatial variance and correlation patterns discovered in these studies were consistent with those observed in arid land bunchgrass distributions. The spatial variance increased with increasing transect lengths, and the spatial correlation decreased with increasing intersegment distance (i.e., the distance between transects). The distance at which spatial correlation is zero provides an estimate of ecological scale.

Based on the patterns in spatial correlation and variance for each of the three communities studied, the appropriate transect lengths and intersegment distances were determined to estimate the percentage cover of either bunchgrass, mussels, or subalpine fir. The efficiency of

these sampling strategies was then compared with standard or alternate sampling strategies. In all cases, sampling strategies based on these new variance and correlation methods produced estimates of percentage cover with generally smaller bias and smaller mean squared error. This process was repeated using statistically independent Poisson, negative binomial, and uniform random variables. The results showed that in the independent case (i.e., no correlation among simulated variables) only transect length affected efficiency.

In light of these results, it is apparent that spatial correlation and variance must be considered in the design of ecological research programs. However, the use of spatial correlation and variance for determining ecological scale does not address all problems associated with measurements obtained over landscapes. For example, the method does not, and was not designed to, determine the presence of multiple scales on a landscape. Although it is possible that Grieg-Smith's (1983) analysis of variance on varying plot sizes and spectral analyses could possibly meet this need, PNL research at a single-scale level suggests new methods will be needed. Because

FIGURE 1. Intraclass Correlation as a Function of Intersegment Distance Based on 128-m Transects. The results illustrated were calculated from cover measurements of 2000-m transects of wheatgrass, one on the ALE Reserve and one at INEL. A 2000-m transect of winterfat was also measured at INEL. The zero correlation point is the distance at which 128-m segments are independent of one another, or where the factors that affect cover are no longer similar. Dashed lines are 95% confidence limits about the zero correlation.

of this possibility, the effect of multiple scales on the design of sampling programs remains a fertile field for research. A second weakness of using spatial correlation and variance is that they only provide estimates of transect lengths and inter-segment distance in the form of ranges. For example, to maintain independence, the distance between wheatgrass transects must be in the wide range between 300 and 600 m. Finally, these results need to be extended to detect changes based on long-term landscape-level monitoring. This extension may involve no more than determining that a significant change in ecological scale has taken place (as a function of time). However, this concept is untested, and more importantly, a variance of the estimate of ecological scale would also need to be derived so a statistical test of change could be developed.

The second major area of research was directed to paradigms for sampling waste sites. Four classes of sampling were found that may be applied sequentially. Initial surveys may be conducted using an analytical approach, to determine whether there are substantial differences in sub-regions of a site. These differences, if found, can then be assessed in more detail, based on pattern sampling plans. Evaluation of the patterns thus found will then facilitate (and make cost-effective) any required sampling for inventory. In some instances, preliminary attention to the efficient estimation of model parameters (sampling for modeling) is needed to estimate movement of contaminants that depend on models of sub-surface features and groundwater flows.

Often an initial goal of environmental sampling at chemically contaminated sites is to define the contamination's extent and location. This goal is distinct from classical survey sampling, which typically estimates a mean or total concentration. However, the cost of laboratory analyses to determine the mean or total concentration for many dispersed chemicals may not permit a sufficiently intense classical sampling grid. Often only a few such soil, water, or air samples can be analyzed. Consequently, sampling programs at chemically contaminated sites must consider coordinated strategies for field sampling and laboratory testing of samples. PNL research in environmental study design has found acceptable ways of compositing (mixing of either laboratory or field samples) that reduce costs and allow inferences over large areas.

The intent of sampling programs to detect contaminant hot spots or migration is to ensure that

all areas with potential major health or environmental risks are found and characterized. Two methods have been developed for increasing the probability of detecting such areas. The first approach was to develop sampling design recommendations specifically for the purpose of sampling for detection. In concurrent research, group testing (compositing) methods were developed to identify excessive contaminant levels in the individual components of composited samples. Together, these two approaches permit more extensive and better use of samples while permitting minimal analytical costs.

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Dynamics of Wild Populations: Detecting and Measuring Change

L. L. Eberhardt

One of the major problems in attempting to develop principles of theoretical ecology is the lack of suitable field data for testing the models thus developed. Any attempts to apply the results to various present-day concepts of global threats may thus be hampered by an inability to show that the models have any basis in reality. Therefore, one of the chief goals of this project is to develop data for these models from real populations. A second major purpose of the research is to develop and evaluate techniques to provide numerical assessments of trends and other evidence of change in such populations.

One effort under this project in FY 1988 was devoted to evaluating the validity of the simplest model of change in an age-structured population:

$$N_t = N_0 \lambda^t \quad (1)$$

where N_0 is an initial population size, N_t its value at time t , and λ the finite population growth rate (often expressed as e^r). Monte Carlo simulation models of several different types of populations were constructed, and four ways of assessing rates of change were tested:

1. solving Eq. (1) directly, using first and last observations
2. using log-linear regression
3. fitting by non-linear least-squares
4. analyzing a series of successive pairs of values by "jackknifing."

The major finding from these studies is that the various methods of analysis do not provide reliable measures of the stochastic variability in the simple underlying growth process (i.e., the standard statistical confidence limits are much too narrow, including roughly half of true values in the cases tested). If the fitted models are used for short-term (a few years) predictions of future population sizes for the same simulated trend, performance improves, but is still unsatisfactory in terms of the theory as usually accepted.

Initial investigations have been carried out on several sets of long-term data that will be extremely valuable in assessing long-term global change in arctic environments. The two major data sets thus far investigated are on the Pacific walrus and the northern fur seal. Detailed data on walrus are available for about 60 years, and a general reconstruction of past trends is feasible back to about 1860.

A good general understanding of the fur seal population is available back to 1787, when commercial harvests of the seals on the Pribilof Islands began. Figure 1 shows detailed data on the population under varying regimes of exploitation are available since 1912, with the best records on the numbers of pups. Data on other components for the interim period (1925 to 1950) are available that will permit reasonably accurate reconstruction of those years for the pup population. Analysis of this population is particularly important because some of the most

extensive data on any natural population are available since the 1950s, but as yet no satisfactory explanation is available of the persistent downward trend that has occurred since about 1960. A detailed initial modeling study of the trend was conducted; this analysis and data set will be particularly valuable in studying global-change phenomena.

Intensive oceanographic and atmospheric study of the Pacific waters has also been underway for many years, in connection with fisheries management and the broader implications of various environmental phenomena. Continued study of the fur seal data will contribute a new dimension to the general understanding of these phenomena, because of the extensive migrations of the seals in the western Pacific during the pelagic stage of their life cycle. In particular, the substantial recent downward trend in the population may well be found to be a result of a widespread environmental perturbation of some sort (no commercial harvests of the population are currently conducted).

In other work, research on a wide variety of species and circumstances continues. Further examples for developing methodology for population projections have been studied (Eberhardt 1987), data for further tests of hypotheses about populations have been located and used in preliminary assessments (Eberhardt 1988a), and further studies of the theory of age structures (Eberhardt 1988b) and calibrating indices (Eberhardt and Simmons 1987) have continued.

References

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FIGURE 1. History of Abundance of the Pribilof Fur Seal Population

Quantifying Energy-Related Effects on Mobile Species

J. M. Thomas, J. R. Skalski, and S. G. Smith

Band-recovery techniques, catch curve analyses, and open-population models all provide survival estimates for animal populations. However, in the fields of animal diseases and toxicology, more than mere survival estimates are desired, for the focus is on relating changes in survival rates to concomitant variables and exposures. Quantitative methods for survival are studied by relating extrinsic factors, such as energy production, habitat alterations, and chemical releases, to the survival rate of biota.

Biologists use an array of techniques to estimate animal survival rates. Among them are band-recovery models, open-population models, and catch-curve analyses. However, population investigations should not conclude with a series of survival rates. Concerns about human- and nature-induced effects on wild populations have increasingly focused on how fluctuations in survival rates may be related to concomitant factors. This focus on establishing correlative, if not causal relationships, is a relatively recent development in statistics despite a long history of interest in effects (e.g., hunting pressure affecting hunting regulations, annual climatic events affecting overwinter survival, and the effects of chemical use on local populations).

Two areas of statistical theory that limit the transition of survival studies from estimation to assessment and hypothesis testing include 1) a lack of well-defined field study designs focused on assessing environmental impacts on survival rates, and 2) a lack of statistical models for analyzing tag-recapture or radiotelemetry data in a hypothesis-testing framework. This research has been directed at approaches to rectify these deficiencies.

Three response models, two sampling models, and two field designs were examined to derive the best experimental strategy. In the case of interventions (e.g., installing an energy facility or spraying large areas with herbicides), results indicate that a replicated experiment (five control and exposed plots, resampled for each of two preintervention and postintervention periods) based on an exponential survival model and the concept of proportional hazards has the statistical

properties necessary to detect a 20% reduction in survival caused by exposure with a Type I error of 0.10 and power of 90%.

The concept of proportional hazards requires that birds or small mammals that reside on treatment and control plots have the same ratio of hazard during the experiment (animals must be either radio-tagged, or a mark-release recapture study must be conducted so that the population is closed). Because of this requirement, the resampling periods must be scheduled very close together. At this time, model parameters have been included to account for plot-to-plot and period-to-period variability as well as the effect of chemical insult or other intervention. Moreover, likelihood ratio tests have been devised to specifically assess environmental impacts in our experimental framework. These likelihood tests were compared with the outcome from two-sample t-tests using simulation techniques. The results show that under circumstances likely to be encountered in field studies (i.e., few plots, modest numbers of animals marked or tagged per plot, and high plot-to-plot variability), the approach developed is superior to a simple t-test of treatment and control survival estimates.

Future Research

The theoretical ecology program comprises projects that will probably be combined. The resulting single program will focus on theoretical and quantitative ecology. Research areas selected for emphasis include:

- providing a paradigm to explain observed scale differences at more than two NERP sites
- developing models that describe multiple scales and testing those models on NERP sites
- proposing a methodology for satellite-based, landscape-level change detection at arid sites
- proposing optimal sampling times, distances, or other independent variables to obtain minimal confidence limits for parameters in models that describe large-scale ecological phenomena
- exploring Bayes sampling methods as cost-effective ways to detect the amount and pattern of chemicals at hazardous waste sites.

Relating environmental hazards to animal survival is also anticipated to be an active area of future research. Applications will cover relating salmon smolt survival during outmigration to river flow

levels, overwinter survival to ambient conditions, and carry-over capacity and the effects of energy development and facility maintenance on animal survival.

Exploratory Research

Exploratory research projects make substantial contributions to OHER Environmental Sciences Programs and other DOE and related research. PNL places a high priority on this basic research, which captures ideas and develops technologies that provide the foundation for subsequent research to understand environmental processes. In FY 1988, exploratory research projects investigated: the biogenic cycling of volatile nitrogen-containing species, remote thermal data for estimation of evapotranspiration, the interactions of contaminants and particles, new concepts for extrapolating ecological processes to landscape and regional scales, the variability and speciation of mercury, the detection of genetically engineered microorganisms in the environment, and databases for evaluating salmon spawning habitat on DOE lands.

Biogenic Cycling of Volatile Nitrogen-Containing Species

B. D. McVeety and S. O. Farwell (University of Idaho)

The objective of this project is to develop and demonstrate the technical capability necessary for determining the spatial and temporal fluxes of volatile nitrogen species produced by soil/plant systems in semiarid environments. This demonstrated capability will be coupled with existing technologies and used to support a detailed evaluation of the relevance of both terrestrial and marine biogenic cycling processes to the global inventories of carbon, sulfur, and nitrogen.

The known, volatile compounds of major significance to the biogeochemical nitrogen cycle are NH_3 , NO , NO_2 , N_2O , N_2 , and possibly organic amines [CH_3NH_2 , $(\text{CH}_3)_2\text{NH}_2$]. Therefore, first-year efforts were concentrated on developing and validating a sensitive analytical methodology for field-based emission measurement of these specific gases. The system design has been kept flexible, allowing for the detection and identification of other, as of yet unknown, volatile nitrogen-containing compounds that may also play a significant role in nitrogen budgets.

A nitrogen-specific detector was fashioned from a commercially available Monitor Labs (San Diego, California) 8440E NO chemiluminescence detector. One of the more important modifications made to the detector included a redesigning of the detection cell, minimizing its volume to better accommodate the small flow rates typical of capillary gas chromatography (GC). Another important modification was the addition of a catalytic oxidation inlet system that uses a platinum tube heated to 1000°C . This catalytic conversion system transforms the nitrogen-containing species eluting from a high-resolution GC column to NO

for subsequent $\text{NO} + \text{O}_3$ chemiluminescence detection. A gas sample cryogenic concentration/injection system was also built and fitted to the inlet side of a Hewlett-Packard (Avondale, Pennsylvania) 5880 GC. Together, this injection/detection system has been successfully tested using various dilutions of gas standards. The detection limit of the detector is about 10 picograms of nitrogen.

On initiation of this project, a set of ultra-low-loss permeation tubes was purchased, and a set of gravimetric permeation tubes was prepared for instrument calibration purposes. The ultra-low-loss tubes (loss rates of $< 1 \text{ ng/min}$) are better suited for field use than the gravimetric permeation tubes, because their loss rates are relatively insensitive to moderate shifts in temperature ($\pm 5^\circ\text{C}$), and they can be used with minimal dilution. The gravimetric tubes are necessary to use as primary standards for calibrating the low-loss tubes, as their loss rates are high enough that they can be accurately determined by carefully weighing the tubes over a period of 6 to 8 months.

Until recently, the loss rates of the low-loss permeation tubes have been assumed based on theoretical calculations. Sufficient data are now available on the rate of weight loss from the gravimetric tubes that accurate loss rates can be assigned to the low-loss permeation tubes. This accomplishment has 1) allowed the conversion efficiency of our catalytic oxidation system to be determined for each of the nitrogen-containing compounds studied, and 2) provides a set of accurate calibration standards to use in the future stages of this project.

The initial FY 1989 task will be the refinement of the chromatographic system for the separation/quantification of volatile nitrogen-containing species. The second task will be interfacing the

calibrated detection scheme with the dynamic enclosure chambers currently used at PNL for plant-gas exchange experiments.

Later, measurements of volatile nitrogen-containing compounds emitted from plant-soil systems will begin. Current plans are to join with plant-gas exchange/microbial community experiments being conducted on other PNL research projects. These experiments will test the performance of the collection, separation, and detection systems, and identify and resolve any major problems before field tests begin in the spring. In addition, biogenic nitrogen emission rates will be measured on plant-soil systems representative of the ALE Reserve ecosystem.

Two separate tasks are planned for FY 1989. The first objective is to make field measurements of biogenic nitrogen emissions at the ALE Reserve. In arid ecosystems, biological activity peaks during the spring when temperature and moisture are optimum. It is expected, although not confirmed, that elevated biogenic emission rates of nitrogen-containing compounds will coincide with this period of high biological activity. Temporal emission rates for three seasons will be collected by the beginning of FY 1990.

The second objective for the spring/summer of FY 1989 is to conduct controlled experiments to identify those environmental parameters that play a key role in determining biogenic nitrogen fluxes. These experiments will be conducted on intact soil/plant systems housed in microcosms, and will build on the information derived from the initial experiments conducted during the winter of FY 1989. These experiments will provide data essential for the interpretation and modeling of the measured biogenic emissions.

Remote Thermal Data for Determining Evapotranspiration in Arid Landscapes

P. A. Beedlow

Environmental impacts frequently affect large areas of land. To assess and mitigate these impacts effectively and economically, important ecosystem processes must be measured on the landscape scale. Evapotranspiration (ET) is one such important process that is difficult to determine for heterogeneous arid environments such as the Hanford Site.

The objectives of this project were to determine how Landsat 5 data can aid understanding the

energy exchange processes, such as ET, that affect the heat and water balance of arid ecosystems, and to develop techniques to measure those processes over landscapes and regions.

This last year, FY 1988, was the third and final year of this exploratory research task. Studies conducted in each year addressed different aspects of measuring the energy exchange processes that affect heat and water balance in arid landscapes. This research was considered to be especially important because the processes involved in maintaining the energy balance over large land areas are also largely responsible for maintenance of continued ecosystem functioning. The improved understanding of the energy dynamic processes is providing new insights into the integration of the hydrological, ecological, and atmospheric processes that are amenable to remote spectrometric measurement. The major technical accomplishments over this 3-year period include:

FY 1986. The accuracy of Landsat TM thermal data for estimating surface temperature in arid terrestrial environments was assessed. Accurate measurement of surface temperature is critical because it is a major driver of arid-land ET. The findings indicated that TM data can be used to estimate surface temperature to within 1°C of ground truth data if the TM data are adjusted for local atmospheric effects and for the emissivity of vegetation and soil.

FY 1987. Models of ET based on energy balance processes were compared with actual ET measurement from precision lysimeters located at ALE's Watershed Research Facility. The most appropriate existing ET model derived for arid-land agriculture proved inaccurate when compared with lysimeter values. Further investigation determined that surface temperature alone accounted for 90% of the variability in ET.

FY 1988. Ground truth instruments for calibrating satellite measures of energy balance were tested. Specially designed portable micrometeorological stations were developed for measuring energy balance parameters within major landscape units on the Hanford Site. These stations were tested in combination with large precision weighing lysimeters at the Watershed Research Facility to evaluate energy-based models of shrub-steppe ecosystem functioning. The models provide a major new tool for laboratory investigations of the

driving forces influencing ecological-hydrological processes over large land areas.

This research project has focused on developing the capability to quantitatively measure evapotranspiration, one of the integrated regulating mechanisms that influence the ability of ecological systems to maintain their integrity. The ability to measure and interpret the response of key ecosystem processes on a landscape-level basis will provide new opportunities in assessing and monitoring environmental changes associated with large-scale energy development and use.

Comparison of Techniques to Elucidate Adsorption Phenomena in Aqueous Solutions

M. J. Peloquin, J. R. Morrey, and J. E. Amonette

The goal of this exploratory research is to understand the chemistry of interactions between contaminants and subsurface particles. To do this, the objective of FY 1988 research was to determine which of three techniques, Fourier-Transform infrared attenuated reflectance spectroscopy (FTIR), nuclear magnetic resonance (NMR) or near-infrared (IR), would be applicable to the movement of adsorbed contaminants in the subsurface.

Aqueous suspensions of spherically shaped colloidal hematite were examined using FTIR.

Magnetic properties of hematite made it impossible to obtain good NMR separation. The near-IR spectra require the measurement of overtone spectra, which were generally weak, and these spectra cannot be obtained at energies below 6700 wavelengths. Hence the major effort was focused on FTIR measurements.

Three different crystals were used in the FTIR study: germanium, KRS-5, and ZnSe. Sufficient signal could not be obtained from the germanium crystal. The KRS-5 crystal produced a spectrum of the water adsorbed to colloidal hematite. This spectrum showed a strong doublet with peaks at 3500 and 3150 cm^{-1} , which revealed two oxygen-hydrogen (O-H) stretching modes (ostensibly two types of water near the surface). The O-H bending peak at 1620 cm^{-1} shows that the water environment is altered near the particle surface. However, because KRS-5 has a finite solubility in water, it was rapidly degraded and could not be effectively used over a long period of time.

Measurements with the ZnSe crystal revealed large anomalous, but reproducible, time-dependent spectral changes. These changes occurred even when pure water was placed around the crystal. Figure 1 illustrates these changes where the initial spectrum at 5 min is subtracted from each of the others before display; thus, it becomes the reference for all of the others. In Region A, the

FIGURE 1. FTIR Infrared Vibrational Spectra of Water Near or Adsorbed to the Surface of ZnSe. All scans are ratioed against a water background taken 2 min after the water touched the crystal. All times are calculated from the initial contact of water on the crystal. Unexpected time dependency implies either a high activation energy for adsorption or a need for cooperative events to occur. The behavior of the spectrum in Region A is not currently understood.

optical absorbance has a time-dependent decrease up to and beyond 300 min (25°C), but between 300 and 1110 min, a dramatic change occurs. No significant changes occur after 1110 min. These changes are not understood, but they most likely relate to adsorption mechanisms. In Region B, the water O-H stretching frequency has a time-dependent shift from a higher to a lower frequency, indicating gradual adsorption of water on the surface. Region C shows the stretching frequency of the adsorbed water. Region D is the bending mode of water. The absorption band broadens with adsorption, indicating that the adsorption sites provide a wider range of vibrational environments than the structure of non-adsorbed water. The slow processes imply either fairly large activation energies for adsorption or the need for a cooperative mechanistic phenomenon.

The FTIR technique is valuable in probing the molecular nature of liquid-solid interfaces. These results have important implications in research that could be conducted under the new Sub-surface Transport Initiative, the Environmental Center of Excellence, and in the Molecular Science Research Center. Other work under a related project "Applied Quantum Mechanics" correlated predictions of the vibrational frequency of adsorbed particles with the experiments described in this section.

Optimal Measurement Scales and Experimental Design for Detecting and Predicting Global Ecosystem Change

J. M. Thomas, V. I. Cullinan, and M. A. Simmons

The goal of this project is to develop a methodology for quantifying landscape changes that result from global or regional environmental impacts. The results of this project will be fundamentally significant, because they will allow predictions about desertification, and applicable, because they will provide a way to monitor landscape-level changes.

To achieve this goal, project objectives are to 1) develop quantitative methods to scale measurements of arid-land ecological processes to the landscape level, 2) provide a theoretical link between landscape-level measurements and analytical sampling methods for studying global change and the processes that cause desertification, and 3) develop methods to quantify landscape change resulting from global or regional factors.

In FY 1988, statistical models were extended that determine ecological scale by using estimates of variability and correlation across a landscape. The method works for field data based on 1-m cover estimates and for Landsat satellite reflectance data contained in 30-m² pixels collected from the same site. Ecological scale is the level of resolution that provides the greatest efficiency in studying particular phenomena.

PNL scientists used statistical measures of the strength of interrelationships between ecological processes to define the most appropriate level of scale for research; in this case, optimal transect size and dispersion over the landscape were determined. To obtain data to test the model, plant-cover measurements were made at two sites in the shrub-steppe habitat: the ALE Reserve in southeastern Washington, and the INEL near Idaho Falls, Idaho.

Based on the observed plant cover, the optimal transect size for the two plant species studied, a perennial grass, *Agropyron spicatum*, and a shrub, *Eurota lanata*, was found to be about 100 m. Such a transect is much longer than normally used or mentioned in the ecological literature. Cover estimates are usually made over a 1-m² area and rarely over 10 m². A correlation model was used to estimate the optimal dispersion of these 100-m-long transects over the landscape. At the INEL site, the processes that influence cover were found to be different beyond 300 m for the two plant species studied. At ALE, this distance was more than 400 m. These distances define an environmental area of influence, which is the distance at which two points in the landscape no longer experience the same habitat conditions.

Because the optimal scale based on field data from two sites was on the order of 100 m, it was hypothesized that Landsat images, composed of 30-m² pixels, might show the same correlation and variance patterns. To explore this hypothesis, satellite images of the ALE site were compared with results from the ground-based data. Data from forty 2000-m transects were analyzed using the same models and methods used for the ground-cover data. The general response patterns were similar.

Based on these results, it is concluded that 1) the scale where similar processes no longer influence percentage cover is between 300 and 400 m, depending on the location and plant species measured, and 2) the optimal transect segment

length is about 100 m for both species. Landsat imagery provided a similar estimate of scale.

These results imply that long-term ecological monitoring or monitoring to detect global change needs to consider the size and spacing of sampling units over landscapes. Moreover, if ground truth for satellite-based measures can be obtained, then Landsat methodology may offer an inexpensive satellite-based means to assess global change.

These study results were presented at the Landscape Ecology Symposium in Albuquerque in March 1988, and at the Ecological Society of America meeting, August 1988 in Davis, California.

Variability and Speciation of Atmospheric Mercury

N. S. Bloom

Mercury is one of the most mobile and toxic of the trace elements in the environment. Much of the mercury in aqueous and atmospheric systems is introduced by human activities such as coal burning. The proportion of this mercury from energy-related activities will most likely rise as world energy consumption rises. Atmospheric mercury, washed to the soil and aqueous systems by rain, may recycle back into the air through volatilization from plants, or accumulate in marine biota. Consequently, the potential exists for accelerated increases in global atmospheric and aqueous mercury levels as a result of energy production and use. These increases in mercury may show up in monitored environmental systems such as fish, resulting in regulatory limit violations and potentially causing a human health hazard.

Thus, this project has two objectives: to obtain a record of long-term changes and shorter-term variability in the atmospheric mercury burden, and investigate the chemical speciation and mechanisms of precipitation scavenging for mercury. A 20-ft sampling tower and pump house were erected on the Pacific Ocean-facing bluffs of Cheekah Peak, at a site currently managed by the University of Washington atmospheric sciences department. Daily samples have been collected at the site since July 1988.

In addition, a novel method for determining the speciation of organomercury compounds in water was developed. This method uses derivatives of the compounds (volatile ethyl-analogs), which are then separated by cryogenic gas chromatography

and detected by cold vapor atomic fluorescence. Currently, this is the most sensitive and selective method in use, and will be described in two papers to be published in the open literature.

Environmental Detection and Fate of Genetically Engineered Microorganisms

J. K. Fredrickson

The current focus of this research project is to establish capabilities at PNL to detect and establish the fate of genetically engineered microorganisms deliberately or accidentally released to the environment.

In FY 1986 and 1987, transposable genetic elements were inserted into common soil bacteria to develop and refine gene probe methods of detecting bacteria in soils and sediments. Few techniques are available to detect and enumerate microorganisms in environmental samples with the sensitivity required for adequate risk assessment.

The detection procedure developed was rapid and sensitive. As few as 10 cells per gram of soil ($1 \text{ cell in } 10^7$) could be detected. Results from this work, conducted in collaboration with Dr. D. F. Bezdicek of Washington State University (now a PNL associate staff scientist), have been published in *Applied and Environmental Microbiology* as "Enumeration of Tn5 Mutant Bacteria in Soil by Using a Most-Probable-Number-DNA Hybridization Procedure and Antibiotic Resistance," by J. K. Fredrickson, D. F. Bezdicek, F. J. Brockman, and S. W. Li (54:446-453).

One disadvantage to the detection method developed was that it required growth of the target microorganisms, which led, in late FY 1987 and FY 1988, to direct quantitative extraction of DNA from soils and sediments and detection of specific genes using a gene probe. Direct detection methods avoid problems with culturing microorganisms, such as physiological changes that can limit a microorganism's culturability, and the transfer of recombinant genes to other microorganisms. Bound or strongly associated humic materials had to be removed from the DNA using CsCl density gradient ultracentrifugation to prevent nonspecific binding of DNA probes. The results indicate that DNA can be quantitatively extracted from soil using organic solvents. This DNA is comparable with that extracted from lysates of bacteria recovered from soil using a repeated centrifugation procedure.

For some soils, significantly greater amounts of DNA were extracted per unit weight using the direct extraction procedure than when bacterial cells were removed and then lysed. This suggests that either DNA from eukaryotes (fungi, protozoa, etc.) is being extracted, or that significant quantities of free DNA are present in soil in protected microsites or sorbed to clay.

A paper, "Detection of a Genetic Marker in Total DNA Extractions from Soil," by B. Denovan and J. K. Fredrickson, presented these results at the 1988 American Society for Microbiology Annual Meetings.

Modeling Salmon Spawning Habitat Through Remote Sensing and Geographic Information System Techniques

D. D. Dauble

Accurate descriptions of salmon spawning habitat are needed by resource managers and technology developers before the potential impacts of human activities on important species can be assessed. Current techniques used for change detection in aquatic systems are limited, both in their ability to quantify changes in population size and to provide qualitative descriptions of spawning locations.

The objective of this study was to establish a database and to evaluate the technology needed to assess concept feasibility.

Three activities were initiated in FY 1988: 1) synthesis of data on Columbia River salmon populations relevant to change detection and concept modeling requirements, 2) organization and evaluation of Columbia River hydrology data relevant to definition of spawning locations, and 3) identification and evaluation of remote sensing techniques (aerial photography) to be used for parameter descriptions.

The 40-year database on chinook salmon spawning at Hanford was summarized by location. River flow and temperature data during peak spawning intervals were also collected from historical records. Mapping data located include U. S. Geological Survey maps that have been digitized and placed in the Geographic Information System, aerial photographs of the Hanford Reach (1930 and 1976), and bathymetric descriptions of several point locations in the Reach. Additional data on velocity and substrate for one major spawning area were also collected. This information is useful for change detection and impact analyses of current and planned DOE activities in the Hanford Reach of the Columbia River.

Educational Institution Interactions

As part of its mission, PNL contributes to the expansion of science and technology education by building partnerships and developing collaborative programs with high schools, colleges, and universities in the region. Through these programs, students and teachers are provided access to some of the Laboratory's unique facilities, such as the advanced Biogeochemical Laboratories, the NERP, MSL, Aerosol Research Center, Watershed Research Facility, and Subsurface Environmental Research Facility.

The Northwest Organization for Colleges and Universities for Science (NORCUS) serves as the mechanism for student and faculty appointments to these programs. The programs are coordinated by the PNL offices of University Relations and Pre-University Education.

Pre-University Interactions

PNL scientists participate in pre-university education activities such as 1) classroom instruction and demonstrations; 2) student apprenticeships, workshops, and field days in the laboratory; 3) teacher workshops and cooperative studies; and 4) technical support and assistance to local and regional schools.

During FY 1988, PNL hosted a DOE national high school honors program. This program is designed to introduce students traditionally underrepresented in science and engineering fields to the scientific method, and to acquaint them with the technology used to evaluate the environment and resources available to mitigate natural and human disturbances.

The institute focused on field experiences at ALE, the Remote Sensing Research Center, and the MSL. In FY 1989, PNL will join other national laboratories in conducting a 2-week honors institute for students from every state. Several students also participated in the Student Research Apprenticeship Program, Youth Opportunity, and summer employee programs.

Researchers also worked in the laboratory with teachers participating in the national DOE-sponsored Research in Science and Technology Program. This program gives teachers an opportunity to participate in "hands-on" science for 4 weeks during the summer. In addition, Environmental Sciences Department researchers made presentations at more than 20 local and regional schools through the Sharing Science with Schools Program. These presentations covered subjects such as wildlife biology, ecology, and marine biology.

University Interactions

Through NORCUS, PNL provides university undergraduates, graduates, postdoctoral scientists, and faculty an opportunity to work together in research activities and participate in seminars, scientific meetings, and symposia.

During FY 1988, 17 university faculty, 18 graduate, and 12 undergraduate students participated in Environmental Sciences programs; some of their research projects are summarized below. The Environmental Sciences Department also hosted visiting scientists during the year for workshops, seminars, and DOE program reviews.

In addition, PNL staff also are members of the graduate and teaching faculty at a number of universities, including the University of Washington, Washington State University, and the University of Idaho. Their faculty appointments build on both PNL and university programs by sharing ideas and technology through research and teaching interactions.

National Environmental Research Park

Faculty and graduate students conducted a variety of research projects on the Hanford NERP during the past year. These included research on water relations and photosynthesis in big sagebrush (Dr. H. Black of Washington State University [WSU]), soil microbial ecology (Dr. D. Bezdicsek of WSU), arid land soils (Dr. A. Busacca of WSU), evapotranspiration (Dr. L. Fritchen of the University of Washington), arid land balance (Dr. D. Hillel of the University of Massachusetts, Amherst), geographical information systems (Dr. W. Smith of Central Washington University), the site's wintering bald eagle population

(S. Eisner of the University of Montana), the ecological behavior of coyotes on ALE (Dr. F. Knowlton of Utah State University), and remote sensing (Dr. D. Gibbons of Fort Lewis College, Colorado).

Future research opportunities may be available for students in restoration ecology because DOE plans to revegetate 1- to 15-ha patches of native vegetation disturbed by the now-terminated Basalt Waste Isolation Project. The effort will focus on planting native species and restoring animal populations to predisturbance levels.

Biogeochemical Phenomena

A U.S. Department of Agriculture-Agricultural Research Service staff member has collaborated on nitrogen studies being conducted on the ALE Reserve. Also, a post-doctoral research associate from Washington State University was funded from these projects to investigate the competitive ability of microorganisms in the rhizosphere and the influence on cation mobility.

Subsurface Science

Subsurface science research has been conducted in close coordination with university scientists. Through subcontracts and NORCUS appointments, PNL is collaborating with scientists from Oregon State University (OSU), University of Wisconsin, Purdue University, University of Florida, Florida State University (FSU), University of Illinois (UI), Virginia Polytechnic Institute (VPI), and Princeton University. For example, Dr. J. Westall of OSU is providing expertise and research perspective on the interfacial science and mechanistic modeling of organic and inorganic solute adsorption, currently unavailable at PNL.

Collaboration with academic subsurface investigators has continued as part of the DOE Deep Microbiology program. This collaboration was enhanced through visits to PNL by FSU and UI investigators for seminar presentations and program discussions. Dr. D. Balkwill of FSU will continue to cooperate through his work on elucidating the variation and diversity of heterotrophic bacteria in aquifers; collaboration with Dr. B. Rittmann of UI is expected to be expanded in the area of modeling subsurface microbiological processes and bioremediation.

Faculty and graduate students from VPI and Princeton are performing some of the modeling necessary to test hypotheses on multiphase flow. Drs. J. Parker and R. Lenhard of VPI are modeling idealized two-dimensional flow cell experiments, and making codes available to PNL to better evaluate flow experiments. At Princeton, Dr. G. F. Pinder is directing graduate research on the interphase transfer between isolated organic liquid globules and water in porous media.

Marine Sciences

A graduate student from the University of Washington College of Fisheries initiated a doctoral research project to characterize the resistance to the hemic neoplasia in different Pacific Rim populations of the mussel in comparison with isoenzyme patterns in the populations. This research should allow more precise selection of individuals for genetic uniformity in studying their ability to cause the disease to go into remission. Two additional doctoral degree candidates, registered at the University of Washington, initiated graduate programs this year and will work on research related to this program.

Also, a program of collaborative research is planned for 1989 with the Center for Marine Biotechnology of the University of Maryland at Baltimore. Research collaboration continued with the French Government Institute for the Utilization of Marine Resources in La Tremblade, France. Through this collaboration, the monoclonal antibodies to the cancerous mussel blood cells were produced and characterized.

Theoretical Ecology

Cooperative research is currently being performed with staff at the University of Washington's Center for Quantitative Sciences and the population dynamics group at the University of Minnesota. The theoretical projects support a postdoctoral scientist who has been conducting research on quantitative methods in landscape ecology and a doctoral student who is working on new methods for analysis of survival data under intervention scenarios. These appointees as well as other university scientists strengthen our research capabilities in theoretical and quantitative ecology.

Technology Transfer

Technology originating from research by DOE or other government agencies and industry may have great value in solving many of the pressing problems of the nation, but improved mechanisms are required in order to facilitate transfer to and implementation in sectors that may make the best use of the information. This cross-fertilization of technology and ideas can have a synergistic effect, adding to the usefulness of the research. Contract arrangements between DOE and Battelle Memorial Institute allow for inventive ways of accelerating the development of technology. Examples of technology transfer activities resulting from DOE, Environmental Protection Agency, and Department of Defense research are summarized below.

Detecting and Managing Change in Terrestrial Ecosystems

PNL is supporting the Department of Defense (DOD) in landscape analysis and management by developing and applying remote sensing techniques for the environmental management of the U.S. Army's Yakima Firing Center and Fort Lewis. Similar techniques are being developed for the Department of Energy for management of its national sites; the technology from each program is contributing to the other. This year for the DOD, a primary effort was focused on developing environmental change detection maps, fire maps, and vegetation/landcover maps. Preliminary change detection maps, prepared from French Systeme Probatoire d'Observation de la Terre (SPOT) satellite imagery, show areas impacted by army training and construction activities. Landcover status and training area boundaries have also been added for reference. SPOT and Landsat imagery are now being analyzed to determine the optimal signatures for prior fires and the degree of restoration. The accuracy of vegetation/landcover maps was determined by analyzing and comparing 180 randomly selected map locations with aerial photographs for the nine vegetation/landcover classes established.

Biogeochemical Phenomena

Battelle, Intertec Group, and Dow Corporation are planning a joint venture to develop and test a new product designed to control termites, wood boring insects, and wood rotting fungi. The product is composed of a pesticide encapsulated in a matrix of polymer plastic that provides a controlled release. This design is based on the patented DOE-funded chemical biobarrier system developed earlier by D. A. Cataldo, J. Cline, P. Van Voris, F. Burton, and G. Skeins, which inhibits plant root growth at Hanford low-level waste sites.

An intensive study of the behavior and sensitivity of selected acid rain watershed codes to forecast future acidification of watersheds was performed for the EPA. When calibrated for a control catchment, the ILWAS code provided a reliable simulation of a catchment recovery resulting from the removal of acidic components from precipitation and application of the water through irrigation. Recovery from long-term acidification at this site was indicated by an increase in pH and a decrease in nitrate and sulfate in the soil drainage. Similar studies are under way at a site of intense acidic sulfate deposition and an experimental acidic manipulation site. Early investigations by DOE in developing geochemical models played an important role in establishing the capability of this effort. The DOE is using the results of the current studies in planning its research contributions to the congressionally mandated 1990 assessment of acid rain in the United States.

Subsurface Transport

Researchers in PNL's OHER Subsurface Transport program have invented a device, submitted for patent, for measuring the concentration and relative distribution of water and organic liquids in soil. The potential for technology transfer applications is great. For example, the device can be used to detect leaks from underground tanks of gasoline, diesel and jet engine fuels, and industrial solvents.

The device, costing less than one dollar, is constructed from a light-emitting diode and a light-detecting cell on either side of a small (4-mm-thick by 1-cm-square) plate. The plate, if made of porous fritted glass, preferentially absorbs water; if made of porous fritted polyethylene, it absorbs organic liquids. The amount of liquid the porous plate absorbs is related to the amount of the liquid in the surrounding soil. In turn, the amount and capacity of the absorbed

liquid changes the amount of light reaching the light-detecting cell. Because the cell responds to the varying light with a change in electrical resistance, the reading on an ohm meter reflects the amount of liquid in the soils or sediments. For organic liquids in the subsurface, the reading indicates the level of contamination.

The device is a major advance in the technology available for measuring liquids in partially saturated soil and other similar media. It allows in situ measurements; it is not compromised by the electrical conductivity of soluble ions; it requires little maintenance; and it is much cheaper than previous methods.

Two studies funded by the Environmental Protection Agency (EPA) are using capabilities developed in the DOE subsurface transport program. In a recently completed study for the EPA Office of Research and Development, estimates of octanol/water partition coefficients and water solubilities were reported for selected high-molecular weight chlorinated dioxins and dibenzofurans. Accurate measurements of the physicochemical properties of hazardous compounds is important in exposure assessment. An ongoing study funded by the same EPA office is evaluating the status of knowledge of the influence of volatilization and particle resuspension processes on the atmospheric concentration of contaminants at hazardous waste sites. The results are being used to identify research needed to evaluate the relative influence of vapor versus particle transport of low-vapor-pressure, highly sorbed organic compounds downwind from hazardous waste sites. Thus, the results from both these studies are helping EPA guide regulatory policy aimed at making sound decisions for

existing hazardous waste problems, and help anticipate problems created by future waste disposal practices.

Marine Sciences

During FY 1988, numerous requests were received from regional fish and shellfish farms and from federal and state government agencies to provide advice or research on health management or disease problems of fish and shellfish. This advice and research applies to both farmed and natural populations of fish and shellfish. Because no other organization provides this assistance, PNL did so where feasible. Over 50 such assistances were provided. In conducting these activities, specific research methodologies or specific disease discoveries resulting from OHER-sponsored research conducted on this program since 1985 were used.

Research projects supported by OHER at the Marine Sciences Laboratory have developed analytical chemistry techniques for the analysis of ultra-low concentrations of metals and specific chemical forms of metals in water, air, sediment, and tissue. These techniques have been applied to other projects funded by the U.S. Department of Interior, U.S. Army Corps of Engineers, EPA, Electric Power Research Institute (EPRI), and American Smelting and Refining Company. For instance, EPA and EPRI-funded research on the geochemical cycling of mercury in acidified lakes is using OHER-developed technology in the analysis of total mercury and methylmercury. In these lakes, acidified by acid rain, the mercury concentrations in edible fish have been shown to be elevated.

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Salt Lake City, UT 84112

Assistant Secretary
Environment, Safety & Health
Department of Energy
EH-1, FORS
U.S. Department of Energy
Washington, DC 20585

S. I. Auerbach
Oak Ridge National Laboratory
P. O. Box X
Oak Ridge, TN 37830

J. A. Auxier
IT/Radiological Services
Laboratory
1550 Bear Creek Road
P. O. Box 549
Oak Ridge, TN 37831

F. I. Badgley
13749 NE 41st Street
Seattle, WA 98125

D. L. Balkwill
Department of Biological
Sciences
309 Nuclear Research Bldg.
Florida State University
Tallahassee, FL 32306

R. M. Baltzo
Radiological Safety Division
University of Washington
Seattle, WA 98105

R. W. Barber
EH-131, GTN
U.S. Department of Energy
Washington, DC 20545

A. D. Barker
Battelle Columbus Laboratories
505 King Avenue
Columbus, OH 43201

J. R. Barker
Office of Environmental Audit
and Compliance
U.S. Department of Energy
Washington, DC 20545

W. W. Barker, Chairman
Department of Biology
Central Washington University
Ellensburg, WA 98926

N. F. Barr
ER-72, GTN
U.S. Department of Energy
Washington, DC 20545

J. E. Baublitz
Office of Nuclear Energy
NE-20, GTN
U.S. Department of Energy
Germantown, MD 20545

J. R. Beall
ER-72, GTN
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P. A. Beedlow
U.S. Environmental Protection
Agency
200 SW 35th St.
Corvallis, OR 97330

M. H. Bhattacharyya
Argonne National Laboratory
BIM Div., Bldg. 202
9700 South Cass Avenue
Argonne, IL 60439

W. R. Bibb
Energy Programs and Support
Division
U.S. Department of Energy
P.O. Box B
Oak Ridge, TN 37831

R. A. Black
Department of Botany
Washington State University
Pullman, WA 99164

R. P. Blaunstein
EH-132, GTN
U.S. Department of Energy
Washington, DC 20545

L. Bliss
Department of Botany
University of Washington
Seattle, WA 98105

H. S. Bolton
National Oceanic &
Atmospheric Administration
HG Hoover Building,
Room 5222
Department of Commerce
LAX 1
Washington, DC 20230

C. M. Borgstrom
Acting Director, NEPA
EH-25, Room 3E080
U.S. Department of Energy
1000 Independence Avenue,
SW
Washington, DC 20585

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NE-22, GTN
U.S. Department of Energy
Washington, DC 20545

A. Brodsky
Allen B. Consultants
16412 Kipling Road
Derwood, MD 20855

W. Broecker
Lamont Geological Observatory
Columbia University
Palisades, NY 10964

D. R. Buhler, Chairman
Toxicology Program
Oregon State University
Corvallis, OR 97331

R. J. Bull
Associate Professor of
Pharmacology/Toxicology
College of Pharmacy
Pullman, WA 99164-6510

G. Burley
Office of Radiation Programs,
ANR-458
Environmental Protection
Agency
Washington, DC 20460

W. W. Burr, Chairman
Medical & Health Sciences
Division
Oak Ridge Associated
Universities
P.O. Box 117
Oak Ridge, TN 37830

L. K. Bustad
College of Veterinary Medicine
Washington State University
Pullman, WA 99164-7010

S. Calcese
Water Resources Center
University of Wisconsin
1975 Willow Drive
Madison, WI 53706

J. T. Callahan
Associate Program Director
Ecosystems Studies Program
National Science Foundation
Washington, DC 20545

R. J. Catlin
Electric Power Research
Institute
3412 Hillview Avenue
P.O. Box 10412
Palo Alto, CA 94303

H. H. Cheng
Department of Agronomy &
Soils
Washington State University
Pullman, WA 99164-6420

G. Chesters, Director
Water Resources Center
University of Wisconsin
1975 Willow Drive
Madison, WI 53706

Chief
Game Management Division
Department of Game
600 North Capitol Way
Olympia, WA 98501

J. A. Coleman
NE-24, GTN
U.S. Department of Energy
Washington, DC 20545

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ER-15, GTN
U.S. Department of Energy
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ER-75, GTN
Department of Energy
Washington, DC 20545

J. Corey
Environmental Sciences
Savannah River Laboratory
Drawer E
Aiken, SC 29801

Council on Environmental
Quality
722 Jackson Place, NW
Washington, DC 20503

T. S. Crawford
Chicago Operations Office
U.S. Department of Energy
9800 South Cass Avenue
Argonne, IL 60439

T. V. Crawford
Atomic Energy Division
Savannah River Laboratory
E. I. duPont de Nemours &
Company
Aiken, SC 29808

J. Cushman
Water Resources Research
Center
Purdue University
Lilly Hall
West Lafayette, IN 47907

J. T. Davis
San Francisco Operations
Office
U.S. Department of Energy
1333 Broadway
Oakland, CA 94612

F. G. Dawson
Battelle Memorial Institute
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G. DePlanque, Director
U.S. Department of
Energy-EMEL
375 Hudson Street
New York, NY 10014

Director
New England Marine
Laboratories
Washington Street
P.O. Box 1637
Duxbury, WA 02332

Director
Washington State Department
of Ecology
Olympia, WA 98501

K. R. Dixon
Washington Department of
Wildlife
600 North Capitol Way
Olympia, WA 98504

T. J. Dobry, Jr.
DP-226.3, GTN
U.S. Department of Energy
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H. Drucker
Argonne National Laboratory
9700 South Cass Avenue
Argonne, IL 60439

A. P. Duhamel
ER-74, GTN
U.S. Department of Energy
Washington, DC 20545

D. N. Edgington
University of Wisconsin
Center for Great Lakes Studies
P.O. Box 413
Milwaukee, WI 53201

C. W. Edington, Director
Board on Radiation Effects
Research
National Research Council
2101 Constitution Avenue, NW
Washington, DC 20418

L. F. Elliott
U.S. Cotton Research Station
USDA-ARS
17053 Shafter Avenue
Shafter, CA 93263

R. J. Engelmann
11701 Karen
Potomac, MD 20854

R. Eppley
Scripps Institute of
Oceanography
LaJolla, CA 92307

R. D. Evans
Department of Botany
Washington State University
Pullman, WA 99164

S. J. Farmer
17217 77th Avenue W.
Edmonds, WA 98020

W. O. Forster
ER/OHER/ERD
U.S. Department of Energy
Washington, DC 20545

A. Gahler
Environmental Protection
Agency
Region X Laboratory
P.O. Box 549
Manchester, WA 98353

C. George
Office of Defense Programs
DP-12, GTN
U.S. Department of Energy
Germantown, MD 20545

T. F. Gesell
Idaho Operations Office
U.S. Department of Energy
785 DOE Place
Idaho Falls, ID 83402-4149

W. C. Ghiorse
Microbiology Department
415 Stocking Hall
Cornell University
Ithaca, NY 14853

H. Giles
Strategic Petroleum Reserve
FE-531, FORS
U.S. Department of Energy
1000 Independence Avenue,
SW
Washington, DC 20585

R. D. Gilmore, President
Environmental Health Sciences,
Inc.
Nine Lake Bellevue Building
Suite 104
Bellevue, WA 98005

M. Goldman
Department of Radiological
Sciences (VM)
University of California
Davis, CA 95616

G. Goldstein
ER-74, GTN
U.S. Department of Energy
Washington, DC 20545

F. B. Golley
Institute of Ecology
University of Georgia
Athens, GA 30601

D. H. Hamilton, Jr.
ER-72, GTN
U.S. Department of Energy
Washington, DC 20545

W. R. Hansen
MS-F643
Los Alamos National
Laboratory
P.O. Box 1663
Los Alamos, NM 87545

F. Harrison
Biomedical and Environmental
Research Program
Lawrence Livermore
National Laboratory
C-523
University of California
P.O. Box 808
Livermore, CA 94550

D. W. Hayne
Department of Statistics
North Carolina State University
Box 8203
Raleigh, NC 27695-8203

F. Hutchinson
Department of Molecular
Biophysics & Biochemistry
Yale University
260 Whitney Avenue
P.O. Box 6666
New Haven, CT 06511

H. Ishikawa, General Manager
Nuclear Safety Research
Association
P.O. Box 1307
Falls Church, VA 22041

K. L. Jackson, Chairman
Radiological Sciences Group
SB-75
University of Washington
Seattle, WA 98195

A. Janetos
U.S. Environmental Protection
Agency
401 M. St. SW
Washington, DC 20460

A. W. Johnson
Vice President for Academic
Affairs
San Diego State University
San Diego, CA 92182

L. J. Johnson
Idaho National Engineering Lab
IRC MS 2203
P.O. Box 1625
Idaho Falls, ID 83415

R. Johnson
Office of Radiation Programs
(ANR-460c)
Environmental Protection
Agency
401 M Street, SW
Washington, DC 20460

G. Y. Jordy, Director
ER-30, GTN
U.S. Department of Energy
Washington, DC 20545

R. F. Kendall
Bartlesville Energy Research
Center
U.S. Department of Energy
P.O. Box 1398
Bartlesville, OK 74005

H. Kibby
U.S. Environmental Protection
Agency
200 SW 35th St.
Corvallis, OR 97330

T. Kimball
National Wildlife Federation
1512 16th Street, NW
Washington, DC 20036

M. Kosorok
Department of Statistics
Brigham Young University
Provo, UT 84602

R. T. Kratzke
EH-131, GTN
U.S. Department of Energy
Germantown, MD 20545

D. Krenz
DOE - Albuquerque Operations
Office
P.O. Box 5400
Albuquerque, NM 87115

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Environmental Protection
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Los Angeles, CA 90024-1786

J. N. Maddox
ER-73, GTN
U.S. Department of Energy
Washington, DC 20545

J. R. Maher
ER-65, GTN
U.S. Department of Energy
Washington, DC 20545

C. R. Mandelbaum
ER-32, GTN
U.S. Department of Energy
Washington, DC 20545

J. Mangeno
Department of the Navy
Nuclear Propulsion Directorate
0808 National Center ND2,
3rd Floor Mail Room
Washington, DC 20362-5101

B. Manowitz
Energy and Environment
Division
Brookhaven National
Laboratory
Upton, Long Island, NY 11973

R. S. Marianelli
Office of Basic Energy Science
U.S. Department of Energy
Washington, DC 20545

O. D. Markham
Radiological and Environmental
Sciences Laboratory
U.S. Department of Energy
785 DOE Place
Idaho Falls, ID 83402

K. Marsh
Lawrence Livermore National
Laboratory
P.O. Box 808
Livermore, CA 94550

W. H. Matchett
Graduate School
New Mexico State University
Box 3G
Las Cruces, NM 88003-0001

R. A. Mayes
Chicago Operations Office
U.S. Department of Energy
9800 South Cass Avenue
Argonne, IL 60439

H. M. McCammon
ER-75, GTN
U.S. Department of Energy
Washington, DC 20545

C. B. Meinhold
Radiological Sciences Division
Bldg. 703M
Brookhaven National
Laboratory
Upton, Long Island, NY 11973

M. L. Mendelsohn
Biomedical and Environmental
Research Program
Lawrence Livermore National
Laboratory, L-452
University of California
P.O. Box 5507
Livermore, CA 94550

F. J. Molz
Department of Civil Engineering
Auburn University
238 Harbert Engineering Center
Auburn, AL 36849

J. J. Morgan
California Institute of
Technology
Mail Code 138-78
Pasadena, CA 91125

W. F. Mueller
New Mexico State University
Box 4500
Las Cruces, NM 88003-4500

I. Murarka
Electric Power Research
Institute
3412 Hillview Avenue
P.O. Box 10412
Palo Alto, CA 94304

T. Murphy, Director
Environmental Research
Laboratory
EPA - Corvallis
200 SW 35th St.
Corvallis, OR 97330

J. R. Naidu
Safety and Environmental
Protection Division
Brookhaven National
Laboratory
Building 535-A
Upton, Long Island, NY 11973

N. S. Nelson
Office of Radiation Programs
(ANR-461)
Environmental Protection
Agency
401 M Street, SW
Washington, DC 20460

J. M. Nenhold
Department of Wildlife
Resources
Utah State University
Logan, UT 84322-5200

W. R. Ney, Executive Director
National Council on Radiation
Protection and Measurements
7910 Woodmont Avenue
Suite 1016
Washington, DC 20014

C. R. Nichols
Idaho Operations Office
U.S. Department of Energy
785 DOE Place
Idaho Falls, ID 83404

Nuclear Regulatory
Commission
Advisory Committee on Reactor
Safeguards
Washington, DC 20555

E. O'Donnell
Mail Stop NLS-260
Nuclear Regulatory
Commission
Washington, DC 20555

W. S. Osburn, Jr.
ER-75, GTN
U.S. Department of Energy
Washington, DC 20545

R. E. Pacha
Department of Biological
Sciences
Central Washington University
Ellensburg, WA 98926

A. Patrinos
Office of Health and
Environmental Research
U.S. Department of Energy
19901 Germantown Rd.
Germantown, MD 20874

G. Petersen
Department of Agriculture
Pennsylvania State University
University Park, PA 16802

G. Pinder
Princeton University
School of Engineering/Applied
Science
Princeton, NJ 08544

W. Piver
National Institutes of
Environmental Health
Sciences
P.O. Box 12233
Research Triangle Park,
NC 27709

J. S. Puskin
Office of Radiation Programs
(ANR-461)
Environmental Protection
Agency

401 M Street, SW
Washington, DC 20460

R. Rabson
Division of Biological Energy
Research
ER-17, GTN
U.S. Department of Energy
Washington, DC 20545

D. P. Rall, Director
National Institutes of
Environmental Health
Sciences
P.O. Box 12233
Research Triangle Park,
NC 27709

J. M. Ramirez
Allied Corporation
Bendix-Kansas City Division
Dept. 144, Mail Code OB-29
P.O. Box 61959
Kansas City, MO 64141-6159

R. D. Reed, Chief
Rocky Flats Area Office
Albuquerque Operations Office
U.S. Department of Energy
P.O. Box 928
Golden, CO 80402-0928

Regional Director
EPA Regional Office
1200 6th Avenue
Seattle, WA 98101

D. Reichle
Oak Ridge National Laboratory
P.O. Box X
Oak Ridge, TN 37830

C. A. Reilly, Jr.
Argonne National Laboratory
Bldg. 203
9700 South Cass Avenue
Argonne, IL 60439

C. R. Richmond
Oak Ridge National Laboratory
4500N, MS-62523
P.O. Box 2008
Oak Ridge, TN 37831-6253

C. Riordan
Office of Research &
Development
Environmental Protection
Agency
Washington, DC 20460

B. Rittman
Department of Civil
Engineering
University of Illinois
Urbana, IL 61801

C. Roberts
Site and Health Standards
Nuclear Regulatory
Commission
Washington, DC 20555

J. S. Robertson
ER-73, GTN
U.S. Department of Energy
Washington, DC 20545

S. L. Rose
ER-73, GTN
U.S. Department of Energy
Washington, DC 20545

R. D. Rosen, Tech. Librarian
Environmental Measurements
Laboratory
U.S. Department of Energy
376 Hudson Street
New York, NY 10014

D. M. Ross
EH-121, GTN
U.S. Department of Energy
Washington, DC 20545

L. Sagan
Electric Power Research
Institute
3412 Hillview Avenue
P.O. Box 10412
Palo Alto, CA 94304

M. Schulman
ER-70, GTN
U.S. Department of Energy
Washington, DC 20545

R. S. Scott, Acting Director
Office of Environmental Audit
EH-24, FORS
U.S. Department of Energy
Washington, DC 20585

G. H. Setlock
Rockwell International
North American Space
Operations
Rocky Flats Plant, MS-T452A
P.O. Box 464
Golden, CO 80402

R. B. Setlow
Brookhaven National
Laboratory
Upton, Long Island, NY 11973

R. Shikier
Battelle - Seattle
4000 NE 41st Street
Seattle, WA 98105

D. B. Shieler
Battelle Memorial Institute
505 King Avenue
Columbus, OH 43201

P. H. Silverman
Lawrence Berkeley Laboratory
Donner Laboratory, Room 466
University of California
Berkeley, CA 94720

J. Simmons
Bioaffects Analysis Branch
Environmental Protection
Agency
401 M Street, SW
Washington, DC 20460

W. K. Sinclair, President
National Council on Radiation
Protection
7910 Woodmont Avenue
Suite 1016
Bethesda, MD 20814

J. Skalski
Center for Quantitative Science
HR-20
3737 15th Avenue, N
Seattle, WA 98195

D. A. Smith
ER-72, GTN
U.S. Department of Energy
Washington, DC 20545

G. S. Smith
New Mexico State University
Box 3-1
Las Cruces, NM 88003-0001

L. M. Smith
Marine Science Center
Environmental Protection
Agency
Newport, OR 97365

M. Smith, Director
Savannah River Ecology
Laboratory
University of Georgia
Savannah River Plant
P.O. Box A
Aiken, SC 29801

G. A. Smithwick
EH-20, FORS (7A-097)
U.S. Department of Energy
Washington, DC 20585

J. Snow
ER-6, FORS
U.S. Department of Energy
Washington, DC 20585

L. E. Sommers
Department of Agronomy
Colorado State University
Ft. Collins, CO 80523

J. N. Stannard
University of California
17441 Plaza Animado #132
San Diego, CA 92128

R. W. Starostecki
NE-40, GTN
U.S. Department of Energy
Washington, DC 20545

R. J. Stern
EH-10, FORS
U.S. Department of Energy
Washington, DC 20585

D. K. Stevens
ER-11, GTN
U.S. Department of Energy
Washington, DC 20545

L. F. Stickel
Patuxent Wildlife Research
Center
Laurel, MD 20810

E. T. Still
Kerr-McGee Corporation
P.O. Box 25861
Oklahoma City, OK 73125

P. M. Stone
ER-6
U.S. Department of Energy
Washington, DC 20585

J. M. Suflita
Department of Botany &
Microbiology
University of Oklahoma
Norman, OK 73019

G. Swartzman
Center for Quantitative Studies
3737 15th Avenue
University of Washington
Seattle, WA 98115

J. Swinebroad
EH-12, GTN
U.S. Department of Energy
Washington, DC 20545

Technical Information Service
Savannah River Laboratory
Room 773A
E. I. duPont de Nemours &
Company
Aiken, SC 29801

R. G. Thomas
ER-72, GTN
U.S. Department of Energy
Washington, DC 20545

L. O. Tiffin
Swan Valley Route
Seeley Lake, MT 59868

A. W. Trivelpiece
Oak Ridge National Laboratory
Oak Ridge, TN 37830

A. Valocchi
Department of Civil
Engineering
University of Illinois
Urbana, IL 61801

P. K. M. Van der Heijde
International Groundwater
Modeling Center
4600 Sunset Avenue
Indianapolis, IN 46208

C. R. Vest
Battelle, Pacific Northwest
Laboratory
Washington Operations
370 L'Enfant Promenade,
Suite 900
901 D Street, SW
Washington, DC 20024

G. L. Voelz
Los Alamos National
Laboratory
MS-K404
P.O. Box 1663
Los Alamos, NM 87545

B. W. Wachholz
Radiation Effects Branch
National Cancer Institute
EPN, Room 530
8000 Rockville Pike
Bethesda, MD 20892

R. A. Walters, Assistant to the
Associate Director
Los Alamos National
Laboratory
MS-A114
P.O. Box 1663
Los Alamos, NM 87545

W. W. Weyzen
Electric Power Research
Institute
3412 Hillview Avenue
P.O. Box 10412
Palo Alto, CA 94303

F. W. Whicker
Department of Radiology &
Radiation Biology
Colorado State University
Ft. Collins, CO 80521

R. P. Whitfield
DOE - Savannah River
Operations Office
P.O. Box A
Aiken, SC 29801

D. L. Willis
Department of General Science
Oregon State University
Corvallis, OR 97331

F. J. Wobber
U.S. Department of Energy
14 Goshen Court
Gaithersburg, MD 20879-4403

R. W. Wood
PTRD, OHER
ER-74, GTN
U.S. Department of Energy
Washington, DC 20545

R. G. Woodmansee
Department of Range Science
Colorado State University
Fort Collins, CO 80523

S. R. Wright
Savannah River Operations
Office
P. O. Box A
Aiken, SC 29801

Zhu Zhixian
Laboratory for Energy-Related
Health Research
University of California
Davis, CA 95616

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University of California Study
Center
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Abt. Nuklearchemie
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5300 Bonn 1
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Library & Information Services
Bhabha Atomic Research
Centre
Central Complex
Trombay, Bombay-400 085
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Sanitaire
Commissariat à l'Énergie
Atomique
BP No. 6
F-92265 Fontenay-aux-Roses
FRANCE

G. Bengtsson, Director-General
Statens Stralskyddsinstitut
Box 60204
S-104 01 Stockholm
SWEDEN

D. J. Beninson
Gerencia de Protección
Radiológica y Seguridad
Comisión Nacional de Energía
Atómica
Avenida del Libertador 8250
2° Piso Of. 2330
1429 Buenos Aires
ARGENTINA

A. Bianco
International Atomic Energy
Agency
V.I.C.
P.O. Box 200
A-1400 Vienna
AUSTRIA

A. Brink
SASOL-One Limited
P.O. Box 1
Sasolburg 9570
REPUBLIC OF SOUTH AFRICA

Cao Shu-Yuan, Deputy Head
Laboratory of Radiation
Medicine
North China Institute of
Radiation Protection
P.O. Box 120
Tai-yuan, Shan-Xi
THE PEOPLE'S REPUBLIC OF
CHINA

M. Carpentier
Commission of the European
Communities
200 rue de la Loi
J-70 6/16
B-1049 Brussels
BELGIUM

M. A. Carregado
Gerencia de Protección
Radiológica y Seguridad
Comisión Nacional de Energía
Atómica
Casilla de Carreo 40
1802 Aeropuerto Int.
Ezeiza
ARGENTINA

Chen Xing-An
Laboratory of Industrial
Hygiene
Ministry of Public Health
2 Xinkang Street
Deshengmenwai, Beijing
THE PEOPLE'S REPUBLIC OF
CHINA

R. Clarke
National Radiological Protection
Board
Harwell, Didcot
Oxon OX11 0RQ
ENGLAND

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Communities
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BELGIUM

Deng Zhicheng
North China Institute of
Radiation Protection
Tai-yuan, Shan-Xi
THE PEOPLE'S REPUBLIC OF
CHINA

G. M. Desmet
Commission of the European
Communities
DG XII
200 rue de la Loi
B-1049 Brussels
BELGIUM

Director
Commissariat à l'énergie
Atomique
Centre d'Etudes Nucléaires
Fontenay-aux-Roses (Seine)
FRANCE

Director
Commonwealth Scientific and
Industrial Research
Organization
Aspendal, Victoria
AUSTRALIA

Director
Laboratorio di Radiobiologia
Animale
Centro di Studi Nucleari Della
Casaccia
Comitato Nazionale per
l'Energia Nucleare
Casella Postale 2400
I-00100 Roma
ITALY

D. Djuric
Institute of Occupational and
Radiological Health
11000 Beograd
Deligradoka 29
YUGOSLAVIA

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Biblioteca
2780 Oeiras
PORTUGAL

L. Feinendegen, Director
Institut für Medizin
Kernforschungsanlage Jülich
Postfach 1913
D-5170 Jülich
FEDERAL REPUBLIC OF
GERMANY

R. M. Fry, Head
Office of the Supervising
Scientist for the Alligator
Rivers Region
P.O. Box 387
Bondi Junction NSW 2022
AUSTRALIA

G. B. Gerber
Radiobiology Department
Commission of the European
Communities
200 rue de la Loi
B-1049 Brussels
BELGIUM

A. R. Gopal-Ayengar
73-Mysore Colony
Mahul Road, Chembur
Bombay-400 074
INDIA

A. Grauby
Service d'Etudes et de
Recherches sur l'Environnement
Commissariat à l'énergie
Atomique
Caderache, No. 1
BAT 153 13115
St. Paul Les Durance
FRANCE

M. D. Hill
Assessments Department
National Radiological Protection
Board
Chilton, Didcot
Oxon OX11 0RQ
ENGLAND

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Agency
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A-1010 Vienna 1
AUSTRIA

K. E. Lennart Johansson
National Defense Research
Institute
FOA 45 1
S-901-82 Umeå
SWEDEN

H.-J. Klimisch
BASF Aktiengesellschaft
Abteilung Toxikologie, Z470
D-6700 Ludwigshafen
FEDERAL REPUBLIC OF
GERMANY

G. H. Kraft
c/o GSI Postbox 110541
Planck Str.
D-6100 Darmstadt
FEDERAL REPUBLIC OF
GERMANY

T. Kumatori, Director
National Institute of
Radiological Sciences
9-1, Anagawa-4-chome
Chiba-shi 260
JAPAN

J. R. A. Lakey, Director
Department of Nuclear
Sciences & Technology
Royal Naval College,
Greenwich
London SE10 9NN
ENGLAND

Li De-Ping, Professor and
Director of North China
Institute of Radiation
Protection, NMI
Tai-yuan, Shan-Xi
THE PEOPLE'S REPUBLIC OF
CHINA

Librarian
Australian Nuclear Science &
Technology Organization
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AUSTRALIA

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Freshwater Institute
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CANADA

Librarian
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SWITZERLAND

Librarian
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D-75 Karlsruhe 1
FEDERAL REPUBLIC OF
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Fisheries Laboratory
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JAPAN

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FRANCE

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DENMARK

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DENMARK

B. Lindell
National Institute of Radiation
Protection
Fack S-104 01
Stockholm 60
SWEDEN

A. M. Marko, Director
Atomic Energy Commission of
Canada, Ltd.
Biology and Health Physics
Division
Chalk River Nuclear
Laboratories
P.O. Box 62
Chalk River, Ontario KOJ 1J0
CANADA

H. J. Metivier
Chef de la Section de
Toxicologie et
Cancerologie Experimentale
Institut de Protection et de
Sûreté Nucléaire
Département de Protection
Sanitaire
Service de Pathologie
Experimentale
BP No. 12
91680 Bruyères-le-Châtel
FRANCE

C. Myttenaere
Laboratoire de Physiologie
Végétale (VYVE)
Place Croix du Sud, 4
B-1348 Louvain-La-Neuve
BELGIUM

J. C. Nènot, Deputy Director
Département de Protection
Centre d'Etudes Nucléaires
BP No. 6
F-92260 Fontenay-aux-Roses
FRANCE

J. P. Oliver
ENEA (OECD) Health and
Safety Office
38, Blvd. Suchet
Paris
FRANCE

O. Pavlovski
Institute of Biophysics
Ministry of Public Health
Givopisnaya 46
Moscow D-182
USSR

V. Prodi
Department of Physics
University of Bologna
Via Imerio 46
I-40126 Bologna
ITALY

Reports Librarian
Harwell Laboratory, Bldg. 465
UKAEA
Harwell, Didcot
Oxon OX11 ORB
ENGLAND

P. J. A. Rombout
Inhalation Toxicology
Department
National Institute of Public
Health and Environmental
Protection
P.O. Box 1
NL-3720 BA Bilthoven
THE NETHERLANDS

M. Rzekiecki
Commissariat à l'énergie
Atomique
Centre d'Etudes
Nucleaires de Cadarache
BP No. 13-St. Paul
Les Durance
FRANCE

G. Smith
National Radiological Protection
Board
Chilton, Didcot
Oxon OX11 ORQ
ENGLAND

H. Smith
International Commission on
Radiological Protection
P.O. Box 35
Didcot
Oxon OX11 ORJ
ENGLAND

J. W. Stather
National Radiological Protection
Board
Building 383
Chilton, Didcot
Oxon OX11 0RQ
ENGLAND

M. J. Suess
Regional Officer for
Environmental Hazards
World Health Organization
8, Scherfigsvej
DK-2100 Copenhagen
DENMARK

Sun Shi-quan, Head
Radiation-Medicine Department
North China Institute of
Radiation Protection, MNI
P.O. Box 120
Tai-yuan, Shan-Xi
THE PEOPLE'S REPUBLIC OF
CHINA

G. Tarroni
ENEA-PAS-FIBI-AEROSOL
Laboratorio Fisica Sanitaria
Via Ercolani 8
I-40138 Bologna
ITALY

J. W. Thiessen
Radiation Effects Research
Foundation
5-2 Hijiyama Park
Minami-Ku
Hiroshima 732
JAPAN

D. Van As
Atomic Energy Board
Private Bag X 256
Pretoria 0001
REPUBLIC OF SOUTH AFRICA

Vienna International Centre
Library
Gifts and Exchange
P.O. Box 100
A-1400 Vienna
AUSTRIA

Wang Hengde
North China Institute of
Radiation Protection
P.O. Box 120
Tai-yuan, Shan-Xi
THE PEOPLE'S REPUBLIC OF
CHINA

Wang Renzhi
Institute of Radiation Medicine
11# Tai Ping Road
Beijing
THE PEOPLE'S REPUBLIC OF
CHINA

Wang Ruifa, Associate Director
Laboratory of Industrial
Hygiene
Ministry of Public Health
2 Xinkang Street
P.O. Box 8018
Deshengmenwai, Beijing
100088
THE PEOPLE'S REPUBLIC OF
CHINA

Wang Yibing
North China Institute of
Radiation Protection
P.O. Box 120
Tai-yuan, Shan-Xi
THE PEOPLE'S REPUBLIC OF
CHINA

Wei Lu-Xin
Laboratory of Industrial
Hygiene
Ministry of Public Health
2 Xinkang Street
Deshengmenwai, Beijing
100088
THE PEOPLE'S REPUBLIC OF
CHINA

B. C. Winkler, Director
Licensing
Raad Op Atomic
Atoomkrag Energy Board
Privaatsk X 256
Pretoria 0001
REPUBLIC OF SOUTH AFRICA

Wu De-Chang
Institute of Radiation Medicine
27# Tai Ping Road
Beijing
THE PEOPLE'S REPUBLIC OF
CHINA

ONSITE

DOE Richland Operations
Office (2)

E. C. Norman
M. W. Tiernan/R. D. Freeburg

Hanford Environmental Health
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