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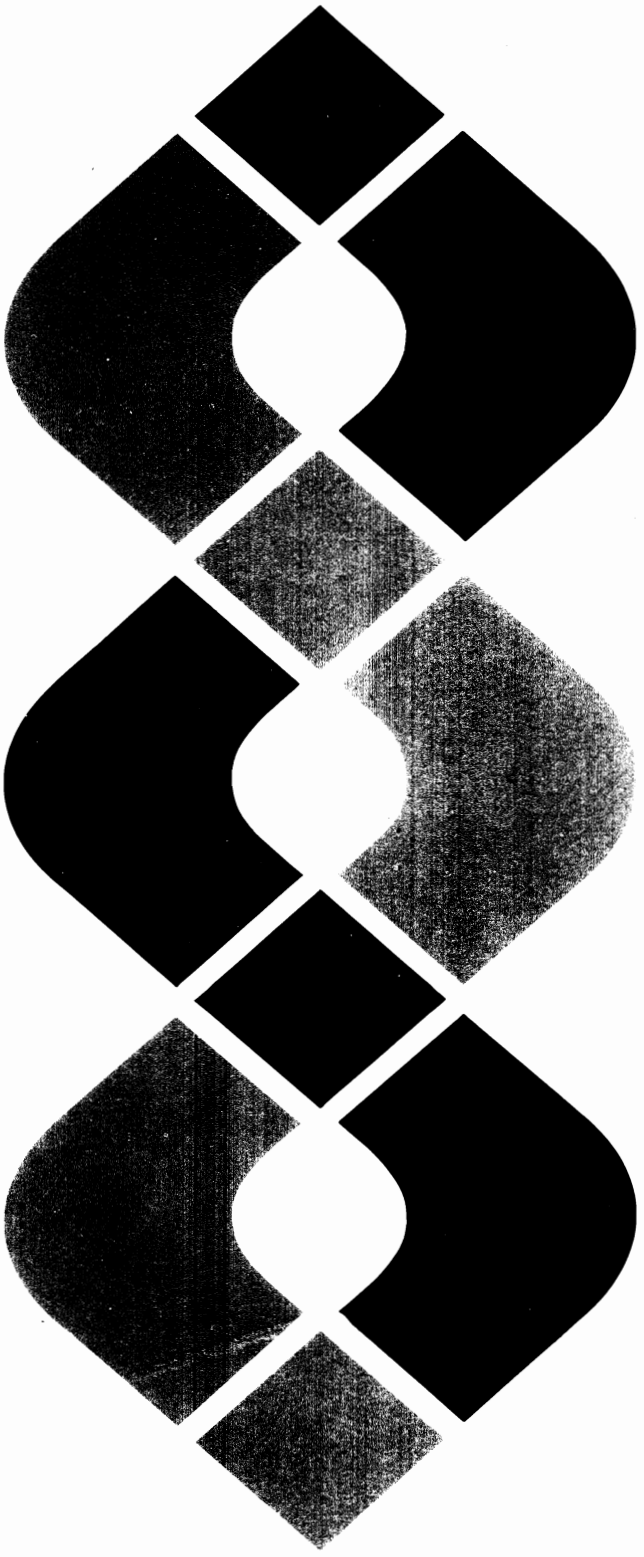

Pacific Northwest Laboratory
Annual Report for 1977
to the DOE Assistant Secretary
for Environment

February 1978

Part 2

Ecological Sciences

Prepared for the
U.S. Department of Energy
under Contract EY-76-C-06-1830

Battelle
Pacific Northwest Laboratories

PNL-2500 PT2

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Pacific Northwest Laboratory Annual Report for 1977 to the DOE Assistant Secretary for Environment

Part 2 Ecological Sciences

by
B. E. Vaughan and Staff Members
of Pacific Northwest Laboratory

February 1978

Battelle
Pacific Northwest Laboratories
Richland, Washington 99352

PREFACE

The 1977 Annual Report from Pacific Northwest Laboratory (PNL) to the DOE Assistant Secretary for Environment introduces a new cover. The earth-green color used on past annual reports has been replaced by the "environmental colors," blue and green. The cover's abstract design is not intended to represent anything specific, but we would not be unhappy if it suggests something environmental or biological to the reader. The blue and green color pattern on the cover is different for each part of this report to help distinguish the five parts.

The five parts of the Report are oriented to particular segments of our program. Parts 1-4 report research performed for the DOE Office of Biomedical and Environmental Research. Part 5 reports progress on all other research performed for the Assistant Secretary for Environment, including the Office of Environmental Control Technology, Office of Technology Impact, and Office of Operational and Environmental Safety.

Each part consists of project reports authored by scientists from several PNL research departments, reflecting the interdisciplinary nature of the research effort. Parts 1-4 are organized primarily by energy technology, although it is recognized that much of the research performed at PNL is applicable to more than one energy technology.

The parts of the 1977 Annual Report are:

Part 1: Biomedical Sciences		
	Program Manager - W. R. Wiley	R. C. Thompson, Report Coordinator D. L. Felton, Editor
Part 2: Ecological Sciences		
	Program Manager - B. E. Vaughan	B. E. Vaughan, Report Coordinator J. L. Helbling, Editor
Part 3: Atmospheric Sciences		
	Program Manager - C. L. Simpson	R. L. Drake, Report Coordinator C. M. Gilchrist, Editor
Part 4: Physical Sciences		
	Program Manager - J. M. Nielsen	J. M. Nielsen, Report Coordinator G. M. Garnant/L. Carson, Editors

Part 5: Control Technology, Overview, Health, Safety, and Policy Analysis

Program Managers - N. E. Carter
D. B. Cearlock
J. C. Fox
D. L. Hessel
H. V. Larson
S. Marks

W. J. Bair, Report Coordinator
R. W. Baalman, Editor

Activities of the scientists whose work is described in this Annual Report are broader in scope than the articles indicate. Knowledge and experience obtained by PNL staff in carrying out research in the Environment, Health, and Safety Research program have contributed to many other DOE interests. These include assistance in the preparation of several Environmental Development Plans for the Assistant Secretary for Environment, preparation of environmental statements for which the Laboratory is responsible, key membership in several national and international organizations, and numerous responses to the media on research projects of public interest.

W. J. Bair, Manager
S. Marks, Associate Manager
Environment, Health, and Safety Research Program

Previous Reports in this Series:

Annual Report for

1951	W-25021, HW-25709
1952	HW-27814, HW-28636
1953	HW-30437, HW-30464
1954	HW-30306, HW-33128, HW-35905, HW-35917
1955	HW-39558, HW-41315, HW-41500
1956	HW-47500
1957	HW-53500
1958	HW-59500
1959	HW-63824, HW-65500
1960	HW-69500, HW-70050
1961	HW-72500, HW-73337
1962	HW-76000, HW-77609
1963	HW-80500, HW-81746
1964	BNWL-122
1965	BNWL-280, BNWL-235, Vol. 1-4, BNWL-36I
1966	BNWL-480, Vol. 1, BNWL-481, Vol. 2, Pt 1-4
1967	BNWL-714, Vol. 1, BNWL-715, Vol. 2, Pt 1-4
1968	BNWL-1050, Vol. 1, Pt. 1-2, BNWL-1051, Vol. 2, Pt. 1-3
1969	BNWL-1306, Vol. 1, Pt. 1-2, BNWL-1307, Vol. 2, Pt. 1-3
1970	BNWL-1550, Vol. 1, Pt. 1-2, BNWL-1551, Vol. 2, Pt. 1-2
1971	BNWL-1650, Vol. 1, Pt. 1-2, BNWL-1651, Vol. 2, Pt. 1-2
1972	BNWL-1750, Vol. 1, Pt. 1-2, BNWL-1751, Vol. 2, Pt. 1-2
1973	BNWL-1850, Pt. 1-4
1974	BNWL-1950, Pt. 1-4
1975	BNWL-2000, Pt. 1-4
1976	BNWL-2100, Pt. 1-5

FOREWORD

In Part 2 of these volumes, all work funded in the Ecological Sciences by DBER has been brought together. This covers ecological effort in the following budget activity categories:

RK-02-03-01 (Biotic Transport and Conversion)
RK-02-03-02 (Environmental Effects)
RK-02-03-03 (Energy Related Supporting Research)
RK-02-03-04 (Environmental Operations and DOE Support)

For convenience, organization charts for the several participating departments and sections of PNL will be found at the back of the report.

For this annual report, we have grouped individual research projects (schedule 189's) according to the energy technology area they primarily support. Schedule 189's are indicated at the top of each blue divider page by boldface dot and title followed by a description of how these efforts are important to technology considerations. In the abstracts that follow, principal investigators are listed, who may be contacted for further information about individual projects. This format should make the report useful not only to DBER, but to managers of technology programs as well. We hope the technology manager can go to an appropriate blue section of this report, then find immediately those ecological problems PNL is working on that are pertinent to his concerns. In several instances; e.g., shale oil program, coordinated efforts lie across biomedical and organic chemistry areas outside the purview of this report. We have cross referenced to appropriate parts of the four-volume Annual Report, in those cases where companion work exists in another volume.

In ongoing discussions with technology representatives, a frequent misunderstanding turns up over measurement levels suitable for environmental acceptability, as regards energy residuals. In most cases, any such numbers are highly provisional and cannot be applied generally to different landscape or other habitat types--a point not generally recognized. Much in existing ecological data bases deals with environmental processes generically, whereas the measurement of technological impact will usually require data on interacting communities of organisms at pertinent locations. Also, organisms of the natural environment show extremely wide range in their susceptibility to damage, depending on climatic, ecosystems, and geologic factors of a variable nature. For these reasons, impact assessment and optimal management strategies must be site specific. In research reported here, those investigations concerned with underlying environmental processes are reported in the Multitechnology Section. Investigations that have been systematized well enough to treat technology problems directly have been reported in an appropriate technology section.

Numerous projects described herein arise out of PNL's direct experience with industry and other agencies concerned with energy installations, including the Nuclear Regulatory Commission, Electric Power Research Institute, U.S. Army Corps of Engineers, American Petroleum Institute, private utility companies, and the Environmental Protection Agency. Opportunities for PNL to contribute to the preparation or review of environmental impact assessments have provided valuable insight into research needs and many of those needs have, indeed, been implemented, with DBER's support, in the programs reported here. Several studies were prompted also by other divisions of the DOE as interdigitating effort with the DBER program, notably work for the Division of Magnetic Fusion Energy (DMFE) on tritium, lithium, and other fusion products. This includes, also, work for the Division of Nuclear Fuel Cycle and Production (DNFCP) funded through intercontractor agreements with the Rockwell-Hanford Co. (radioecological problems concerned with fuel reprocessing and waste sites).

The Hanford site was formally dedicated as a National Environmental Research Park (NERP), on March 18, 1977. In establishing the Hanford NERP, major segments of the 750 square-mile site have been set aside for applied research in ecology, open to all qualified research scientists. Charter intent is to stimulate: 1) baseline observations, 2) field studies of response to man-made disturbances, 3) predictive modeling of impacted ecosystems, and 4) public demonstration of alternate land use at the energy activity sites. Use policy and procedures have evolved from those developed earlier on the Arid Lands Ecology (ALE) Reserve. Within the Hanford NERP, the ALE Reserve (110 square miles) remains a protected area intended for long-range modulation studies (i.e., nondestructive studies) of grasslands and desert steppe shrub ecosystems. A brochure has been prepared describing the NERP.[a] Research publications will be listed in another report.[b]

Among other developments, Dr. Burton E. Vaughan appeared before the Honorable George E. Brown's Subcommittee on Environment and Atmosphere, July 28, 1977, to describe PNL's experience in operating the ALE Reserve over the past decade. During this time, the ALE Reserve appears to be the only national location that has been maintained fully to NSF standards for Biosphere Reserves. Also, Dr. Raymond E. Wildung, PNL, was appointed to the NAS/NRC Committee on Accessory (Trace) Elements, with membership in three subpanels, Coal, Oil Shale, and Uranium. Apart from organic residuals from fossil fuel processes, increasing environmental burdens of trace elements continue to occupy attention. During this reporting year a refereed symposium volume was published, covering the biological implications of trace metal accumulation.(c) Also, Dr. Richard F. Foster, PNL, assumed responsibility to develop Environmental Standard Review Plans. This project is designed to bring to better focus proper environmental issues, to carry out environmental analysis with scientific rigor, and to provide consistency both in approach and criteria, for assessments in support of license application.

Burton E. Vaughan
Subprogram Manager
Ecological Sciences

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Richland, WA 99352

(b) A Bibliography of Environmental Research
BNWL-SA-4655 Rev. 3
Ecosystems Department
Battelle-Pacific Northwest Laboratories
P.O. Box 999
Richland, WA 99352

(c) Biological Implications of Metals in the Environment
H. Drucker and R. E. Wildung (eds.) June 1977
CONF 750929 Natl. Tech. Info. Service
U.S. Dept. Commerce
Springfield, VA 22161

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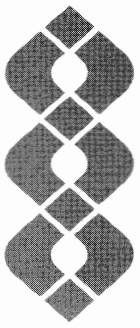

1.0 Land Use and Coal Technology

LAND USE AND COAL TECHNOLOGY

- **Hanford National Environmental Research Park (NERP) and ALE Administration**
- **Terrestrial Ecology**
- **Restoration of Surface-Mined Lands**
- **Fossil Fuel Effluents in Freshwater Ecosystems**

The U.S. Department of Energy has extensive land holdings in the arid and semi-arid regions of the Western United States. One of the most strategically situated land holdings is in the State of Washington. The Hanford Site currently contains the obsolete facilities developed for wartime plutonium production and also contains large amounts of radioactive waste materials. To provide a measure of safety and security, production facilities were buffered by a large land area unoccupied and unused by people. Today, this buffer zone provides some of the most desirable land for long- and short-term terrestrial and aquatic ecological baseline research in the United States. In recognition of this research potential, the Arid Lands Ecology Reserve and the Hanford National Environmental Research Park were established to promote the use of the Hanford Site for ecological research, especially studies related to energy technologies and their potential for environmental impacts.

Coal is currently regarded as the most dependable interim source of energy in the United States. To meet expected demands, coal needs to be mined in large quantities and may be mined predominantly in locations of sparse precipitation. Often the most economical way to extract coal is through surface mining. It is expected that following coal extraction the pits will be filled with overburden, graded to approximate original contour, native topsoil applied to prescribed depths and planted with climatically adapted herbs, shrubs or trees. Because primary productivity in dry regions is characteristically low, it is realistic to expect, if the above procedure is followed, that the revegetated surfaces will also produce little phytomass in the years following restoration. Appropriate data are needed for accurate estimation of the economic feasibility of a particular restoration practice or its alternative.

Throughout 1977 Annual Report:

- Bullets denote 189 titles.

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Research on land restoration at PNL is aimed at purposeful modifications of topography to collect and direct rainfall (water harvest) and to determine the best arrangement of suitable soil in an engineered design to produce a more favorable environment for plant growth. A better plant growth environment permits a wider selection of plant species, some of which could be harvested as food. The purpose is to provide a way to grow useful plants on reclaimed surface mines in arid regions without a need for irrigation. A brief description of the proposed experimental design and some initial data are included in this section of the Annual Report.

The current environmental program in synfuels at PNL concerns: 1) chemical characterization of residuals from SRC, CO_2 receptor, and several in-situ coal burning facilities (See PNL 2500, Part IV, Physical and Technological Programs), 2) occupational health and toxicological studies (See PNL 2500, Part I, Biomedical Sciences), and 3) potential impact studies on organisms of the natural environment (this report). Dr. Sidney Marks is coordinator of the comprehensive program.

Coal conversion is designed to eliminate or greatly reduce the noxious elements of conventional coal combustion that are released as waste byproducts. However, aqueous effluents from coal conversion may have deleterious effects on biota inhabiting lakes, ponds and streams. Conventional coal combustion steam-electric stations characteristically release CO_2, SO_2 ozone and trace metals into the atmosphere which present certain long-term ecological problems to terrestrial habitats.

This section of the annual report is divided into two segments. The first deals with land use and the second part concerns the effects of coal byproducts on freshwater ecosystems.

LAND USE

- **Hanford National Environmental Research Park (NERP) and ALE Administration**
- **Terrestrial Ecology**
- **Land Restoration**

Principal Investigators: W. H. Rickard, L. E. Rogers and R. H. Sauer

Other Investigators: R. E. Fitzner, D. W. Uresk, J. F. Cline and J. M. Thorp

Technical Assistants: H. A. Sweany, M. J. Harris, M. A. Combs, C. A. Lee, V. D. Charles, K. A. Gano and L. F. Nelson

Norcus Appointees: J. N. Fitzner and W. D. Steigers

Consultant: J. T. Rotenberry (Ornithology)

The National Environmental Research Park (NERP) encourages the use of Hanford Site ecological habitats and facilities through graduate students from universities and colleges and for senior investigators with funding support from sources other than U.S. DOE. Terrestrial ecology studies are mostly confined to the Arid Lands Ecology Reserve and are concerned with short and long-term impacts relating to energy technologies and perturbations of natural origin in shrub-steppe ecosystems. These projects provide the kind of ecological information needed to meet the intent of the National Environmental Policy Act (NEPA) as well as advancing the state of ecological knowledge in the scientific community at large. Land restoration studies are aimed at testing alternative ways to successfully revegetate surface-mined lands in the most arid parts of the Western United States.

HANFORD NATIONAL ENVIRONMENTAL RESEARCH PARK (NERP) AND ALE ADMINISTRATION

NERP and ALE Administration Progress

The Hanford National Environmental Research Park (NERP) was dedicated in March 1977. The NERP includes the Arid Lands Ecology (ALE) Reserve a 120 square mile land area located on the western edge of the Hanford Site dedicated to ecological research. The Reserve is administered by the Pacific Northwest Laboratories. The remainder of the Hanford Site is also available for ecological research but administrative NERP decisions are subject to approval by other U.S. DOE contractors and the Richland Operations office (RL).

One of the important purposes of the NERP is to promote the use of U.S. DOE lands for ecological research of national and regional interest funded by sources other than U.S. DOE. In calendar year 1977, three different offsite studies were granted access to the

ALE Reserve. All are funded by the National Science Foundation. These are listed below:

Organization	Investigator	Research topic
Wash. State Univ.	Dr. R. N. Mack	Plant demography
Univ. of Wash.	Dr. G. J. Kenagy	Ecophysiology of rodents
Oregon State Univ.	Dr. J. A. Wiens	Ecology of shrub-steppe birds

These studies add to the ecological data base of the ALE Reserve, which has been accumulating since 1967, as well as providing a carefully protected research location for field research by the offsite investigators.

Another purpose of the NERP is to provide facilities to onsite NORCUS graduate student appointees to conduct research projects that are approved thesis topics. Two appointees worked on the NERP in 1977. They are listed below:

Student Appointee	Sponsoring University	Thesis project
J. Fitzner	Wash. State Univ.	Life history long-billed curlew (Ph.D.)
W. Stiegers	Brigham Young Univ.	Radiotracking mule deer fawns (MS)

These studies were conducted on NERP lands other than the ALE Reserve. Satisfactory progress was made in both projects. PNL provided transportation, laboratory and office space, radiotransmitters and receivers, and specialized consultant services.

A third task is to provide surveillance of the ALE Reserve and the western boundary of the NERP against unauthorized public trespass and stray livestock intrusions that would interfere with ongoing long-term ecological studies. Fence maintenance and airplane patrol service were performed by a private contractor.

L. E. Rogers and R. E. Fitzner participated in the NERP Symposium held at the Los Alamos Scientific Laboratory, New Mexico, in August 1977. Their presentations will be published in the symposium proceedings in 1978.

Use of the Hanford NERP by the scientific community was promoted by selected visits to universities in the Pacific Northwest and to an international audience in Iceland by W. T. Hinds.

The designation of the Hanford Site as a National Environmental Research Park is expected to accelerate the orderly accumulation of ecological information concerning the shrub steppe ecosystem in North America and make it one of the scientifically best known arid land areas in the United States.

TERRESTRIAL ECOLOGY

Primary Productivity: Response to the 1977 Drought

The plant growing season for 1977 was characterized by unusually low precipitation. Precipitation at three study sites harvested annually for aboveground primary productivity during the October to May growing season totaled 8.84 cm at upper Snively field, 6.63 cm at lower Snively field, and 6.05 cm at ALE Headquarters.

Productivity of herbaceous plants, as measured by hand clipping of live parts at peak yield at three study sites, was the lowest measured in 10 yr of record (Table 1.1).

TABLE 1.1. Average Herbage Production g/m^2 ± SE (dry wt) of Three Plant Communities on the ALE Reserve During the 1977 Growing Season (n=20)

Community	Herbage	Reproductive Parts[a]	Total
Cheatgrass (upper)	66 ± 11	13 ± 1.2	79
Cheatgrass (lower)	9 ± 3	5 ± 2.0	14
Bunchgrass (ALE)	9 ± 3	Trace	9

[a] Reproductive parts = flowering heads of cheatgrass with ripened caryopses.

Threatened and Endangered Species

The American bald eagle has historically used the Hanford Reach of the Columbia River as a winter foraging ground and records have been made of their abundance since the 1960's (Figure 1.1). The eagle population increased from a low level of about five birds in the 1960's to a high of about 15 birds in the 1970's. Eagles generally appear in mid-November, reach peak numbers in late November through January, and begin to leave by mid-February. By March, all eagles have usually departed. An interesting change in population

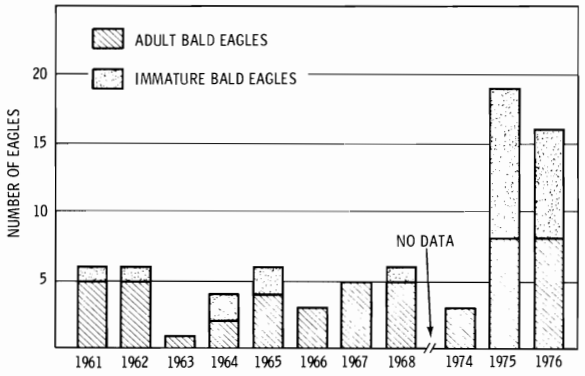

FIGURE 1.1. Peak Abundance of Bald Eagles on the Hanford Site During the Years 1961 - 1976

structure occurred in 1975 and 1976 when about one-half of the population was comprised of immature birds.

Although the bald eagle population has been generally declining throughout the United States, the winter population along the Columbia River appears to be increasing. All the reasons for the apparent increase in eagle use of the Hanford Site are, of course, not known but the presence of spawned-out salmon and the availability of hunter-wounded waterfowl that make their way back to the refugium of the Hanford Site are probably attractants as well as the isolation from people provided by the Hanford Site boundaries.

Invertebrate Studies: Darkling Beetle Distribution

Darkling beetles characteristically occur in species assemblages or groups of overlapping populations. There have been very few investigations designed to document how these assemblages are distributed through their environment.

The seasonal distribution of adult darkling beetles occupying the Hanford Site shows that adults tend to be present through much of the year when weather conditions are most favorable (March to October). Others, such as Philolithus densicollis and Stenomorpha puncticollis, are restricted to only a few months of the year, September to November. Eleodes obscura probably occurs earlier than May and later than August. This is a scarce (although conspicuous) beetle and has probably just not been collected during these periods. Eleodes humeralis occurs from June to October but is present in very limited numbers except during the August-September period, when it is fairly abundant.

Table 1.2 shows the percentage distribution of darkling beetle captures among three soil texture classes: sand, sandy-loam, and loam. The term loam, as used here, describes soil texture comprised of less than 50% sand; sandy-loam refers to soil containing 50-70% sand; and sandy soil consists of more than 70% sand. Some species, such as Anemia californica, Conisattus nelsoni, and Eusattus muricatus, were only found in sandy soil and are probably limited to this soil type. Blapstinus was restricted to the loam soil type, but the situation is less clear than for the sand-inhabiting species. Blapstinus greatest abundance occurred in Upper and Lower Snively old fields, so they may be more closely associated with the cheatgrass community type rather than soil texture per se. A few (9% of total captures) occurred in the hopsage study location, possibly indicating an association with loam soils. This study site is dominated by hopsage with a sparse understory of cheatgrass and is located at lower elevations near sandy soil study locations where Blapstinus was absent. Oxygonodera hispidula occurs in several community types all of which have loamy soils. It is most abundant in the sagebrush/bunchgrass community where it commonly is found within tussocks of bluebunch wheatgrass. It will probably never occur extensively where this community type is largely absent. Coniontis setosa occurred at nearly all study sites but was only abundant on loamy soils. This species was particularly abundant in cheatgrass and hopsage communities. Two congeneric species, C. lanei and C. ovalis, occur at higher elevations on Rattlesnake Mountain. None of the Coniontis species appear to be an important component of the low elevation darkling beetle fauna. Philolithus densicollis was widely distributed across all soil textures. Philolithus adults comprise an important segment of the

TABLE 1.2. Distribution of Darkling Beetle Captures Across Soil Texture Classes

Species	Sand	Sandy-Loam	Loam	Total Number Collected
	------ % ------			
Anemia californica	100	0	0	8
Blapstinus sp.	0	0	100	86
Coniontis setosa	7	7	86	108
Conisattus nelsoni	100	0	0	302
Eleodes granulata	0	92	8	25
Eleodes hispilabris	56	27	17	98
Eleodes humeralis	68	16	16	6
Eleodes nigrina	68	13	18	22
Eleodes novoverrucula	7	21	72	29
Eleodes obscura	50	0	50	2
Eusattus muricatus	100	0	0	57
Oxygonodera hispidula	0	7	93	46
Philolithus densicollis	25	60	15	419
Stenomorpha puncticollis	0	38	62	197

ground-dwelling beetle population at lower elevations during fall months. Stenomorpha puncticollis occurred primarily in areas with loam and sandy-loam soils. These beetles are restricted to the higher elevations of the ALE Reserve and are probably not restricted to any specific soil texture types.

It seems reasonable to assume that soil type is of critical importance in relation to larval development. Adults, being mostly wingless, are probably more or less restricted to small areas where the larvae develop. Habitat selection, therefore, is possibly influenced by two factors: 1) oviposition preferences by adult females, and 2) differential mortality of larvae in various soil textures.

Avifauna of an Isolated Streamside Plant Community

Snively Canyon supports a permanent spring brook and a dense, deciduous tree-shrub plant community along its shore. The canyon is unique in that it is surrounded by a large matrix of native shrub-steppe vegetation that has not been cultivated nor grazed by domestic livestock for a period of 35 yr. Unlike other spring-brook habitats in southeastern Washington, Snively Canyon is virtually free from interference resulting from the presence of a resident human population. For these reasons, a record of bird use of the canyon plant community represents as near pristine condition as can be found anywhere in the steppe region.

The results of the 30 surveys conducted over a 2 yr period are summarized in Table 1.3. Although the table may appear overly-detailed, we feel the uniqueness of this area (particularly the lack of human disturbance and its relative rarity and isolation) warrant the inclusion of these data for subsequent comparative purposes. Altogether 3,298 individuals of 81 species were recorded; the average count contained about 105 individuals of 16 species. Of these 81 species, 34 are "accidental" or of irregular occurrence within the canyon, even though a few are abundant in the surrounding shrub-steppe. Nineteen of the remaining 47 may be considered "dominant," which is to say that each contributed at least 1% (33) of the total number of individuals seen. In the absence of a more rigorous analysis these dominants may be considered diagnostic of the two major seasonal assemblages of birds: spring/summer breeders and fall/winter/spring migrants.

TABLE 1.3. Thirty Surveys Conducted Over 2 yr Period for Bird Use of Snively Canyon

Species	Status	# Individuals	# Counts	"Diversity"
Goshawk *(Accipiter gentilis)*	A	2	2	2.0
Cooper's Hawk *(A. cooperii)*	A	3	3	3.0
Sharp-shinned Hawk *(A. striatus)*	A	1	1	1.0
Marsh Hawk *(Circus cyaneus)*	R/B	15	11	9.8
Rough-legged Hawk *(Buteo lagopus)*	A	2	1	1.0
Red-tailed Hawk *(B. jamaicensis)*	M/-	7	6	5.4
Swainson's Hawk *(B. swainsoni)*	A	2	1	1.0
Golden Eagle *(Aquila chrysaetos)*	M/-	7	5	4.5
Merlin *(Falco columbarius)*	A	6	2	1.8
Kestrel *(F. sparverius)*	M/B	18	14	10.8
Sage Grouse *(Centrocercus urophasianus)*	R/B	15	2	1.1
California Quail *(Lophortyx californicus)*	R/B	37	9	3.5
Chukar *(Alectoris chukar)*	R/B	323	21	8.2
Ring-necked Pheasant *(Phasianus colchicus)*	R/B	32	12	9.1
Killdeer *(Charadrius vociferous)*	A	1	1	1.0
Band-tailed Pigeon *(Columba fasciata)*	A	2	1	1.0
Mourning Dove *(Zenaida macroura)*	M/B	347	18	10.4
Great Horned Owl *(Bubo virginianus)*	AX	1	1	1.0
Long-eared Owl *(Asio otus)*	M/B	10	7	6.3
Common Nighthawk *(Chordeilus minor)*	A	4	1	1.0
Rufous Hummingbird *(Selasphorus rufous)*	A	4	3	2.7
Common Flicker *(Colaptes cafer)*	M/-	25	14	11.0
Hairy Woodpecker *(Dendrocopus villosous)*	M/-	4	4	4.0
Eastern Kingbird *(Tyrannus tyrannus)*	M/B	105	5	1.3
Western Kingbird *(T. verticalis)*	A	3	3	3.0
Say's Phoebe *(Sayornis saya)*	A	1	1	1.0
Willow Flycatcher *(Empidonax trallii)*	M/-	8	4	3.6
Dusky Flycatcher *(E. oberholseri)*	A	1	1	1.0
Western Wood Pewee *(Contopus sordidulus)*	M/-	15	6	4.8
Olive-sided Flycatcher *(Nuttallornis borealis)*	A	1	1	1.0
Horned Lark *(Eremophila alpestris)*	R/B	32	11	5.8
Barn Swallow *(Hirundo rustica)*	M/B	112	14	8.9
Tree Swallow *(Iridoprocne bicolor)*	A	1	1	1.0
Black-billed Magpie *(Pica pica)*	R/B	99	25	14.0
Raven *(Corvus corax)*	R/B	13	9	7.3
House Wren *(Troglodytes aedon)*	A	2	1	1.0
Winter Wren *(T. troglodytes)*	M/-	9	6	4.8
Bewick's Wren *(Thyomanes bewickii)*	A	1	1	1.0
Rock Wren *(Salpinctes obsoletus)*	M/B	15	11	7.8
Canyon Wren *(Catherpes mexicanus)*	A	1	1	1.0
Long-billed Marsh Wren *(Telmatodytes palustris)*	A	2	1	1.0
Robin *(Turdus migratorius)*	M/-	46	13	8.6
Varied Thrush *(Ixoreus naevius)*	M/-	19	9	6.1
Townsend's Solitaire *(Myadestes townsendi)*	A	4	3	2.7
Hermit Thrush *(Hylocichla guttata)*	M/-	13	3	1.9
Golden-crowned Kniglet *(Regulus satrapa)*	M/-	24	5	4.2
Ruby-crowned Kinglet *(R. calendula)*	M/-	181	14	3.5
Northern Shrike *(Lanius excubitor)*	A	2	1	1.0
Loggerhead Shrike *(L. ludovicianus)*	R/B	9	7	6.2
Starling *(Sturnus vulgaris)*	M/B	154	11	7.9
Solitary Vireo *(Vireo solitarius)*	M/-	19	3	1.5
Red-eyed Vireo *(V. olivaceus)*	A	4	1	1.0
Warbling Vireo *(V. gilvus)*	A	4	2	2.0
	A	6	3	2.0
Nashville Warbler *(Vermivora ruficapilla)*	A	32	12	9.1
Yellow Warbler *(Dendroica petechia)*	A	4	2	1.6
Yellow-rumped Warbler *(D. coronata)*	M/-	88	7	3.0
Townsend's Warbler *(D. townsendi)*	M/-	17	3	1.6
Yellow-breasted Chat *(Icteria virens)*	M/B	15	8	6.8
MacGillivray's Warbler *(Oporornis tolmiei)*	M/-	10	3	2.8
Wilson's Warbler *(Wilsonia pusilla)*	M/B	44	5	2.9
Western Meadowlark *(Sturnella neglecta)*	R/B	461	23	14.7
Red-winged Blackbird *(Agelaius phoenecius)*	A	4	3	2.7
Brewer's Blackbird *(Euryphagus cyanocephalus)*	M/B	36	8	2.7
Brown-headed Cowbird *(Molothrus ater)*	M/B	12	3	2.3
Northern Oriole *(Icterus galbula)*	M/B	84	10	8.0
Western Tanager *(Piranga ludoviciana)*	M/B	7	5	2.5
Black-headed Grosbeak *(Pheucticus melanocephalus)*	A	2	2	2.0
Evening Grosbeak *(Hesperiphonia vespertina)*	A	3	1	1.0
Lazuli Bunting *(Passerina amoena)*	M/B	62	11	8.6
House Finch *(Carpodacus mexicanus)*	A	2	1	1.0
Pine Siskin *(Spinus pinus)*	M/-	88	7	3.0
American Goldfinch *(S. tristis)*	M/-	11	3	2.1
Rufous-sided Towhee *(Pipilo erythrophthalmus)*	A	6	3	2.0
Savannah Sparrow *(Passerculus sandwichensis)*	M/-	10	3	2.2
Vesper Sparrow *(Pooicetes gramineus)*	M/B	109	11	4.4
Lark Sparrow *(Chondestes grammacus)*	A	7	2	2.0
Sage Sparrow *(Amphispiza belli)*	A	3	3	3.0
Dark-eyed Junco *(Junco hyemalis)*	M/-	306	16	6.5
White-crowned Sparrow *(Zonotrichia leucophrys)*	M/-	126	12	5.5
Golden-crowned Sparrow *(Z. atricapilla)*	A	1	1	1.0
Song Sparrow *(Melospiza melodia)*	R/B	66	22	16.3

Status = pattern of occurrence on surveys
- A = "accidental", less than 10 individuals seen on less than four surveys
- M = migrants
- R = year-round residents
- B = breed in canyon
- - = do not breed in canyon

Seasonality of Litterfall in Steppe Shrubs

Leaffall is a conspicuous seasonal event in the deciduous forests of the eastern United States. It signals the end of the growing season and the onset of winter dormancy. Leaffall also occurs in western conifer forests but the process is not nearly so obvious because conifers usually retain leaves year-round. Leaffall occurs in steppe shrubs but the seasonal pattern of leaffall has not been described prior to this report.

Five species of steppe shrubs provide almost all the shrub cover on the Hanford Site. These are big sagebrush, Artemisia tridentata, spiny hopsage, Grayia spinosa, greasewood, Sarcobatus vermiculatus, bitterbrush, Purshia tridentata and rabbitbrush, Chrysothamnus nauseosus. All of these dominate shrub cover at particular places but big sagebrush is by far the most abundant species.

A special litterfall collector was made and placed beneath the canopy spread of individual bushes. The collector consisted of a decimeter long piece of nonmetallic irrigation pipe having an inside diameter of 15.6 cm. This section was cemented to a 0.5 dm length of the same diameter pipe with a piece of fine mesh nylon screen sandwiched between the two sections.

Greasewood is able to maintain a more or less full complement of leaves throughout the summer months because it obtains water by sending roots into a perched permanent water table. Leaffall in greasewood is completed with the onset of winter temperatures rather than the onset of summer drought as with hopsage. Hopsage is leafless for about 7 months each year and greasewood for about 4 months. Sagebrush and rabbitbrush are autumn blooming species that are never completely leafless. Litterfall contributions during fall and winter months for these plants mostly represents flower parts.

Seasonality of litterfall in steppe shrubs is a useful bit of knowledge that can be applied to ecological monitoring of the effects of salt drift from cooling towers associated with nuclear and fossil fueled energy stations as well as to the gaseous and particulate airborne effluents from coal-fired steam-electric stations expected to be built in the shrub-steppe regions of the Pacific Northwest in the near future. The main advantage of litterfall collection is that it requires little manpower, provides a continuously operable means of litter collection at all times, and is nondestructive. The same shrubs can be sampled year after year and the collected litterfall chemically analyzed for energy facility effluents (Table 1.4).

TABLE 1.4. Average + SE (n=4) Monthly Litterfall Grams per Collector for Five Shrub Species During 1976-77 on the Arid Lands Ecology Reserve

	Species				
Month	Greasewood	Hopsage	Bitterbrush	Rabbitbrush	Sagebrush[a]
May	.184 ± .052	.081 ± .038	.133 ± .035	.080 ± .025	.010 ± .010
% Total	3.6	3.4	4.5	2.1	3.7
June	.663 ± .175	.864 ± .290	.387 ± .057	.198 ± .050	.699 ± .161
% Total	13.1	36.1	13.1	5.1	25.6
July	.643 ± .157	1.008 ± .180	.854 ± .106	.306 ± .047	.807 ± .064
% Total	12.7	42.2	28.9	7.9	29.6
August	.612 ± .170	.441 ± .097	1.270 ± .441	.189 ± .021	.623 ± .093
% Total	12.1	18.4	43.0	4.9	22.8
September	.365 ± .059	0	.308 ± .130	.281 ± .055	.108 ± .015
% Total	7.2	0	10.4	7.2	4.0
October	.680 ± .099	0	0	.206 ± .035	.052 ± .007
% Total	13.5	0	0	5.3	1.9
November	1.933 ± .816	0	0	.562 ± .066	.130 ± .037
% Total	38.3	0	0	14.5	4.8
December	0	0	0	.227 ± .018	.021 ± .004
% Total	0	0	0	5.8	0.7
January	0	0	0	1.259 ± .129	.156 ± .030
% Total	0	0	0	32.4	5.7
February	0	0	0	.573 ± .187	.122 ± .019
% Total	0	0	0	14.8	4.5
March	0	0	0	0	0
% Total	0	0	0	0	0
April	0	0	0	0	0
Total	5.050	2.39	2.95	3.88	2.73

[a] (n = 8)

Mineral Content of Steppe-Shrub Litter: Baseline Values

Historically, the shrub-steppe region of Washington has been relatively free from airborne mineral depositions of energy facility effluents such as cooling tower drift and emissions from coal-fired steam-electric power stations. However, because of an anticipated need for additional electric power the low population areas of the arid shrub-steppe region are expected to be chosen as new sites for energy production stations either nuclear or fossil fueled.

Operating cooling towers and coal-fired stations are expected to add to the mineral content of steppe-shrubs through direct foliar retention and soil buildup over a long period of years. Unless mineral contents of steppe-shrub tissues are determined in advance, it is not possible to determine if increases in mineral content of shrub tissues actually results from the airborne effluents of energy facilities.

Chemical analyses of freshly fallen litter from five species of steppe-shrubs are presented in Table 1.5. These data show that greasewood litter is high in sodium, hopsage litter is high in potassium and magnesium, bitterbrush litter is high in calcium, magnesium and sulfur and sagebrush litter is high in iron and zinc.

TABLE 1.5. Chemical Analyses of Freshly Fallen Litter From Five Species of Steppe Shrubs

	%						ppm				
	S	P	K	Mg	Na	Ca	Fe	Mn	B	Cu	Zn
Greasewood											
Jun	0.56	0.28	2.7	0.16	5.8	0.80	910	150	52	22	26
Jul	0.49	0.12	1.5	0.11	8.8	0.65	700	150	47	12	14
Aug	0.51	0.11	1.5	0.12	9.0	0.65	410	150	62	11	8
Sep	0.38	0.06	0.91	0.11	7.0	0.77	330	130	55	8.5	7
Oct	0.58	0.08	1.9	0.13	10	0.65	390	120	51	8.5	8
Nov	0.58	0.10	1.4	0.12	10	0.65	390	130	55	10	10
Hopsage											
Jun	0.65	0.10	4.0	1.3	0.08	1.8	900	200	58	6	16
Jul	0.65	0.11	4.3	1.4	0.10	1.7	800	160	55	6	9
Aug	0.26	0.12	13	1.5	0.06	1.9	550	180	80	8	10
Rabbitbrush											
Jul	-	0.22	2.2	0.28	0.15	1.7	1600	140	85	22	13
Aug	-	0.18	1.1	0.32	0.10	2.2	940	150	52	14	19
Sep	0.36	0.08	1.0	0.26	0.00	1.9	930	130	75	20	21
Oct	0.32	0.16	1.8	0.32	0.09	1.6	630	110	90	20	16
Nov	0.36	0.08	1.3	0.28	0.10	1.6	660	120	100	22	19
Dec	0.37	0.11	1.4	0.40	0.04	1.7	320	130	62	13	10
Jan	-	-	-	-	-	-	-	-	-	-	-
Feb	0.30	0.11	1.2	0.37	0.04	1.8	600	120	57	13	10
Mar	0.17	0.05	0.76	0.28	0.03	2.1	450	120	62	12	10
Bitterbrush											
Jul	0.60	0.12	2.3	1.4	0.03	2.4	600	160	62	7	20
Aug	0.80	0.13	1.9	1.9	0.04	3.1	500	160	57	6	10
Sep	0.74	0.08	2.5	2.2	0.02	3.5	350	140	52	5	8
Oct	0.66	0.04	3.3	2.3	0.04	2.9	400	170	100	10	12
Sagebrush											
Jul	0.21	0.22	1.2	0.16	0.04	1.2	1500	150	60	15	26
Aug	0.23	0.17	1.0	0.18	0.04	1.6	1600	120	57	14	21
Sep	0.19	0.12	0.95	0.16	0.04	1.4	1400	130	57	16	24
Oct	0.23	0.20	1.1	0.16	0.07	1.2	1600	110	100	19	22
Nov	-	-	-	-	-	-	-	-	-	-	-
Dec	0.23	0.12	1.0	0.14	0.08	1.2	1600	84	82	20	20
Jan	-	-	-	-	-	-	-	-	-	-	-
Feb	0.19	0.16	0.80	0.12	0.03	1.0	1200	110	100	19	24
Mar	0.19	0.20	0.72	0.12	0.03	1.0	1400	80	76	19	31

RESTORATION OF SURFACE-MINED LANDS

Land Restoration

Present and future energy needs require the extraction of coal from arid and semi-arid regions of the United States. The disposition of the spoil banks after mining continues to raise questions because the spoil banks disrupt the previous ecosystem and are usually not aesthetically pleasing. The conventional approach has been to level the spoil banks, topsoil, seed, irrigate, and fertilize the new ground to affect biologic productivity at least equivalent to that before mining. An alternative approach is to leave spoil banks instead of leveling them and use the steep slopes of the banks to collect precipitation. The runoff from the slopes irrigates the soil placed in the valley between the banks to enhance the growth of valuable crop plants. This latter approach has the advantage of requiring less earth moving, no irrigation water and potential for increased production of useful crops for long periods of time.

Simulated spoil banks approximately 100 m long, 17 m apart and 3 m high were constructed on the Hanford Site. The width of the top soil surface between the banks is 4 m. There are six valleys, three running NS and perpendicular to the strongest winds, three running EW and parallel to the wind and one area with no adjacent slopes. The two aspects were chosen to test the effect of wind direction and slope aspect on plant production.

The steep slopes forming each valley have been experimentally covered to increase slope stability and runoff. Butyl rubber, 1/16 in. thick, was placed on the slopes of one NS valley in October 1976 to provide a maximum runoff treatment and reference for the other treatments. Paraffin with an approximate melting point of 54°C was sprayed in a melted state with specially designed equipment in May 1977 in another NS valley, and in a EW valley in August 1977. Rubberized asphalt was sprayed in an EW valley in late September 1977.

Instrumentation to measure soil water content, canopy air temperature, soil temperature, wind speed and direction and insolation have been installed to follow the effect of differences in aspect, windspeed and slope treatment on plant productivity.

The first crop of wheat (Nugains, a soft, white winter wheat) was planted in October 1976 in the NS valleys and harvested in July 1977. To exclude deer and rabbits a fence was built. Four varieties of grapes were planted in the NS valleys. The same grape varieties will be planted in the EW valleys in the spring of 1978 after the fencing of that area is complete.

The selected study site is the most arid in Washington. Only 60 mm of rain fell between 1 October 1976, and 1 July 1977. This extreme minimum (normally 150 mm) did not percolate past 30 cm deep in the untreated valleys. In contrast, in the valley with rubber sheet slope treatments soil water percolated beyond the deepest moisture sensors (90 cm). The soil water data are shown in Figure 1.2 where it is clear that only the valley with rubber sheeting had sufficient soil water to sustain plant growth. These values of soil water content are minimums

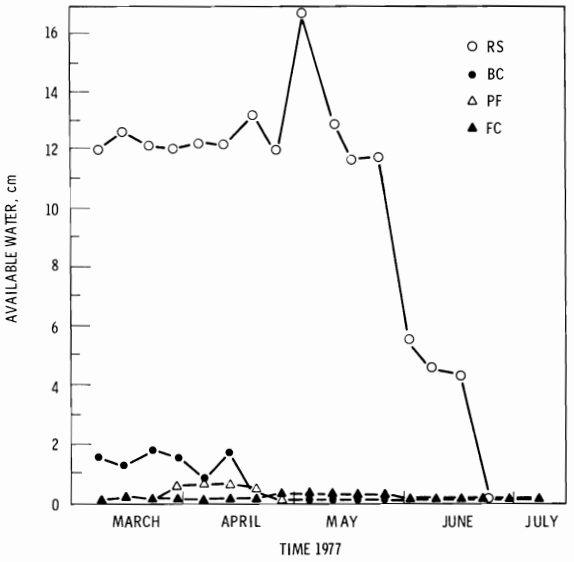

FIGURE 1.2. Centimeters Available Soil Water of Top 10 cm of Soil Profile in the Valley With Rubber Sheeting (RS) and Controls (BC, PF, FC). Available Water is that at Soil Water Potential Greater than -15 Bars. BC is Valley with Bare Slopes. FC is Area with no Adjacent Slopes. PF is Valley Set Aside for Future Parraffin Treatment.

since the total depth of percolation is not known; water probably went much deeper than the 90 cm. It should be noted that the paraffin and rubberized asphalt were not in place for the little rain received during the 76-77 growing season.

Wheat production in the controls was negligible due to the severe drought. Nevertheless, some wheat grain was produced in the valley with rubbersheeting. However, it is believed that the yield would have been greater if the newly planted seeds had not been eaten by a large rodent population. Rodents also harvested the heads of wheat as the crop ripened. To reduce rodent damage, the mice are being live trapped and released sufficiently far away to prevent return.

- **Fossil Fuel Effluents in Freshwater Ecosystems**

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This research is intended to provide predictive information on potential impacts of coal combustion and coal conversion (i.e., solvent refining) on water quality and aquatic biota. Essentially, we are attempting to identify coal byproducts that reach aquatic environments, determine the compounds and elements occurring in greatest concentrations, quantify the behavior and fates of these compounds and elements, assess the potential of coal byproducts for adverse impacts, and define direct and indirect effects that can result on aquatic biota. The present program was evolved from combination of two FY-76 189's entitled: 1) Effects and Behavior of Fossil Fuel Effluents in Freshwater Ecosystems, and 2) Effects of Coal Utilization on Freshwater Biota. Present emphasis is on assessment of potential environmental impacts of whole effluent, effluent components and residual wastes derived from processing of solvent refined coal by a combination of field and laboratory research.

COAL COMBUSTION

Accumulation of Trace Elements in Freshwater Ecosystems

As a final phase of studies on trace metals in freshwater ecosystems at western coal-fired generating plants, data are presented from field samples at Valmont Plant, Colorado and at the Hale and Huntington Plants, Utah.

Table 1.6 summarizes data from three interconnected lakes comprising the cooling system at Valmont and from Baseline Lake, a nearby "control" water body. Sediments from Leggett Lake, which directly receives plant cooling water effluents and ash disposal pond effluents, appear to have higher concentrations of Mn, Cu, Zn, and Pb than sediments from Valmont, Hillcrest, or Baseline Lakes, although Hillcrest Lake had high concentrations of Cu and Zn. Concentrations of Ni and Cu in seston were higher at Leggett Lake than Baseline Lake; the opposite was true for Mn, Fe, and Pb in seston, but Cr and Zn concentrations were similar. Trace element concentrations in bottom organics were similar to those in the sediments. Bottom organic material and cattails were absent from Baseline Lake, thus no comparison of trace element concentrations could be made.

Fish were collected from Leggett and Baseline Lakes, but no species were common to both. Analyses of whole body minus gut contents showed that, in general, most trace elements were more concentrated in fish from Leggett Lake, particularly in May. Comparison

TABLE 1.6. Trace Element Concentrations in Valmont Plant Site Samples, ppm dry wt

	Mn	Cr	Fe	Ni	Cu	Zn	Pb
Core-sediments							
Leggett Lake, May	551-	83	36400	20.8	197	257	167
Leggett Lake, Nov.	578	69	37501	43.6	205	300	216
Valmont Lake, Nov.	322	53	25413	18.7	22.4	92.1	38.7
Hillcrest Lake, Nov.	365	57	32752	31.2	150	217	87.1
Baseline Lake, Nov.	343	67	30427	27.7	25.3	100	47.4
Seston							
Leggett Lake, May	142	28	8000	41	67	92	19
Leggett Lake, Nov.	101	15	2726	26.2	89	71	13
Baseline Lake, Nov.	328	32	17160	16.7	16	104	34
Bottom Organics							
Leggett Lake, May	526	213	30100	43	166	281	86
Leggett Lake, Nov.	750	81	26830	48.5	146	269	106
Cattails							
Leggett Lake, Nov.	517	5.6	127	3.1	3.5	15	2.8
Fish							
Leggett Lake							
Carp, May	83	17	766	3.5	22	1478	13
Carp, Nov.	11	7	281	2.3	3	260	2.1
Bluegill, May	16	14	279	2.6	5.7	155	3.3
Bluegill, Nov.	11	8	143	2.2	6.0	101	2.7
Baseline Lake							
Rainbow, Nov.	3	6	64	1.7	2.0	58	
Black Crappie, Nov.	7	6	100	~1.5	1.2	60	2.2
Bullheads, Nov.	8	6	140	2.6	4.9	62	2.5
Suckers, Nov.	22	5	113	1.9	1.6	45	2.9

of Leggett Lake data shows a tendency for higher values in May than November, especially for carp. Concentrations of all elements in water were below drinking water standards in all lakes. Neither Mo nor Cd was detected in any biological or sediment samples.

Table 1.7 presents trace element data from Cladophora (sessile green alga) and Pteronarcys californica (stonefly nymph) collected above and below the discharge of cooling water from the Hale Plant into the Provo River, Utah and Daphnia from the Huntington Canyon Plant evaporating pond.

TABLE 1.7. Trace Element Concentrations in Utah Site Samples, ppm dry wt

	Mn	Cr	Fe	Ni	Cu	Zn	Pb
Hale Plant, Sept.							
Cladophora, above outfall	7429	34	19720	<15	95	186	54
Cladophora, below outfall	17470	53	18240	<26	213	212	70
Pteronarcys, above outfall	1244	4	1672	6	162	200	10
Pteronarcys, below outfall	1640	20	1767	12	50	186	7.3
Potamogeton	5774	<6	1555	9.6	60	209	11
Huntington Canyon Plant, Sept.							
Daphnia, Evap. pond	358	21	5081	<6.7	344	278	5.0

Cladophora collected below the Hale Plant discharge in the Provo River contained higher concentrations of all trace elements except Fe and Ni, which were essentially the same in populations above and below the discharges. P. californica contained higher concentrations of Mn, Cr, and Ni below the outfall, while Cu was lower and Fe, Zn, and Pb were about the same in both populations. Potamogeton sp., a pondweed growing in the discharge canal at the Hale Plant, contained lower concentrations of Mn, Cr, Fe, Cu, and Pb than did Cladophora. This was probably due to greater adsorptive uptake by Cladophora, which has a much greater surface to volume ratio. Daphnia was the only organism present in the Huntington Canyon Plant evaporating pond. They contained relatively high concentrations of Cu and Zn.

It is apparent that certain trace elements are being concentrated in the aquatic food web at the Valmont Plant. There is, however, no evidence that they are detrimental to the ecosystem as a whole.

Permeability Tests of Carty Reservoir Soils

In preparation for studies of trace metal uptake kinetics in sediments of Carty Reservoir (now being constructed near Boardman, Oregon), a series of permeability tests were performed in the laboratory to aid experimental design. Tests were done using standard permeability test chambers containing intact sediment cores from the three dominate terrestrial communities in the Boardman area: 1) sagebrush, 2) needle-and-thread grass, and 3) cheatgrass. K (velocity) was determined from the equation:

$$K = \frac{Q}{(At)}\left(\frac{l}{h}\right)$$

where:

K = velocity in cm/sec
Q = total discharge in cm^3
A = cross section area of soil column in cm^2
t = total time in seconds
l = length of soil column in cm and
h = hydraulic head in cm.

Three test replicates were performed for each community soil type, each lasting 30 sec. Results are shown in Table 1.8.

The data indicate more rapid water movement through soil columns from needle-and-thread grass areas than through soil columns from cheatgrass and sagebrush areas. Higher permeability rates would be expected in soil from the grass communities since root masses tend to loosen soil and provide channels for water movement. However, permeability rates

TABLE 1.8. Permeability of Carty Reservoir Sediments Taken From Three Terrestrial Community Types

Community	K (cm/sec X 10^{-3} ±SD X 10^{-3})
Sagebrush	2.1 ± 0.2
Needle-and-thread grass	2.8 ± 0.06
Cheatgrass	1.8 ± 0.07

in cheatgrass soil columns were lower than in open, sandy soil columns collected near sagebrush.

Similar tests were run on three cores collected from areas that will be just underwater, at mid-depth, and at the greatest depth of Carty Reservoir. Permeability rates were 5.3 x 10^{-4}, 8.1 x 10^{-5}, and 3.7 x 10^{-3} cm/sec, respectively. The range indicates a wide variety of soil textures and types in the reservoir area. A series of permeability coefficients were determined by Shannon and Wilson, Inc., geological consultants for PGE in various parts of the reservoir site. These were done through the open end of uncased borings, a situation greatly different from ours. Their permeability values ranged from 2 x 10^{-4} to 1 x 10^{-3} cm/sec, and averaged 6.8 x 10^{-4} cm/sec.

Accumulation of ^{54}Mn and ^{109}Cd by Periphyton

Studies were initiated to determine transfer coefficients between the trace metals ^{54}Mn and ^{109}Cd and periphyton with methods described by Cushing and Rose (1970. Limnol. Oceanogr. 15:762-767). For ^{54}Mn, uptake and retention experiments were performed with both living periphyton under constant light and killed periphyton under constant light. A single uptake and retention experiment was done with ^{109}Cd using living periphyton under constant light.

Quasi-equilibrium conditions were reached in both cases for ^{54}Mn after 160-190 hr exposure. Approximately 40% more ^{54}Mn was accumulated by the living periphyton community than by killed periphyton. This figure was higher than that found previously for accumulation of ^{65}Zn, and indicates that adsorption was an important mode of uptake. Retention experiments indicated that ^{54}Mn was more tenaciously bound to periphyton than ^{65}Zn. On the average, less than 8% of the accumulated ^{54}Mn was lost after about 200 hr of washing with unspiked water.

Quasi-equilibrium conditions for ^{109}Cd were not attained until about 230 hr, considerably longer than required for ^{54}Mn or ^{65}Zn. Circulation of unspiked water resulted in loss of only 7% of the accumulated ^{109}Cd after 110 hr.

COAL CONVERSION

This phase deals with a developing technology, the solvent refining of coal. Studies were initiated in mid-1976 at a pilot solvent refined coal (SRC) plant operated under ERDA (DOE) contract by the Pittsburg and Midway Coal Mining Company (PAMCO) at Fort Lewis, Washington.

Solvent refining involves mixing of crushed coal with a recyclable coal-derived solvent, and heating to about 825°F in the presence of hydrogen at 10,000 psi or greater. The coal swells and dissolves. Refined coal is obtained after filtration to remove inorganic residue, and evaporation to remove the solvent. The final product has minimal residual ash and sulphur content, and provides optimum quality air emissions when burned. The effluents from the pilot plant are not necessarily representative of effluents from a later demonstration or commercial plant.

New techniques for solvent refining of coal are being developed under the national program to accelerate production and use of coal for energy. Initial research and development for the SRC-I process at the PAMCO pilot plant was complete by February 1977. The plant was then converted to SRC-II process during a 4 month closure and resumed operation in summer 1977. Modification of the refining process is not expected to greatly alter the quantitative composition of liquid effluents passed from the plant. Our research objectives are to:

- Characterize the organic and inorganic constituents in liquid SRC effluent discharged from the pilot plant.
- Determine acute and chronic toxicity of treated SRC effluent, and of key effluent components.
- Measure the accumulation and retention of key SRC effluent components in selected freshwater plants and animals.
- Assess the sublethal physiological and behavioral responses of freshwater biota to key compounds occurring in treated SRC effluent.

Solvent Refined Coal Characterization Studies

Toxicity Tests With SRC Effluent

This phase is essentially biotic monitoring intended to quantify obviously adverse

effects that are directly lethal to aquatic organisms exposed to whole or diluted portions of SRC plant effluent. It is designed to correlate, insofar as possible, biological response with qualitative chemical analyses of effluent samples.

Fish are valuable indicators of toxic components in industrial waste discharges since they exhibit a variety of direct (lethal) and indirect (sublethal) responses. However, a biological response alone will not identify the particular toxicant or combination of toxicants evoking the response, but merely indicate that deleterious material is present.

We conducted 4-day laboratory exposures with SRC effluent drawn from the pipe discharging treated liquid wastes into a small sump. Juvenile rainbow trout were the main test organisms. The test design involved 6 pairs of aquaria containing 0, 20, 40, 60, 80 and 100% effluent and positioned randomly. The tests were static to avoid further dilution and unaerated to minimize loss of volatiles. Summary of results to date is given in Table 1.9.

Exposure mortalities were sporadic, and often attributed to low dissolved oxygen levels or outbreaks of a bacterial pathogen. The effluent appeared to have a narcotic effect in some tests, causing sluggishness and respiratory distress among fish. At this point, only general conclusions can be advanced concerning the extent of toxicity of SRC effluent to aquatic life. First, the toxic component, when present, was apparently a volatile organic compound, possibly derived from the process solvent as well as the dissolved coal. Second, the in-plant treatment (neutralization, ionization, biodigestion) greatly reduces the amount of toxic components in the effluent. Third, toxicity of effluent varies widely from one sampling period to the next.

Apparently many factors affect the quality of SRC effluent, including: 1) condition of charcoal filters in the plant, 2) acid ratio and vitality of bacteria in the biodigesters, 3) the point in a test run when an effluent sample is obtained, and 4) the extent of sorbing of organic compounds to particulate matter in the effluent.

SRC Effluent Characterization

Samples were collected for qualitative and quantitative analyses at the time effluent was collected for bioassays. These samples were usually taken at three locations in the SRC extraction process: 1) untreated effluent leaving the surge reservoir, 2) partially treated effluent leaving the ion

TABLE 1.9. Summary of Procedures and Assessment of Results from 96-Hr Static Laboratory Bioassays with Treated SRC Process Effluent at 17-19°C. Effluent was collected at SRC plant one day before start of each assay.

Date Assay Conducted	Test Organisms	Test Conditions	Assessment
Aug. 3-7, 1976	Rainbow trout fingerlings	nonaerated	No relationship between mortality and effluent concentration. Losses due to low dissolved oxygen.
Aug. 24-28, 1976	Largemouth bass fingerlings	aerated	No mortality attributable to effluent concentration. Some losses due to disease.
Oct. 12-16, 1976	Rainbow trout fingerlings	aerated	Significant mortality (>50%) at effluent concentrations of 80 and 100%, associated with effluent chlorinations.
Nov. 16-20, 1976	Rainbow trout and crayfish	aerated	Significant mortality (>50%) at effluent concentrations of 40, 60, 80 and 100% for rainbows, >60% for crayfish; narcosis evident among crayfish.
Jan. 27-31, 1977	Rainbow trout fry	nonaerated	No mortality in 96 hr but some fish stressed at 80 and 100% effluent concentrations.
Jun. 21-25, 1977	Rainbow trout fingerlings	nonaerated	Limited mortality at higher effluent concentrations.

exchange columns, and 3) treated effluent leaving the biodigestor and oil filters and entering the discharge sump. Inorganic components were quantified by x-ray fluorescence. Organic components were quantified, insofar as possible, by a combination of nitrogen sparging, gas chromatography, liquid chromatography, and other techniques. Many samples in FY-77 proved unsuitable for organic analysis.

Concentrations of 21 elements were measured by x-ray fluorescence in the SRC effluent (As, Br, Ca, Cl, Co, Cr, Cu, Fe, Ga, Hg, K, Mn, Ni, P, Pb, Rb, Se, Si, Ti, V and Zn). At the point of discharge into the effluent pond, none of the identified elements differed greatly in concentration from levels occurring in plant tap water, or in water from nearby Lake Sequalichew. Nor was there evidence of elemental accumulation in water from the SRC effluent pond. In-plant treatment of effluent and dilution with freshwater before discharge may account for low element levels.

Organic analyses to date are less than satisfactory due to difficulty in obtaining reliable qualitative and quantitative determinations with each sample series to correlate with the bioassays. The 36 organics identified were highly aromatic hydrocarbon and, in general, were similar to those in petroleum contaminated samples. The major differences were lower amounts of alkylate compounds relative to the parent aromatics and the presence of phenylether and dibenzofuran (Tables 1.10 and 1.11).

TABLE 1.10. Characterization of Total Extractables from SRC Process Effluent Samples Taken 5/21/76

Sample Location	Sample Volume, ml	Extraction Solvent	Amount of Extractables, mg/l Gravimetric[a]	Infrared[b]	Hydrocarbon Index[c]
V-9105	3960	CH_2Cl_2	19.30	—	—
V-9105	3920	CCl_4	29.90	12.10	0.09
V-9105	4075	CCl_4	21.30	8.00	0.07
V-9108	3770	CCl_4	16.10	5.60	0.15
V-9108	3735	CCl_4	17.60	5.90	0.16
Treated Effluent	3788	CH_2Cl_2	0.50	—	—
Treated Effluent	3755	CH_2Cl_2	0.19	—	—
Treated Effluent	3660	CCl_4	0.75	0.05	0.24
Treated Effluent	3965	CCl_4	0.58	0.06	0.47

[a] Determined by weighing the solvent-free residue from an aliquot
[b] Determined from CH_2 absorption using API Reference Fuel Oil for calibration
[c] (O.D. of C=0) ÷ (O.D. of C-H_2). This ratio is used as an indication of the proportion of hydrocarbons. A ratio of 0.2 or less indicates that hydrocarbons probably represent a majority of the CCl_4-extractable material.

TABLE 1.11. Characterization of Aromatic Components in Water Extracts from SRC Process Effluent Samples Taken 5/21/76

No.	Retention Index	M.W.[a]	Tentative Identification[b]	Amount Found in Given Sample, µg/l[d] V-9105	V-9108	V-9108	Treated Effluent
1	1080	120	Dimethylethylbenzene	54	10	2	[e]
2	1140	132	Methylindan	28	16	21	[e]
3	1200	132	Tetralin	130	37	68	[e]
4	1220	128	Naphthalene	940	510	820	2
5	1220	134	Benzothiophene	60	30	70	[e]
6	1250	146	Methyltetralin	100	60	22	[e]
7	1260	146	Methyltetralin	49	27	10	[e]
8	1280	146	Methyltetralin	27	4	8	[e]
9	1300	146	Methyltetralin	160	34	56	0.5
10	1320	142	2-Methylnaphthalene	800	330	510	2
11	1330	142	1-Methylnaphthalene	200	160	120	0.5
12	1330	148	Methylbenzothiophene[c]	70	30	70	[e]
13	1350	160	Dimethyltetralin	30	11	6	[e]
14	1390	154	Biphenyl	340	170	260	0.5
15	1410	156	Ethylnaphthalene	210	64	62	0.5
16	1420	170	Phenyl Ether	390	240	260	4
17	1420	156	Dimethylnaphthalene	130	35	32	0.5
18	1430	156	Dimethylnaphthalene	90	23	17	[e]
19	1450	156	Dimethylnaphthalene	33	7	8	[e]
20	1480	154	Acenaphthene	62	35	55	0.5
21	1500	168	Methylbiphenyl	170	47	44	1
22	1510	168	Dibenzofuran	78	25	27	[e]
23	1570	166	Fluorene	94	27	23	3
24	1580	168	Methylacenaphthene	59	15	8	2
25	1590	184	Dibenzothiophene[c]	25	8	8	[e]
26	1620	182	Dimethylbiphenyl	34	3	2	[e]
27	1630	182	Dimethylbiphenyl	28	1	1	[e]
28	1660	180	Methylfluorene	2	2	5	[e]
29	1680	180	Methylfluorene	2	1	2	[e]
30	1690	180	Methylfluorene	4	2	3	[e]
31	1740	182	Dimethylacenaphthene	57	15	9	0.5
32	1760	178	Phenanthrene	280	59	96	7
33	1880	192	Methylphenanthrene	100	7	3	2
34	1900	192	Methylphenanthrene	37	4	2	1
35	2060	202	Fluoranthene	76	4	6	0.5
36	2100	202	Pyrene	38	3	3	3

[a] Determined by chemical ionization mass spectrometry using methane as the ionizing gas
[b] Tentative identifications are based on the molecular weight, lack of fragmentation indicated by the mass spectra, and gas chromatographic retention times. In some cases other isomers are equally likely, e.g., dimethylindan instead of methyltetralin or ethylbiphenyl instead of dimethylbiphenyl
[c] Sulfur-containing compounds were confirmed by flame photometric detection
[d] The amounts are reported as µg/liter of original water sample extracted
[e] Not detected, < 0.5 µg/l

Phenol and phenolic compounds (o-creosol, m-creosol, p-creosol) are present in both hydrocarbon and aqueous streams of the SRC process. The combined phenol content in the plant wastewater can reach the 1,000 ppm level before biodegradation and biodigestion treatments. Continued quantitative determinations are being made in cooperation with PNL's Physical Sciences Department (Table 1.12).

Behavioral Studies With Chemical Components

Predator-Prey Study

The main chemical families identified in SRC process waters have been the polycyclic hydrocarbons, phenolics, and quinolines. The SRC process waters were shown to contain several percent organic compounds with phenolic species predominating.

Based on this and the relatively large volume of phenol toxicological information

TABLE 1.12. Analysis of Phenol in Treated SRC Process Effluent at Various Dates, Determined by High Pressure Liquid Chromatography. No cresols or xylenols were detected.

Sample Date	Source/Treatment	Phenol, ppb	Remarks
10/11/76	Plant discharge pipe	6.6 ± 0.6[a]	
10/11/76	Plant discharge pipe	5.2 ± 0.5	2 days later at start of bioassay after 4 days aeration during bioassay
10/13/76	Plant discharge pipe	8.2 ± 0.7	
1/26/77	Plant discharge pipe	6.6 ± 0.6	
4/12/77	Plant discharge pipe	39.0 ± 4.0	
4/12/77	East end discharge pond	7.9 ± 0.7	
4/12/77	West end discharge pond	13.1 ± 1.2	

[a]Samples on 10/11/77 consisted of treated effluent at undiluted strength, taken during backflush operations.

available, phenol was selected as our initial toxicant for investigation. Although conventional bioassay techniques have been used to establish lethal doses of phenol at 10-25 mg/l for trout, evaluation of subtle sublethal effects that influence complex species interaction have not been attempted. Behavioral methods such as predator-prey interactions are more sensitive than conventional bioassay techniques, allowing detection of environmental stress at much lower levels than those resulting in direct mortality.

We are using a predator-prey interaction to determine the effective concentration (EC_{50}) of phenol that influences predator-prey relationships. The design of our experiments requires exposure of juvenile rainbow trout (Salmo gairdneri) to sublethal concentrations of phenol and control trout to river water for a standard period of 96 hr prior to presenting mixed groups of fish to large predators, also rainbow trout. The prepredation exposure concentration was selected on the basis of the reported lethal concentration range for phenol (10-25 mg/l) and on a reported threshold for disturbance in trout of 1.3 mg/l. The test fish were exposed to a range of phenol concentrations (2.5, 1.88, 1.41, 1.05 and 0.79 mg/l) in aquaria supplied by a Mount proportional diluter.

Figure 1.3 is a flow chart showing design of the predator-prey experiment. Juvenile trout are marked with a water insoluble dye for later identification as phenol-exposed or control fish. After exposure to phenol, 25 fish from the two groups are combined and simultaneously presented to predators (adult trout) in a large circular tank. When about 25 fish remain, the survivors are removed and scored.

Results thus far show no significant difference in the rate of predation on exposed and control fish. Considerable difficulty has been encountered in controlling the delivered concentration of phenol from the diluter. The variable concentrations may have masked any effect. However, it is believed that the range of phenol concentrations may be below the effect threshold. A new series of experiments has been initiated

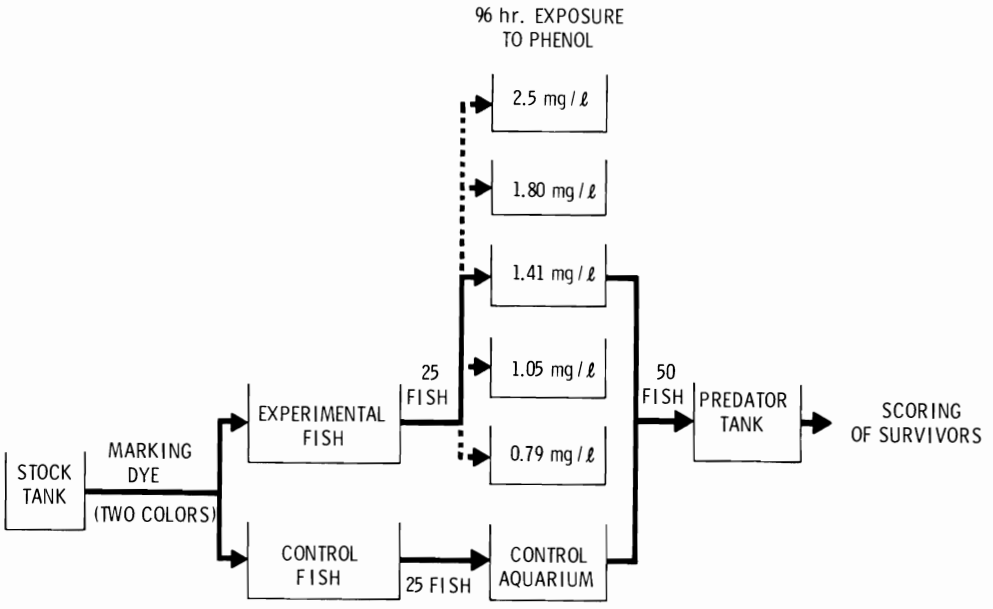


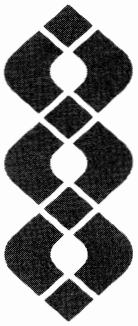
FIGURE 1.3. Flow Chart of Predator-Prey Experiment

with elevated levels of phenol (7.00, 5.25, 3.94, 2.95 and 2.20 mg/l) for prepredation exposure.

Avoidance - Attraction Study

The ability of fish to detect a toxicant can be determined by avoidance of or attraction to sublethal concentrations. Tests with phenol will be conducted in a trough supplied with an equal amount of flowing water entering from both ends. The water exits from the trough through a bottom slit in the middle. The flow characteristics of the trough produce an interface above the slit. If a toxicant enters at one end, fish in the trough are presented with a choice of being attracted to or avoiding the toxicant.

In order to avoid "investigator influence" of the results, tests will be recorded on video tape from which fish distribution can be scored. Only initial tests have been conducted at this time. No behavioral response, avoidance or attraction, was observed on exposure of fish to 1.0 ppm phenol.



2.0 Nuclear Wastes: Fission

NUCLEAR WASTES: FISSION

- **Suspended Particle Interaction and Uptake in Terrestrial Plants**
- **Transuranic Complexation in Soil and Uptake by Plants**
- **Quantitative Aspects of Plutonium Field Studies**
- **Ecological Distribution and Fate of Plutonium and Americium in a Processing Waste Pond on the Hanford Reservation**
- **Use of Analog Elements to Predict the Equilibrium and Behavior of Transuranic Elements in the Environment**
- **Weathering and Aging of Transuranics**
- **Radioecology of Uranium**
- **Radioecology of Waste Management Areas (ROWMA)**
- **Radioecology of ^{129}I and ^{99}Tc**
- **Influence of Soils and Aquatic Sediments on the Chemical Behavior, Transport, and Bioavailability of Pollutants**

One of the highest priorities associated with the continued development and use of fission as a source of energy is the resolution of questions about the long-term fate and ultimate effects of long-lived radionuclides in the biosphere. This long-term behavior generally cannot be inferred with confidence from the short-lived isotopes. The individual studies described in this section range from basic investigations of the behavior and effects of long-lived nuclides at the chemical and cellular level to ecosystems studies which describe inventory, transport, and effects in waste management areas. The investigations and reports listed in this section are designed to differentiate and measure certain processes affecting the biological availability of the radioelements. The specific applications of this and other information to nuclear fuel reprocessing and waste management sites will be found in Section 8 with appropriate publications listed there. Materials of particular concern in connection with Nuclear Fusion, e.g. ^{3}H, are covered in Section 3.

In terrestrial studies, investigations of transuranics in soils and plants have demonstrated the importance of valence state, complexation, competing elements, microbial processes, migration down the soil profile, and weathering cycles in governing transuranic, ^{129}I and ^{99}Tc availability to plants and, in the case of Pu, to the consuming animals. In the latter case, it was demonstrated, for the first time, that ingestion of plant tissues containing Pu may result in greater transfer across the gut compared to gavaging animals with inorganic Pu solutions, underscoring the importance of detailed studies of the soil, plant and animal factors influencing uptake by the ingestion pathway. Further evidence of the importance of the ingestion pathway was provided in studies of foliar interception of airborne transuranic elements in which it was shown that Pu in particles in the respiratory size range were effectively intercepted and retained by plants, and significant quantities of intercepted Pu were transported to roots and seeds. Similar studies on the terrestrial ingestion pathway have been initiated with other actinides including U, Am, Cm, and Np.

Radioecological field studies at PNL were directed toward establishment of pertinent ingestion pathways and exposure levels through description of habitat types, population densities, and, in several instances, dosimetry, for major insects, reptiles, birds, and mammalian species. These studies were extended to agricultural ecosystems through definition of the uptake of long-lived nuclides and digestibility in cattle of several forage species. In studies on a pond ecosystem at the nuclear fuel reprocessing plant, Pu and Am uptake rates were studied for major biotic components including organic floc, algae, fish and ducks. The results indicated that assimilation of transuranics by the biota and export from the pond system were low compared to the total inventory.

Field inventory studies have provided the bases for appropriate environmental sampling and, in conjunction with basic studies of Pu behavior in soils and plants, have resulted in the development of reliable statistical procedures for estimating the concentrations of transuranics in soils. Decisions as to need for land decontamination in former nuclear detonation and waste management areas will be based on this inventory.

The approaches developed in terrestrial and freshwater studies were recently extended to studies of the marine environment. The marine environment may represent an ultimate repository for the mobile forms of transuranic elements and results indicate that geochemical and biochemical factors may influence transuranic mobility and availability to biota. Investigations have provided a basis for description of dispersion mechanisms on the sea floor of transuranics arising from nuclear detonations and a bomber crash. In addition to physical dispersal, biochemical factors may influence transuranic speciation and mobilization resulting in substantial changes in Pu availability as a function of ocean depth.

This is a comprehensive program, encompassing basic, as well as descriptive studies on the behavior and effects of the major long-lived radionuclides. It should provide a better integrated picture of the extent of engineering sophistication required in the nuclear fuel cycle to minimize risks associated with this important energy source.

- **Suspended Particle Interaction and Uptake in Terrestrial Plants**

  Principal Investigator: D. A. Cataldo

The purpose of these studies is to develop an understanding of the behavior and fate of airborne pollutants following interception by plant foliage. Submicron (<1 μm) particles intercepted by plant canopies behave differently, with respect to retention time on foliage, than >10 μm particles. The present studies with $^{241}AmO_2$ show submicronic particles (respiratory size range) to be effectively intercepted and retained on plant foliage. Even under rigorous leaching conditions, >70% of the deposited Am is retained. The data also suggest that retention tends to increase with residence time on foliage. Transport studies show a significant fraction of the foliar deposits to be transported to roots and seed. The quantity transported to these tissues tends to increase following leaching treatments (simulated rainfall).

Investigations are continuing in an effort to understand the behavior of particulate forms of the transuranics in the environment. Areas of study include: 1) an evaluation of the retention characteristics of ^{241}Am, ^{244}Cm and ^{237}Np in addition to ^{238}Pu; 2) the extent of uptake and translocation of the transuranics following foliar contamination; and 3) elucidation of the relationships between particle size and mechanisms associated with their retention and bioavailablity.

Retention of Submicronic Transuranic Particles by Plant Foliage

Previous studies show the retention behavior of submicronic Pu particles intercepted by plant foliage to differ substantially from early concepts of retention half-times for natural and simulated fallout particles having diameters >10 μm. Submicronic particles of ^{238}Pu appear to remain fixed to leaf surfaces of bushbean with <2-8% able to be resuspended by the action of rainfall over a 21-day period following contamination. These studies have been extended to include ^{241}Am.

Foliage of 21-day old bushbean plants were contaminated with freshly prepared $^{241}AmO_2$ using a low-wind speed plant exposure facility. Particles deposited onto foliage have a count median diameter of 0.10 - 0.20 μm.

To study the retention behavior of $^{241}AmO_2$, contaminated foliage was subjected to a simulated rainfall (0.4 cm/10 μm) 1, 7, 14 or 21 days following contamination (Figure 2.1). The retention date obtained for $^{241}AmO_2$ was qualitatively similar to that obtained for $^{238}PuO_2$, while being quantitatively more available for leaching under mild rainfall conditions at 7, 14 and 21 days. However, only 3 to 8% of the foliar deposits could be removed under these mild leaching conditions. Application of more rigorous leaching conditions (1.6 cm/30 min) with either simulated rainwater or acidified rainwater (pH 2.0) using plants with all but contaminated foliage removed, shows an interesting trend. Rainwater, pH 7.0, effectively removes ∿8% of the total foliar deposits, while pH 2.0 rainwater removed ∿27% of the deposited $^{241}AmO_2$. These values are substantially higher than the 4 and 6% removable by rainwater and acidified rainwater, respectively,

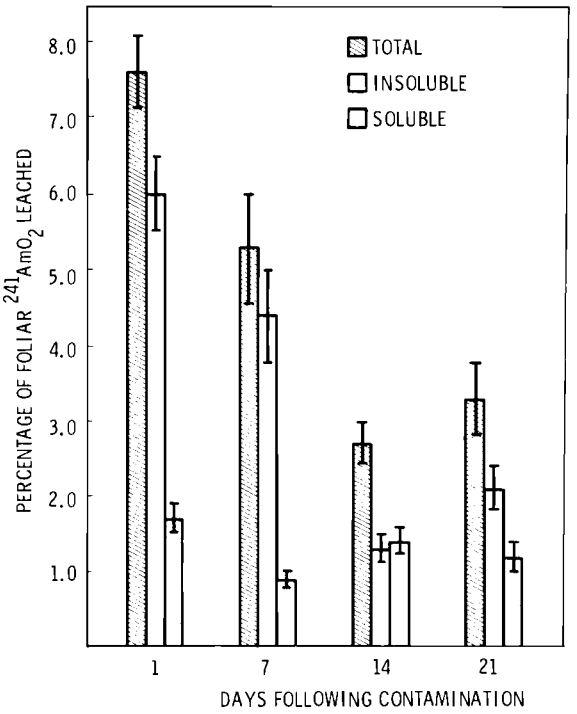


FIGURE 2.1. Leachability of foliar $^{241}AmO_2$ Contaminants as a Function of Time Following Contamination of Bushbean Leaves.

seen for fully prepared $^{238}PuO_2$ suspensions. Fractionation of the leachates resulting from application of acidified rainwater shows 95% of the leachable Am to soluble (passes a 0.01 μ membrane filter).

Data obtained for submicronic $^{241}AmO_2$ particles, and previous data on $^{238}PuO_2$, suggest that retention of submicronic particles by plant foliage is greatly underestimated in light of currently accepted retention half-times of 14 days. This implies the need for revised hazard analysis and further study of chemical modification and foliar absorption that may occur because of extended residence times for submicronic particles on foliage.

Uptake and Translocation of Foliarly Deposited Americium

Earlier studies, with various chemical forms of ^{238}Pu, showed that the availability of Pu deposited on foliar surface for uptake and transfer to bushbean seeds was dependent on chemical form of Pu supplied and increased with the application of a simulated rainfall to contaminated foliage (solution vector). Concentration ratios (CR) using freshly prepared $^{238}PuO_2$, for root and seed tissues were $\sim 5 \times 10^{-6}$ in the absence of rainfall and increased to $\sim 2 \times 10^{-5}$ on application of a simulated rainfall. Similar studies have recently been completed for $^{241}AmO_2$.

Calculation of classical CR values for transfer from contaminated foliage to seed or root tissues yields values of 3×10^{-7} to 8×10^{-6}. Unlike the behavior of Pu, transfer rates were not increased by the application of a simulated rainfall. The use of CR values for evaluating the transfer of transuranic from foliage to seed or root, although useful in making comparisons, should be used with caution. Substantial variation in CR value appears to result from differences in the magnitude of the concentration of transuranic on contaminated foliage. Although data generated for Pu are consistent between experiments, experiments with Am, due to the apparent larger soluble component, appear to have fractionated during aerosol generation and interception, yielding an order of magnitude difference in foliar concentration. However, if one looks at simply the percentage transfer to seed or roots, or the total Am present/g seed, there appears to be 5 to 20 times more Am absorbed and transferred to seed and roots than found for ^{238}Pu (freshly prepared).

Future efforts will be directed towards evaluating the processes controlling foliar absorption and transport of transuranic in plants. Results with both Pu and Am indicate that foliar absorption is a complex process, with chemical form and solubility playing a major role. Studies are being initiated to investigate the absorption of transuranics through foliar surface and its fate following absorption.

• Transuranic Complexation in Soil and Uptake by Plants

Principal Investigators: R. E. Wildung, T. R. Garland,
K. M. McFadden, D. A. Cataldo, H. Drucker and M. Sullivan

The principal objective of these studies is to define the role of soil microbial and chemical processes in influencing the long-term availability, distribution, and form of transuranic elements in plants and animals.

In order to accomplish this objective, basic studies have been undertaken to determine: 1) the effects of transuranic elements on soil microbial populations and processes, 2) the potential for alteration of valence state and formation of transuranic-organic complexes in soil as a function of soil physicochemical properties and soil microbial activity, 3) the influence of soil microbial and plant processes on the kinetics, extent of uptake, sites of deposition, and form of transuranic elements in plants, and 4) the gut transport and form of transuranic elements in animals as related to soil and plant variables.

Emphasis was placed on delineation of the relationships between soil chemical and microbial processes and the role of soil microorganisms in affecting solubilization and transformation of elements considered largely insoluble in soils strictly on the basis of their inorganic chemical characteristics.

Soluble, diffusible Pu in soils (usually less than 0.1% of total) appears to be largely present as particulates of hydrated oxide, but several lines of evidence suggest that microorganisms may influence the solubility of Pu and that the nonparticulate plant-available fraction is stabilized in solution by inorganic or organic ligands of limited concentration in soil. The role of soil microorganisms in influencing the solubility, form and plant-availability of the transuranics is discussed on the basis of the: 1) known chemistry of organic ligands in soils, 2) effects on the soil microflora, and 3) principal microbial transformation mechanisms, including direct alteration (valence state, alkylation), indirect alteration (metabolite interactions, influence on the physicochemical environment), and cycling processes (biological uptake and release after decomposition of tissues).

The toxicity of Pu to microorganisms depends on Pu solubility in soil. However, soil microorganisms are generally resistant to Pu, with toxicity apparently due to radiation rather than chemical effects. Highly resistant bacteria, fungi, and actinomycetes have been isolated from soil, and these organisms have been shown to be capable of transporting Pu into the cell and altering its form in the cell and in solution. The resulting soluble Pu complexes tend to be of higher molecular weight than simple complexes (Pu-DTPA) and negatively charged. The form of Pu, although not well-defined, is dependent upon organism type, carbon source, and time of Pu exposure during growth. These factors, in turn, are a function of Pu source, soil properties, and soil environmental conditions. Knowledge of the relative influence of these factors serves as a valuable basis for predicting the behavior of Pu and other transuranic elements in the soil.

During the current year, investigations have continued to develop basic techniques for understanding the chemistry of the transuranic elements and to measure 1) the solubility and plant availability of Pu as a function of soil properties, 2) chemical speciation (complexes, valences) of Pu in soil extracts, microbial cultures, and plants, 3) microbial transformations of Pu in soil, 4) form and behavior of Pu in plants and 5) availability to animals of Pu in plant tissues. In addition, a literature review of organic complexants in nuclear wastes was completed with partial support from the Environmental Protection Agency. The possibility of mobilization of wastes by complexation will be assessed using basic information derived from the present program.

Pu Valence States

Pu valence states have been determined on a regular basis using a thenyltrifluoroacetone (TTA) extraction discussed in a previous report. The use of complexing agents for measurement of oxidation state was extended to separation by TLC using cellulose plates pretreated with TTA or triisooctylamine (TIOA) and developed in 2M NH_4NO_3-.5 M HNO_3. Pu IV and VI were mobilized differently in these systems, though the separation was not complete. Optimum concentration of ligand required has not been determined. The possibility of diffusion on TIOA plates during standing and of photochemical sensitivity of the TTA plates must be resolved.

The TTA extraction method was used in the presence of several model Pu complexes to determine the effect of complexing agents on the reduction of PuO_2^+ to Pu IV. Ligands were chosen because of their production by plants or microorganisms (gentisic, malic, and photocatechnic acids), and their use in agriculture or industry (DTPA, EDTA). Formation of the corresponding chelates with Pu was demonstrated by TLC. Complexed forms behaved identically on TLC, whether the Pu was supplied initially as Pu IV or as PuO_2^+. Solubility (<.01 μ) of Pu chelates, as listed in Table 2.1, was measured 1 and 12 days after preparation. Aliquots of each filtered solution were preserved in 1.8 M HNO_3 for 24 hr prior to TTA extraction. Initial extraction represents the aliquots filtered 1 day after preparation and acidified 10 days later. Final extraction represents aliquots filtered 12 days after preparation and immediately acidified.

Since benzene alone did not result in extraction of Pu from any of the complex systems, TTA likely results in disassociation of the complex and extraction of Pu IV. Much less likely is the possibility of TTA extraction of the intact complexes. Where Pu IV was initially present, the majority of the soluble Pu remained as extractable Pu IV for most ligands in Table 2.1. In 0.02 M $CaCl_2$, the small fraction of soluble Pu is probably hydrolyzed and therefore unextractable. The marked drop in extractable Pu in the final extraction of gentisic acid suggests the removal of some stabilizing influence, thereby allowing Pu to hydrolyze. A sizeable fraction of Pu was left at the origin in TLC plates of the gentisic acid system, supporting this conclusion. The protocatechnic acid system similarly shows Pu at the origin, and incomplete extraction by TTA.

When PuO_2^+ was the initial Pu form, reduction to Pu IV by ligand action, was evidenced by TLC and by the extraction data. The Pu was extractable from protocatechnic acid, DTPA and EDTA. Since TLC shows all Pu complexed by DTPA and EDTA, evidently the TTA did not completely disassociate the complexes. This is also observed to some degree in the Pu IV system. Gentisic acid, again, showed low final extractability and considerable Pu remaining at the TLC origin. In the system used, the Rf of the Pu-malic acid complex was indistinguishable from that of uncomplexed PuO_2^+, so the efficiency of complex formation cannot be determined. The Pu was extractable from the Pu IV-malic acid system, suggesting that malic acid did not reduce PuO_2^+ to the complexable Pu IV.

TABLE 2.1. Reduction of Pu in the Presence of Organic Liquids

	Pu IV Added				PuO_2^+ Added			
	Soluble		TTA Extractable		Soluble		TTA Extractable	
	Initial	Final	Initial	Final	Initial	Final	Initial	Final
	- - - % - - -							
0.02 M $CaCl_2$	16	25	4	0.5	78	74	0.7	0.2
DTPA	100	78	79	66	89	75	49	48
EDTA	96	81	80	77	77	65	59	59
Gentisic Acid	61	64	59	2	82	80	70	3
Malic Acid	85	82	76	75	86	83	19	16
Protocatechnic Acid	79	77	74	45	83	83	73	74

With some ligands, use of PuO_2^+ apparently aids in complex formation by providing a source which remains more soluble relative to Pu IV and, therefore, more available for complexation, provided the ligand can reduce the species. A comprehensive study using ligands with a broader range of properties is scheduled.

Behavior of Pu in Soil

Studies to define the kinetics of Pu solubility and diffusibility in 28 well-characterized soils representing a range in physicochemical properties have continued. Solubility parameters will be related statistically to soil properties in order to establish a basis for prediction of Pu solubility and plant availability over a broad geographic region. This information, in conjunction with studies of transuranic speciation, microbial transformations, and plant and animal availability of Pu in different soils, should provide a broad predictive base to assess the long-term behavior and effects of Pu in the terrestrial environment.

Further characterization of 28 soils representing a wide range of physicochemical properties has continued. Elemental analysis of major components in the soils was accomplished using standard fusion techniques. Analyses for mercury by atomic absorption revealed a range of <0.02 - 0.45 ppm. Trace elemental analysis by spark source mass spectrometry for elements ranging from Li to U has been completed. In addition, unifoliates from 25-day old soybeans grown on these soils were analyzed by spark source mass spectrometry for trace elemental composition. A range of several orders of magnitude has been observed in the apparent uptake concentration ratios of several elements from the 28 soils. These data will serve as a comparative control data set for a later study of Pu availability to plants from the 28 soils.

In addition to total elemental composition, studies of extractable forms of several elements have been completed. Adsorption by, or reaction with, hydrous oxides of Fe, Al, and Mn is considered to be a major mechanism for sorption of heavy metals in soils. Pyrophosphate, dithionite, and oxalate extractions approximately reflect the organic, inorganic, and amorphous oxide forms, respectively, of Fe and Al. Available Mn oxides were determined by a $NH_2OH \cdot HCl$ reduction technique, while a KOH extraction was used to estimate SiO_2 and Al_2O_3 forms. Range, average, and mean values for these materials in the 28 soils are listed in Table 2.2.

TABLE 2.2. Total and Extractable Elemental Content of 28 Soils

	Range	Average	Mean
		- - - % - - -	
Na - Total	0.085 - 5.9	1.3	1.2
K - Total	0.24 - 1.8	1.3	1.4
Ca - Total	0.22 - 4.8	1.6	1.5
Mg - Total	0.067 - 1.4	0.72	0.62
Ti - Total	0.055 - 0.97	0.48	0.46
Si - Total	4.2 - 37.3	28	28
KOH (SiO_2)	0.075 - 1.0	0.34	0.24
Mn - Total	0.004 - 0.32	0.11	0.087
Amor. Oxides	0.003 - 0.52	0.087	0.048
Al - Total	0.78 - 8.4	5.6	6.1
Extractable:			
Pyrophosphate	0.005 - 1.2	0.13	0.07
Dithionite	0.002 - 0.39	0.052	0.021
Oxalate	0.011 - 1.4	0.20	0.12
KOH(Al_2O_3)	0.008 - 0.26	0.070	0.036
Fe - Total	0.38 - 6.9	3.4	3.5
Extractable:			
Pyrophosphate	0.001 - 1.2	0.14	0.05
Dithionite	0.039 - 0.8	0.18	0.14
Oxalate	0.022 - 1.1	0.25	0.16
P, Available	0.0004 - 0.018	0.004	0.003

In the oxalate extraction for Fe and Al, soils were contacted with the solution for 20 min. Upon reextraction, several soils showed the expected decrease in Al concentration in the second leachate, but a considerable increase in Fe. All soils having this phenomenon were native to the Hanford area and contained considerable magnitite. Oxalate is known to dissolve magnitite and easily weatherable iron minerals. When extractable Fe and Al were monitored for 48 hr, these soils showed a marked increase in extractable Fe, and a less pronounced increase in extractable Al. Some soils showed an initial apparent equilibrium at $\leq$ 2 hr, which may represent the maximum extraction time allowable before mineral dissolution becomes significant.

Behavior of Pu in Plants

Investigations have been concerned with evaluating the physiological processes which affect and possibly regulate the behavior of Pu in plants. These have dealt with the kinetics of Pu accumulation by plants from soils, characterization of Pu form in xylem exudate, and the importance and fate of Pu form on movement from leaves to seed.

Previous studies with six soils varying in properties, showed Pu content of shoot and root tissues to increase in a linear function with plant age. This suggested that the Pu content of plants is a function of plant growth in addition to the concentration of "available" Pu in soil solution, the latter being a function of both soil parameters and chemical speciation of Pu. An analysis of the kinetics of dry matter production and Pu content of shoots of soybean grown on Ritzville soil, and corresponding CR values from germination to maturity shows several trends (Figure 2.2). First, growth (dry matter production) increases exponentially over the 84 day period, with maximum dry matter production occurring after initial flowering; this is characteristic of plants with indeterminant stem meristems. Interestingly, the total Pu content of shoots exhibits similar kinetics. This would suggest that although the available soil Pu may remain constant in this study, the rate of accumulation or flux may be proportional to water flux or transpiration. An important observation in these data is that although dry matter production and Pu accumulation increase in a similar fashion, CR values actually decrease by a factor of 20 over the 84-day period. With other soils, the patterns of dry matter production and Pu content are similar, but CR values decrease by as little as a factor of 3. The behavior of CR values in these studies result from their calculation at one point in time based on two concentrations, one of which (soil concentration) bears little relevance to total available pools. This effect is better seen when comparing Pu uptake data and CR values using a number of techniques and larger quantities of soil with all other factors constant. Methods must be developed to better quantitate Pu availability based on rates of accumulation.

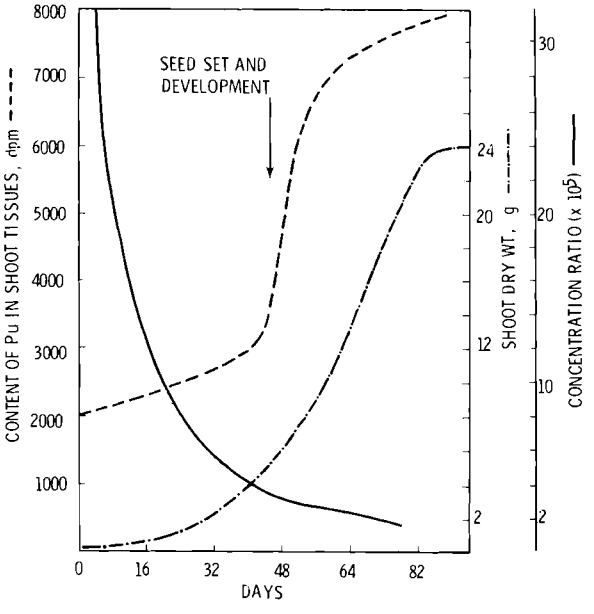


FIGURE 2.2. Comparison of Shoot Pu Content, Shoot dry wt and CR Values Based on Plant Age

Characterization of Pu form in plants has been continuing. Studies with various chemical forms of Pu, have shown organic Pu complexes to predominate in plant tissue, especially with respect to transport forms in xylem exudate. Several organic compounds, present in high concentration in both roots and exudate, are capable of effectively complexing Pu. Unfortunately, chromatographic properties of several of the plant complexes coincide with Pu·DTPA. Methods are being developed to resolve this problem. Studies are also underway to resolve the behavior of endogenous Pu-complexes in plants. These have shown that Pu·DTPA supplied to leaves is readily transported to sink leaves, seeds and pods but not mature leaves (>6% in 24 hr), a characteristic of phloem mobile compounds. Although DTPA and similar chelates are not held to be phloem mobile, these preliminary studies would suggest that they may have a function in maintaining Pu solubility enabling subsequent complexation by endogenous organic compounds which are phloem mobile.

Microbial Transformations of Pu

Mixed Cultures of Organisms

Transformation of the chemical form of a soluble complex of Pu was previously shown, to occur in mixed cultures of Pu-resistant fungi and bacteria. Results of analysis of the exocellular and intracellular soluble fractions of these cultures by gel permeation chromatography, thin-layer chromatography, and thin-layer electrophoresis indicated the formation of components of greater molecular weight than the original Pu·DTPA. It was also shown that modification of the form of Pu·DTPA occurred when added to a mixed culture of organisms which were not previously exposed to Pu. The changes in solubility of the initially soluble Pu complex varied with carbon source and culture (Table 2.3a, b).

Further studies were designed to answer several questions raised by these results. Is the capability to modify Pu as a result of metabolic processes inherent in only a few specific organisms or is this a common phenomenon? Does the growth of different organisms result in different or

TABLE 2.3a. Distribution of Pu in Mixed Microbial Cultures Exposed to Pu at Stationary Growth Phase and Grown on Different Carbon Sources

	Distribution of Plutonium (%) in Cultures			
	Fungi		Bacteria	
Fraction	Mixed Sugars	Organic Acids	Mixed Sugars	Organic Acids
Exocellular Medium	75	42	39	89
Intracellular Soluble	0.49	0.068	8.3	2
Cell Debris	10	42	28	8.7

TABLE 2.3b. Distribution of Pu in Mixed Microbial Cultures Continuously Exposed to Pu and Grown on Different Carbon Sources

	Distribution of Plutonium (%) in Cultures			
	Fungi		Bacteria	
Fraction	Mixed Sugars	Organic Acids	Mixed Sugars	Organic Acids
Exocellular Medium	29	54	46	88
Intracellular Soluble	4.2	0.24	2.7	4
Cell Debris	29	39	31	3.5

similar transformations of Pu? What is the potential for modification of an insoluble form of Pu? Are the modified forms of Pu mobile in soils? The answers to these questions are important to the prediction of the fate of Pu in soils and availability for plant uptake.

Modification of the Chemical Form of Complexed and Hydrolyzed Pu by Pure Cultures of Fungi

To answer the questions arising from the studies of Pu transformations by mixed cultures of soil organisms, 59 individual, pure fungal cultures isolated from soil on the basis of carbon source and resistance to Pu, and other metals, were tested for their ability to modify the chemical form of Pu·DTPA and $Pu(OH)_n$. These studies were conducted in liquid culture using a standard mineral base medium with glucose as the carbon source at a Pu level of 0.02 μg/ml for both chemical forms.

In 50 of the 59 cultures, the level of Pu (complexed and hydrolyzed) had no effect on growth. In seven cultures, a significant reduction in growth was observed for the Pu·DTPA treatment and in two cultures, the $Pu(OH)_n$ treatment reduced growth significantly. In contrast to the distribution observed with Pu·DTPA in the presence of a mixed fungal culture grown on sugars (Table 2.3), only 8 of 59 pure fungal cultures showed less than 5% of the Pu soluble (< 0.01μ) at the end of growth. Surprisingly, the form of the added Pu (i.e., complexed or hydrolyzed) did not appear to be the major factor in the final solubility of Pu in the exocellular medium after growth. In five of the cultures, the final solubility was greater for the initially hydrolyzed Pu form (Table 2.4). It would appear that the diversity in exocellular products from the individual fungal strains was an important factor.

TABLE 2.4. Modification of the Solubility of Complexed and Hydrolyzed Pu by the Growth of Selected Soil Fungal Isolates

	Solubility of Pu in Solution After Growth When Pu Added as			
	Pu·DTPA		Pu(OH)n	
Fungal Isolate #	0.4	0.01	0.4	0.01
	- - - % - - -			
Sterile Control	100	100	92.2	15.4
510	0.51	0.40	81.5	34.4
369	100	96.4	4.39	2.58
527	0.34	0.26	2.36	1.99
401	1.96	0.90	5.82	1.61
326	4.35	0.90	2.73	1.40
458	1.17	0.34	1.01	0.36
451	3.37	2.74	2.86	0.021
453	4.62	1.07	0.37	0.059

With all cultures tested, characterization of the chemical form of the Pu remaining in the exocellular medium by thin-layer chromatography and thin-layer electrophoresis revealed that, in the case of the Pu·DTPA treatments, the Pu·DTPA molecule was either attaching itself to one or more other components or being incorporated intact into

another larger molecule to varying degrees. Under the conditions of thin-layer electrophoresis, the Pu·DTPA was regenerated or freed from the components which previously exhibited different mobility from Pu·DTPA on thin-layer chromatography. In the case of the hydrolyzed Pu treatments, several complexed species were demonstrated, although the bulk of the soluble Pu remained at the origin in both TLC and TLE. This could be explained on the basis of the size of the molecules in association with Pu or its continued presence as collodial Pu.

Soil Mobility of Pu in the Exocellular Medium After Growth of Pure Fungal Cultures

Although previous studies demonstrated that both the complex form of Pu and a hydrolyzed form of Pu were transformed through fungal and bacterial metabolism, the question remained as to whether these new complexes of Pu were mobile in soils and therefore likely available for plant uptake.

A series of soil columns was prepared (Ritzville silt loam, < 2 mm) and 2.0 ml of the clarified (< 0.4μ) exocellular fraction was allowed to pass by gravity through the column. The clear leachate was immediately analyzed for Pu and subjected to TLC and TLE analysis for comparison to similar analyses made immediately after growth.

The fraction of soluble Pu from cultures treated with Pu·DTPA in the leachate from the soil column was generally much greater than from cultures treated with $Pu(OH)_n$ (Table 2.5). Previous studies comprising 260 individual columns with four different metals present in the fungal growth media, showed that the fraction passing the < 0.01μ filter best described "soluble" species. The data for Pu indicate that the 0.01μ filtration removed Pu which in certain cultures would pass through the soil in both complexed and hydrolyzed Pu treatments. The greater fraction of the Pu passing through the soil from the exocellular media cultures treated with Pu·DTPA may be related to the phenomenon previously described. That is, the Pu·DTPA was incorporated in larger molecules at the end of growth and passage through the soil released the very soluble Pu·DTPA. It is also possible that this mechanism played some role in the few cases where the Pu passing soil was much greater than the "soluble" levels in cultures treated with hydrolyzed Pu. Insight into the mechanisms controlling the mobility of the microbially modified Pu will be obtained after chemical characterization studies, currently in progress, are completed.

TABLE 2.5. Soil Mobility of Pu Soluble After Growth of Pure Fungal Cultures Grown in the Presence of Complexed and Hydrolyzed Pu

	Fraction of Initially Soluble Pu Passing Soil Column from Cultures Treated with	
Isolate #	Pu·DTPA	Pu(OH)n
	- - - % - - -	
Controls		
Pu Only	100	4000
Sterile Medium + Pu	78.5	0.7
Isolates		
510	107	0.3
369	2.4	5.8
527	92	2.0
401	200	2.4
326	50.1	0.9
458	276	22.2
451	86.2	4000
453	598	25.0
Remaining Isolates	0.8-395	1.5-131

Organic Complexants in Aqueous Nuclear Wastes

Millions of gallons of liquid and solidified radioactive wastes are currently in storage. Although the organic content is not expected to exceed 1% of the salt cake remaining after evaporation, the actual state of the organics present is unknown and presumed complex.

A literature search, partially funded by the EPA, was aimed at identifying organic complexing agents that could be expected in aqueous process wastes, either through direct addition or due to chemical and radiolytic degradation. Known organic complexing agents used in waste processing procedures are surveyed. Oils and detergents used in routine maintenance are potential sources of complexing agents. In addition, degradation of diluents commonly used in process systems or of the added complexing agents themselves yield products possessing observed chelation ability.

Attention is paid to fission products and transuranics in discussion of the potential for interference with normal sorption of nuclides by soil due to the presence of organic

ligands. Nuclides discussed are Am, Sb, Ce, Cs, Co, Cm, Eu, I, Np, Pm, Pu, Ra, Ru, Tc, U, and Zr. General conclusions are made regarding relative chelate stability for the various classes of nuclides. Actual instances where behavior of a nuclide in soil was altered due to the presence of ligand activity are included (Table 2.6).

TABLE 2.6. Potential Sources of Complexing Agents from Aqueous Process Wastes

Ligands Used Directly in Processes:

TBP	HDEHP	Tartaric Acid
DBBP	NTA	Hydroxyacetic Acid
HEDTA	DTPA	Sugar
EDTA	Citric Acid	

Impurities in, or Degradation Products of, Primary Ligands:

DBP	Short chain hydrocarbons
MBP	H_2MEHP
Butyl alcohol	Tri (2 ethyl hexyl) phosphoric acid
Diluted ether	2 ethyl hexanol
Condensed Phosphates	HDEHP polymer
Polymeric TBP	Tartaric acid
Phosphonic acid	Oxaladic acid
Butanol or Methyl nitrate	

Probable Primary Diluent Degradation Products:

Nitro-paraffins:	nitro ($-NO_2$)
	nitrate esters ($-ONO_2$)
	nitroso (-N = 0)
	isonitro (NOOH)
	polymitro compound
Carbonyl compounds:	carboxylic acids (-COOH)
	carbonyls (-CHO)
Polymers and solids formation:	Olefinic and carboxylic acid dimers
	Polymerized amines

Potential Secondary Degradation Products:

Nitroalcohols
Nitroalefins
Dinitro compounds
Aci-nitro salts
Nitrolic acid
Hydroxamic acids
Oximes

• Quantitative Aspects of Plutonium Field Studies

Principal Investigators: R. O. Gilbert, P. G. Doctor
and L. L. Eberhardt

The purpose of this project is to provide quantitative and statistical support and methodology for the design and analysis of plutonium and other transuranic ecosystem and field studies. Emphasis is on developing methods for sampling and statistical analyses to identify and deal with the extremely high variability in plutonium and other transuranic concentrations in environmental samples and to effectively communicate this information to scientists engaged in Pu field studies. The design problem is being considered in terms of the rather different objectives that may be considered in planning field studies, such as the movement and dynamics of plutonium within and between ecosystem components, estimation of amounts and concentrations in these components, and more generally, sampling to assess the need for "clean-up" or to evaluate hazards. Specific activities are: 1) the publication of a statistics newsletter (TRAN-STAT), 2) research into appropriate statistical methods for the analysis of ratio data and for effectively summarizing and reporting transuranic field data, 3) preparation of a manuscript giving sources of information on sampling and statistical methods, 4) research in evaluating appropriateness of Kriging techniques for estimating spatial pattern and inventories, 5) identification of sampling designs and analyses appropriate for cleanup situations, and 6) maintaining contact with other statisticians and scientists engaged in transuranic field studies.

Statistics for Environmental Transuranic Studies (TRAN-STAT)

TRAN-STAT is an informal newsletter being sent to scientists engaged in environmental transuranic studies. TRAN-STAT is devoted to stimulating an increased awareness of the role that statistics can play in the design and analysis of transuranic field studies. The first issue was distributed in September 1977. Future issues, each discussing a different topic, are expected to be distributed about every 2 months.

Topics are expected to include regression and ratio analysis, nonparametric statistics, methods for analysis of spatial data for estimating concentration contours and/or inventories, and statistical approaches for deciding whether a contaminated area requires remedial action. New and pertinent references to statistical techniques will be included whenever possible, and comments received concerning prior issues will be discussed.

Analysis of Ratio Data

Our interest in ratio data stems from its widespread use in field studies to describe relationships and distributions of transuranic elements in ecosystem components. We are

currently preparing a paper (Doctor and Gilbert, 1977) dealing with the appropriate use of ratios in environmental transuranic studies. Two types of ratios are discussed, concentrations and pure ratios. The appropriate use of ratios in transuranic field studies demands at least some knowledge of the relationship between the two variables (Y and X) composing the ratio Y/X. If the ratio is a concentration (e.g., nCi/g), then Y should be at least approximately proportional to X. If the ratio is meant to express the relationship between two variables Y and X measured in the same units (e.g., ^{238}Pu to ^{239}Pu in nCi), then either Y should be approximately proportional to X, or the process under study should be a function of the ratio Y/X rather than of X or Y individually. Using a ratio when none of the above cases seems reasonable, at best, probably provides no information on the relationship of the two variables and, at worst, could be misleading.

Sources of Information on Statistical Methods

A paper is being prepared that gives sources of information on sampling and statistical methods relevant to transuranic studies. This report also suggests some further statistical research needed on particular problems.

Four categories of objectives are discussed: 1) descriptive sampling, 2) sampling for spatial pattern, 3) analytical sampling, and 4) sampling for modeling. It is important that quite specific objectives be identified in order to devise an efficient sampling plan. Stratified random sampling and double sampling are discussed for estimating a total amount (inventory) over a geographical area. "Sampling for spatial pattern" is discussed relative to the problem of determining the portion of a contaminated area needing cleanup. Many aspects of designing such studies remain unresolved or uncertain. Analytical sampling, or "sampling for comparisons" is discussed relative to interlaboratory comparisons, the frequent occurrence of skewed frequency distributions, and to the common practice of computing ratios of highly variable quantities in transuranic studies. "Sampling for modeling" refers to designs appropriate for estimating rate constants. Problems in this area needing further attention are discussed. Some problems common to all types of studies are also briefly described.

Kriging Techniques for Estimating Spatial Pattern

Kriging is a relatively new technique that takes into account the spatial correlation structure that may exist to obtain improved estimates of spatial pattern and/or inventories. Dr. Pierre Delfiner of the Centre de Geostatistique, School of Mines, Fontainebleau, France, is an expert on Kriging and is on a consulting contract with PNL to provide us with guidance on the applicability of Kriging to transuranic field studies. He has written a paper (partly funded by the Nevada Applied Ecology Group) that presents an approach to estimating average $^{239-240}Pu$ concentrations in soil in Area 13 on the Nevada Test Site. The methods presented in this paper should also be useful at other locations. We are also experimenting with Kriging techniques using a Kriging program called BLUEPACK available on the Nevada Operations Office computer in Las Vegas (accessed via a high speed terminal from Boeing, Richland). This will provide practical experience in the use of Kriging.

Cleanup Studies

We have identified several statistical approaches that may be acceptable (under certain assumptions) for helping to decide whether a contaminated area needs remedial action. These were discussed in the second issue of TRAN-STAT (November 1977). It is presumed that a decision regarding remedial action is to be made for a unit of land from which samples are to be collected. In general, the decision could be based on an average concentration or on the proportion of sample concentrations greater than some specified concentration. The validity of all methods highly depends on obtaining representative samples from all portions of the area being sampled. Other assumptions, such as normally or lognormally distributed data, are also usually necessary. The occurrence of unsuspected "hot spots" or trends in concentrations are problems that could result in wrong decisions based on statistical procedures. Kriging may prove to be the preferred method for estimating average concentrations in some cleanup situations.

Interaction and Communication

To maintain contact with potential users of our work we attended DBER's workshop on

Environmental Research for Transuranic Elements held at Woods Hole, April 19-22, 1977. Further interaction occurs as a result of our work for the Nevada Applied Ecology Group (NAEG) (see next article) and our publication of TRAN-STAT. We view effective communication as the key to the use of improved statistical procedures.

NAEG Transuranic Field Studies on the Nevada Test Site

The purpose of this program [funded by the Nevada Applied Ecology Group (NAEG)] is to provide statistical design and analysis support for environmental transuranic studies being conducted by the NAEG either on or adjacent to the Nevada Test Site (NTS). We have been engaged in this effort since 1971. Our work for the NAEG has led to an awareness of the many statistical problems being investigated in our Quantitative Aspects of Plutonium Field Studies program for DBER.

Specific activities for the NAEG during FY-77 are as follows:

1) Initial design and statistical analysis for the first NAEG nuclear site studies. Objectives of these studies include estimating the spatial pattern and total amount of various radionuclides in soil and vegetation. Several hundred initial soil samples have been collected. The statistical design and analysis procedures used thus far at three nuclear sites were reported at the March 1977 NAEG Information Conference.

2) Design and analysis of a field study to estimate the inventory of $^{239-240}Pu$ in blow-sand mounds as opposed to desert pavement areas on NTS. A major study was designed for which whole blow-sand mounds were collected and aliquots were withdrawn for plutonium analysis. Comparisons were made and total amounts of plutonium were estimated for mound tops, mound bottoms, and desert pavement areas. The change in plutonium to americium ratios in soil over time was investigated in addition to a comparison of portable field instrument (FIDLER) readings of ^{241}Am over mounds as opposed to desert pavement areas. This work is reported in Gilbert and Essington (1977) and Essington et al. (1977).

3) Additional soil and vegetation samples were collected at previously studied safety-shot sites on NTS for obtaining improved estimates of $^{239-240}Pu$ inventory. These revised estimates are not yet available.

4) A special soil sampling study in Area 13 was designed for estimating the variability and correlation between ^{241}Am concentrations in soil samples collected various distances apart. This information is needed in our evaluation of Kriging techniques.

5) A special study was designed to study the relationship between ^{241}Am concentrations and size of aliquot. For each of five aliquot sizes (1 g, 10 g, 25 g, 50 g, and 100 g) twenty aliquots were analyzed for ^{241}Am. The results will be reported at the February 1978 NAEG information meeting.

6) Dr. Pierre Delfiner's work on the applicability of Kriging to plutonium studies is partially funded by the NAEG. As mentioned above he has used data from Area 13 collected by the NAEG for this purpose.

- **Ecological Distribution and Fate of Plutonium and Americium in a Processing Waste Pond on the Hanford Reservation**

Principal Investigators: R. M. Emery, D. C. Klopfer and M. C. McShane

U Pond, located on the Hanford Reservation, has received low-level quantities of plutonium (Pu) and americium (Am) longer than any other aquatic environment in the world. Its ecological complexity and content of transuranics make it an ideal resource for information concerning the movement of these actinides within and out of an aquatic ecosystem.

U Pond has been intensively inventoried for Pu concentrations in the ecological compartments and characterized limnologically in terms of its physicochemical parameters, biological productivity and community structure. This work provides a basis for evaluating the pond's performance in retaining waste transuranics. The quantitative estimation of export routes developed by this study is important in determining how effectively such ponds act as retainers for transuranic wastes. A more complete report has been prepared.

Ecological Export of Pu

One of the major goals of the U Pond study has been to obtain sufficient information about the pond's ecosystem, and the distribution of Pu within it, so that Pu export routes can be assessed quantitatively. Although it is often difficult to measure with reasonable certainty the parameters necessary for describing these export routes, the purpose of this work is to formulate the best expressions of export given the conditions which limit this process. The objectives are to determine ranges of quantities of Pu in the pond's ecosystem and assess the amount of Pu being exported in relation to this inventory. To accomplish this task, estimates were made of the pond's Pu inventory quantities on a basis of minimum, mean and maximum values for each ecosystem compartment, shown in Figure 2.3, to postulate the amount of these inventories that are exported yearly.

In its 33-yr history, U Pond has received an estimated 1 Ci of Pu which appears to have been retained by the sediments. In relative terms, sediments, submerged plants and gastropods have the highest concentrations of Pu, ranging from 3.2×10^{0} to 6.9×10^{2} pCi/g. Emergent plants and the remaining fauna have Pu concentrations ranging from 4.0×10^{-1} to 6.1×10^{1} pCi/g. Emerging insects have the highest Pu concentrations of the latter group, ranging from 3.2×10^{1} to 6.1×10^{1} pCi/g.

The mean Pu inventory of the sediment is 1.7×10^{9} nCi, ranging from 1.3×10^{9} to 2.0×10^{9} nCi of Pu (Figure 2.4). This essentially represents the total pond inventory, since > 99% of the Pu in the pond are found in the sediments. The mean Pu inventory for the biota is 6×10^{6} nCi, ranging from 1×10^{6} to 1×10^{7} nCi (Figure 2.4). Among the biota, plant life contain > 99% of the Pu. Diatoms and pondweed (Potamogeton) alone account for > 99% of the Pu in plants. Emergent insects contain < 1×10^{-1}% of the Pu in biota and < 1×10^{-3}% of the Pu in the pond. The inventory of this compartment has particular relevance, since it is the only direct

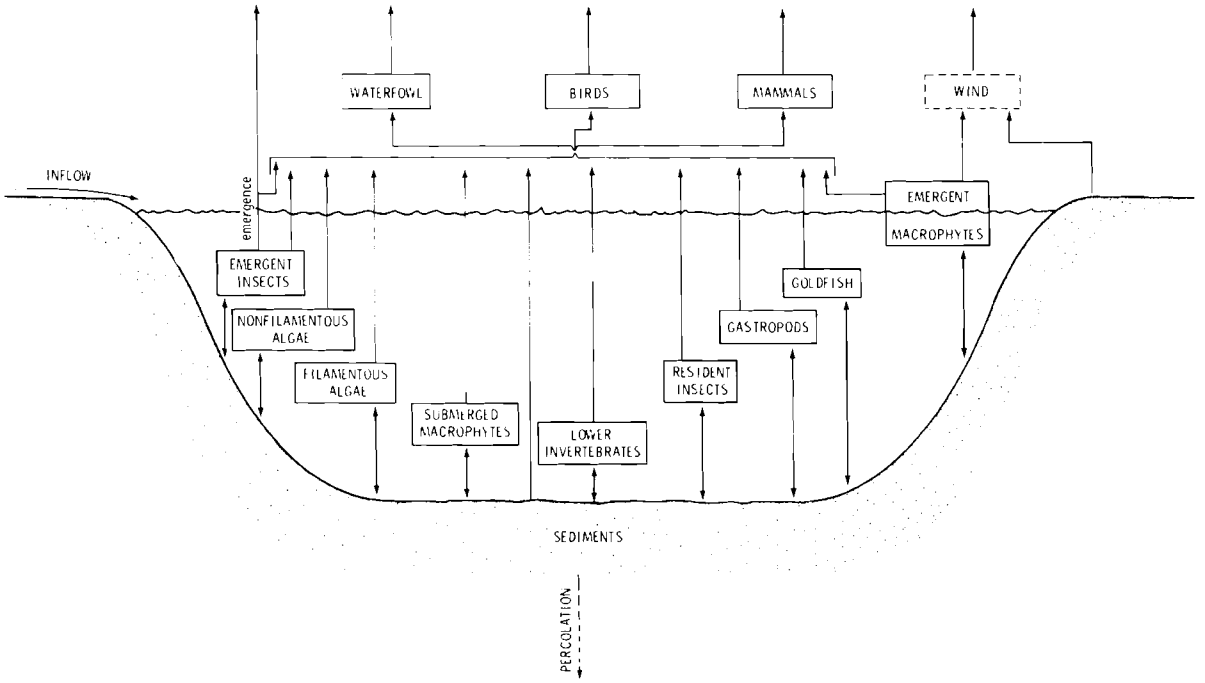


FIGURE 2.3. Schematic Representation of Ecosystem Compartments and Pu Export Routes from U Pond via Biological Mobilization, Wind and Percolation.

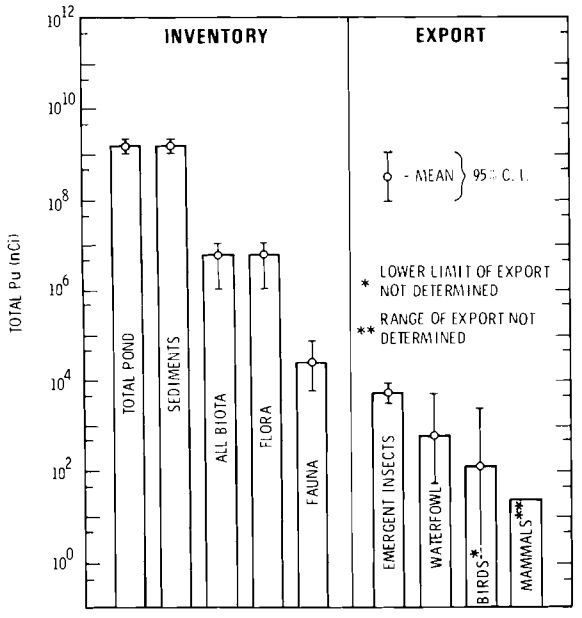


FIGURE 2.4. Inventories of Pu in Ecological Compartments of U Pond Compared with Estimated Quantities that are Exported from the Pond Annually. Export of Pu by percolation or wind do not appear to be significant.

biological route of export from the pond. Remaining pond biota contain < 1 x 10^{-2}% of the total Pu inventory in the pond and can leave the pond only by the forces of external export vectors.

If all emergent insects successfully leave the pond, they could export from 3.5 x 10^{3} to 7 x 10^{3} nCi of Pu. These quantities are more than 5 orders of magnitude lower than the total pond Pu inventory (Figure 2.4). Estimated quantities of Pu annually exported by waterfowl range from 4 x 10^{1} to 4 x 10^{3} nCi, with a mean annual export of 5 x 10^{2} nCi of Pu (Figure 2.4). Other birds appear to export about 1 x 10^{2} nCi of Pu each year, with a maximum of 2 x 10^{3} nCi (Figure 2.4). These export quantities are about 6 orders of magnitude lower than the total inventory of Pu in the pond. Mammals are estimated to annually export a maximum of 3 x 10^{1} nCi of Pu from the pond (Figure 2.4), which is at least 5 orders of magnitude lower than the minimum total Pu inventory of the pond. There is no apparent significant export of Pu from the pond via wind.

U Pond has been exposed to Pu since 1944, longer than any other aquatic system. In its lifetime, it has received about 1 Ci of ^{239,240}Pu and ^{238}Pu from reprocessing operations. This 14-acre pond provides a realistic

illustration of the mobility of Pu in a lentic or nonflowing ecosystem. Although this pond has a rapid flushing rate, it is highly enriched with plant nutrients, ecologically well established with a natural complexity of populations and diversity of communities, and in continuous contact with associated terrestrial life. It appears to effectively bind the Pu discharged into it and prevent it from moving significantly into routes leading to man and other remote life. The environmental behavior of Pu in U Pond appears to be quite similar to that of other aquatic systems having vastly different ecological character. As long as this pond remains in its present condition, the likelihood of it releasing hazardous quantities of Pu to man and his environment is very small.

- **Use of Analog Elements to Predict the Equilibrium and Behavior of Transuranic Elements in the Environment**

Principal Investigators: W. C. Weimer, J. C. Laul and J. C. Kutt

Several naturally-occurring elements with chemical properties similar to those of selected transuranic elements have been chosen and are being examined as potential predictors of transuranic geochemical behaviors. This approach may allow the estimation of the long-term behaviors of transuranic elements in the environment by analyses of the steady-state behaviors of their analog elements. The elements receiving principal attention are the transuranics Am and Cm and their proposed lanthanide element analog Nd.

Incorporation of Naturally-Occurring Rare Earth Elements by Vegetables

The incorporation of several rare earth elements by vegetables grown on a natural soil has been determined. The purpose of this investigation was to evaluate how the uptake of these trivalent elements was influenced by the ionic size of the elements, which changes uniformly across the lanthanide series. The results demonstrate that the plant uptake of these elements is a smooth function of the elements' ionic radii.

The incorporation of nonessential trace elements by plant species is a complex function of many factors. Two of these factors which may be particularly significant are the ionic charge and the ionic radius of the element in question. All of the lanthanide elements commonly exhibit the same environmental oxidation state of +3 and, therefore, offer a unique opportunity to evaluate uptake as a function of ionic radius since there is a decreasing ionic size across the series due to the lanthanide contraction. Also, since the chemical properties of the lanthanide elements and their corresponding actinide elements are in several instances quite similar, an estimate of the potential uptake of the transuranics may be obtained from the rare earth element uptake for elements of the same charge and similar ionic radius.

Samples of the fruiting portions of four vegetable varieties and of contiguous soils were obtained from the Health and Safety Laboratory (HASL) garden plot in Massachusetts. Extreme care was exercised during the collection and processing of the vegetable samples to insure that no surface contamination of dust was transferred into the fleshy portions of the vegetables which were actually analyzed. The contents of the rare earth and other elements in these samples were determined by radiochemical neutron activation analysis and by instrumental neutron activation analysis.

The concentrations of the rare earth elements in these vegetable samples are extemely low, ranging from approximately 30 ppt to 20 ppb for the various rare earths. These values represent the lowest concentrations ever reported for the rare earth contents of any materials. The rare earth concentrations in the plants and in the cultivated soil samples have been plotted as a function of the elemental ionic radius and are depicted in Figure 2.5. In this figure, all of the rare earth concentrations have been normalized to the concentrations in chondrites to facilitate visual interpretation. The rare earth patterns in the vegetables and in the soils on which they were grown vary exactly as a smooth function of the ionic radius. (The apparent discontinuity for Eu is due to the Eu enrichment in chondrites.) The slopes of

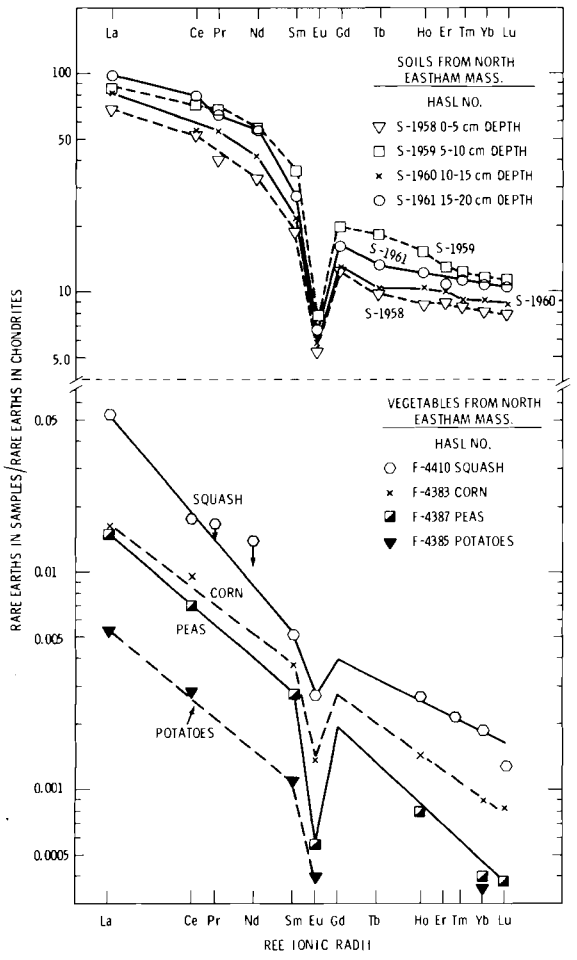


FIGURE 2.5. Rare Earth Patterns in HASL Samples.

these curves (from La to Lu) for the vegetables are nearly the same as for the soils; however, there does appear to be some discrimination against the heavier rare earth elements during uptake. This is probably related to the fact that rare earth patterns for the soils are for the bulk soils, whereas the plant uptake will be from a small portion of this total pool. This pool of plant-available elements may have a slightly different rare earth pattern. Nevertheless, the uptake of these several trivalent lanthanide elements appears to be directly related to each element's ionic radius as the factor of principal influence.

If the ionic radius is the dominant factor affecting the uptake of the lanthanide elements of identical charge and very similar chemical properties (solubility products, hydrolysis constants, complex formation constants), then the ionic radius should also be a major factor in relating the uptake of other trivalent elements with similar properties to the uptake of the lanthanides. Specifically, the uptake of the lanthanide Nd and the actinides Am and Cm should be nearly identical because of the close correlation of the properties of these three elements.

Plant Uptake of Am, Cm and the Chemical Analog Nd

Dryland and irrigated plant species (cheatgrass and snapbeans) have been grown on soils treated with the same chemical forms of Am, Cm and Nd. Under the conditions examined, the concentration ratios for the uptake of these ions from the soils into the plants were nearly identical, indicating that these plant species do not differentiate between these trivalent ions during uptake.

On the basis of several chemical properties including ionic charge, ionic radius, solubility product values, hydrolysis constants, and formation constants, the element Nd has been selected as a potential chemical analog for the two transuranic elements Am and Cm. Previous investigations in our laboratory have demonstrated that purely physical-chemical principles will influence the behavior of Am, Cm and Nd nearly identically in soil suspensions. Therefore, these comparative behavior investigations were expanded to include an evaluation of the ability of plants to discriminate more precisely against one or two of these elements during uptake.

Samples of cheatgrass (Bromus tectorum) and snapbeans (Phaseolus vulgaris) were grown on chemically treated soils in lysimeters on the Arid Lands Ecology (ALE) Reserve and in growth chamber pots at Oak Ridge National Laboratory (ORNL), respectively. The same chemical form of Am, Cm and Nd was added to the soils of these experimental containers prior to planting of the vegetation seeds. For the lysimeter plantings, the nitrate forms of these elements were added to a small portion of soil which was then placed as a layer near the top of the lysimeter and covered with unspiked soil. The soil for the growth chamber plantings was uniformly mixed with the citrate forms of Am, Cm and 146-enriched Nd. The harvested plant samples were analyzed by instrumental and radiochemical neutron activation analysis and by alpha spectrometry.

The concentration ratios (calculated on the basis of the total weights of isotopes and soils in the containers) for the uptake of Am, Cm and Nd are given in Table 2.7. These data show that the uptake of these elements by the snapbeans was identical for each of the elements. The concentration ratios for the uptake by cheatgrass are also very similar, ranging from 2.5×10^{-4} to 6.5×10^{-4}, or a factor of approximately 2.6. This evidence indicates that Nd acts as a good short-term analog element and suggests that it may also serve as a useful long-term predictor of transuranic behavior since physical-chemical and biochemical studies both have demonstrated the like elemental behaviors.

TABLE 2.7. Plant Uptake of Am, Cm, and Nd.

Plant	Concentration Ratio		
	Am	Cm	Nd
ALE Cheatgrass-Leaves and Stems (Lysimeters)	4.8×10^{-4}	2.5×10^{-4}	6.5×10^{-4}
ORNL Snapbeans-Leaves and Stems (Growth Chamber)	1.1×10^{-3}	1.3×10^{-3}	1.2×10^{-3}

• Weathering and Aging of Transuranics

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Technical Assistance: M. A. Combs, M. J. Harris, M. C. McShane, L. F. Nelson and H. A. Sweany

The release of transuranium elements from the nuclear energy industry poses questions for which data are very limited. Information is needed to quantify the movement of radiocontaminants through food chains to assess the potential hazards to man. Quantitative data are required on the transfer of transuranics from soil to vegetation. Of special interest are the effects time (weathering, aging, and associated biological processes in soil) has on the availability of transuranium elements to be taken up by plants from soil. This study provides information on the effects of time on the uptake of transuranics from soil by range and crop plants.

1977 Progress

At present, 360 lysimeters have been prepared and placed in an exclosure located near the U.S. DOE Arid Lands Ecology Reserve (ALE) headquarters. Isotopes of ^{238}Pu, ^{239}Pu, ^{244}Cm, ^{241}Am and ^{237}Np were individually placed in soil contained by the lysimeters. The water-tight lysimeters were constructed from polyvinyl chloride pipe (PVC), 13.2 cm in diameter and 1 m long. The radioisotopes were mixed in either 3.4 kg or 100 g of soil. The 3.4 kg mixtures were placed in the lysimeters in a layer 20 cm thick, 10 cm below the soil surface. The 100 g mixtures were also placed 10 cm below the surface, but were only 0.5 cm thick. The aboveground plant parts were harvested at maturity and divided into selected components. Cheatgrass (Bromus tectorum) and other plants which invaded the lysimeters were separated by species and the entire aboveground plant parts analyzed. The seeds from the peas and barley were analyzed separately from the remainder of the plant. The entire alfalfa plant (three separate harvests) were analyzed. The radiochemical analyses were done by LFE Environmental Analysis Laboratory in Richmond, CA.

The relative uptake of ^{238}Pu, ^{239}Pu, ^{241}Am, ^{244}Cm and ^{237}Np by four different plant species are presented in Table 2.8. The data presented are for plants grown in lysimeters in which the transuranics had been amended to 3.4 kg of soil. There did not appear to be any difference in the uptake of ^{238}Pu and ^{239}Pu. Uptake of the plutonium isotopes was the lowest among the transuranics examined. Comparing the other transuranics to plutonium, the uptake of ^{241}Am was about 20, ^{244}Cm about 10, and ^{237}Np about 4000 times (40,000 when comparing the seeds) greater than ^{238}Pu or ^{239}Pu. The relative uptake of all the transuranics was about 10 times higher in the peas and alfalfa than in the cheatgrass and barley. The concentration of the barley seeds were approximately 10 to 100 times less than the remainder of the plant. The pea seeds were from 100 to 400 times less than the other plant fragments.

TABLE 2.8. Relative Uptake of Transuranium Elements by 4 Different Plant Species. The transuranics were amended to the soil in a 20 cm band located 10 cm below the surface.

$\frac{\text{pCi/g Dry Vegetation}}{\text{mCi/Lysimeter}}$ ± Standard Error[a]

Isotope	mCi per Lysimeter	Cheatgrass	Barley		Peas	
			Seed	Stem-Leaf	Seed	Stem-Leaf-Pod
^{238}Pu	0.108	1.3 ± 0.3	0.21 ± 0.12	5.9 ± 0.6	0.24 ± 0.06	41 ± 11
^{238}Pu	1.06	2.7 ± 0.6	0.10 ± 0.06	15. ± 10.	0.064 ± 0.026	25 ± 8
^{239}Pu	0.103	12 ± 3	0.094 ± 0.013	4.8 ± 2.0	0.0 ± 0.039	58 ± 19
^{239}Pu	0.989	5.0 ± 0.8	0.085 ± 0.017	3.8 ± 1.0	0.089 ± 0.045	37 ± 8
^{241}Am	0.0946	140 ± 20	0.35 ± 0.08	34 ± 6	1.8 ± 0.5	(4.8 ± 1.1) x 10^2
^{241}Am	0.983	48 ± 11	0.16 ± 0.02	9.7 ± 1.0	2.8 ± 0.8	(7.1 ± 1.6) x 10^2
^{244}Cm	0.103	65 ± 25	0.90 ± 0.28	39 ± 5	3.0 ± 1.1	(6.3 ± 1.7) x 10^2
^{244}Cm	1.04	80 ± 17	0.16 ± 0.03	10 ± 2	4.7 ± 0.8	(4.8 ± 0.4) x 10^2
^{237}Np	0.101	(1.1 ± 0.1) x 10^4	(0.43 ± 0.17) x 10^4	(2.7 ± 0.3) x 10^4	(0.37 ± 0.04) x 10^4	(23 ± 7) x 10^4
g dry vegetation per lysimeter		3.87 ± 0.30	14.49 ± 0.48	24.63 ± 0.72	12.52 ± 0.79	11.08 ± 0.31

Isotope	mCi per Lysimeter	Alfalfa		
		1st Harvest	2nd Harvest	3rd Harvest
^{238}Pu	0.108	30 ± 7	13 ± 3	58 ± 42
^{238}Pu	1.06	16 ± 2	22 ± 6	34 ± 13
^{239}Pu	0.103	80 ± 21	17 ± 5	45 ± 20
^{239}Pu	0.989	24 ± 7	14 ± 2	16 ± 2
^{241}Am	0.0946	(60 ± 14) x 10^2	(1.7 ± 0.2) x 10^2	(2.7 ± 0.4) x 10^2
^{241}Am	0.983	(7.0 ± 1.1) x 10^2	(1.7 ± 0.5) x 10^2	(3.5 ± 0.5) x 10^2
^{244}Cm	0.103	(7.8 ± 1.9) x 10^2	(2.3 ± 0.5) x 10^2	(3.8 ± 0.6) x 10^2
^{244}Cm	1.04	(3.5 ± 0.8) x 10^2	(1.5 ± 0.3) x 10^2	(2.5 ± 0.6) x 10^2
^{237}Np	0.101	(21 ± 7) x 10^4	(5.5 ± 1.3) x 10^4	(4.7 ± 1.2) x 10^4
g dry vegetation per lysimeter		7.34 ± 0.79	9.97 ± 0.28	6.29 ± 0.21

[a] $n = 5$

- **Radioecology of Uranium**
- **Radioecology of Waste Management Areas (ROWMA)**

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Radioecology of nuclear wastes was pioneered in the western United States on the Hanford Reservation during the early 1940's. Initially, attention was paid to the Columbia River because it provided coolant water for production reactors. Ecological studies centered upon fish and waterfowl as vectors of radionuclides to people. With the abandonment of production reactors a few years ago, the radionuclide content of Columbia River water has declined and attention is now being directed towards radionuclides released into the environment as a result of chemical processing of irradiated fuel. Places that have received radioactive wastes are solid waste burial grounds, liquid effluent release belowground, surface ditches and ponds, and the terrestrial environment around chemical processing facilities that have received airborne deposits of radioactive debris from stacks.

ROWMA studies reported here are concerned with the degree of uptake of various radionuclides from soil, sediments, and water by terrestrial and aquatic biota, the potential of removal of radioactivity from waste management areas by biota, and the transfer of radionculides between organisms and ecological trophic levels. The long-term objective of ROWMA effort is to delineate and quantify underlying ecological processes affecting transport of radioactivity. A portion of effort is allocated also to site specific problems caused by biotic vectors; i.e., birds, insects and wild animals that cause wider dissemination of radioelements. Important companion studies also are underway. They are funded through local intercontractor support agreements rather than through DBER and involve short-term assistance to Hanford plant personnel concerned with characterization of particular landscape subunits impacted by operation of nuclear facilities. See Section 8.0, for intercontractor studies.

First year studies on the ROWMA projects centered upon characterizing the abiotic environment and the biota of representative waste management areas on the Hanford Reservation.

First year studies using uranium isotopes centered around the growing of plants in uranium-contaminated soil.

Radioecology of Aquatic Systems

Many aspects of the nuclear industry associated with fuels and weapons processing and reactor operations require a continuous supply of freshwater. Much of this water receives only low-level contamination and can be safely discharged into open ditches (streams) and basins (ponds) at the facility sites. These wastewater discharges create aquatic environments that frequently support diverse ecological communities which adapt to the often unusual thermal, chemical and radiological conditions. Such is the case at Hanford, where several large ponds and streams are maintained to receive wastewater from reactor, reprocessing and laboratory operations.

The objective was to obtain data that describe these aquatic environments limnologically and radiologically. The purpose of developing these radioecological profiles was to determine if certain ecological characteristics exist that would set these aquatic systems apart from others not associated with nuclear facilities.

Four ponds and four streams were selected on the basis of waste types currently being received, history of use, and ecological significance. U Pond, located in the 200 West Area, has been receiving wastes from plutonium reprocessing operations for over 30 yr. The Z-19 ditch, which has fed U Pond for the past 5 yr, was the latest of a series of ditches carrying Pu reprocessing wastes to U Pond. B Pond, fed via the B-3 and A-29 ditches, is situated east of the 200 East Area and has recieved chemical sewer wastes and process cooling water from recovery operations since 1945. Gable Mountain Pond, located north of the 200 East Area, has received process cooling water since 1957. West Pond, which occurs naturally, is a mirror of the groundwater table and has no surface water or wastewater sources, although it did receive sanitary wastes at one time. It does, however, contain uranium which may be due to a local natural uranium deposit. The 100-N trench, located at the N-reactor site near the Columbia River, was created in 1965 and receives cooling water from the N-reactor crib.

Gable, B, and U Ponds differed widely in physical dimensions, retention times and sedimentation properties, but were generally similar in chemistry. An exception to this was the unusually high concentrations of nitrogen in B Pond. West Pond showed extreme variation from the other ponds due to the long-term accumulation of dissolved and suspended materials through evaporative concentration. Since West Pond received sanitary wastes during the early development of Hanford, nitrogen and phosphorus concentrations are also disproportionately high. All of the study ditches were generally similar in physicochemical characteristics except B-3 ditch, which was relatively higher in nitrogen but lower in phosphorus concentrations.

The 100-N trench is the only system that showed a difference in biological profile that might be related to a uniqueness in radiological profile. Its radionuclide concentrations and radiation dose rates were much higher than any of the other aquatic sites (Table 2.9). Profiles of algae, macrophytes, and invertebrates indicate a reduced complexity in organism types (Tables 2.10-2.12). However, calculations of diversity indices (Table 2.13) indicated that species and community diversity, and evenness were now the lowest of the aquatic systems studied at Hanford. Also, rates of colonization on bare substrates placed in 100-N trench were higher than on those in B, U, or West Pond (Table 2.13). However, the thermal ranges in 100-N trench, as well as ranges of some chemical parameters, exceeded those of other aquatic systems. Therefore, it may be possible to explain differences in biological profiles between 100-N trench and other systems primarily on the basis of physicochemical conditions.

Radiation Doses to Mice Living on Retired Burial Grounds

The most abundant small mammal living on the burial grounds on the Hanford Site is the Great Basin pocket mouse, Perognathus parvus. This rodent is capable of burrowing to a depth of 4-6 ft if soil conditions permit. Deer mice, which are also captured on the burial grounds, do not normally burrow deeper than 2 ft. This study was initiated to determine if the mice were burrowing deep enough to become exposed to radiation emitted from the buried waste.

The sites examined were the WYE burial ground, 300-North burial ground, 300-6 and 7 burial ground, and a control plot of similar habitat and soil texture. These burial grounds received low-level laboratory wastes which were positioned in trenches 8-10 ft deep and backfilled with cobblestones and gravel.

Live traps were arranged in a grid pattern containing 100 traps spaced at 10 m intervals

TABLE 2.9. Ranges of Radiological Characteristics for Ponds and Streams on the Hanford Reservation. Dosimetry is based on one-month measurements of the sediment-water interface.

	Filterable α, ρCi/ℓ	Particulate α, ρCi/ℓ	Filterable β, ρCi/ℓ	Particulate β, ρCi/ℓ	Dosimetry, mr/wk
Gable Pond	<4.0 - 170	<2.0 - 145	36 - 140	<1.7 - 1,740	143 - 633
B-Pond	<1.1 - 322	<2.1 - 4.3	<30 - 190	7.8 - 111	2.8 - 8.3
U-Pond	<4.5 - 522	<1.5 - 59	<26 - 8,100	<17 - 5,110	183 - 229
West Pond	<13 - 220	<0.8 - 64	<89 - 14,700	<2.6 - 1,740	≤bkg - 3.1
A-29 Ditch	<1.1 - 100	<2.6 - 189	<26 - 773	<16 - 662	≤bkg - 0.3
B-3 Ditch	<1.0 - 15	<2.1 - 51	<37 - 161	<16 - 649	≤bkg - 6.1
Z-19 Ditch	<5.6 - 26,900	<0.1 - 141,000	<37 - 2,310	<1.9 - 3,440	—
100-N Trench	22 - 860[a]		<1,900 - 5,700,000[b]		357,000 - 743,750

[a] Total ± measurements
[b] Total ± measurements

TABLE 2.10. Relative Abundance of Algae Observed in Ponds and Streams on the Hanford Reservation. The letters L (low), M (medium) and H (high) indicate a qualitative spectrum of appearance of these organisms.

Division (Common Name)	Family	Gable Pond	B-Pond	U-Pond	West Pond	A-29 Ditch	B-3 Ditch	Z-19 Ditch	100-N Trench
Cyanaphyta (Blue-green Algae)	Oscillatoriaceae	L	L	M	L	M	M	M	M
	Chroococcaceae	L	L	L	L	L	L		
	Nostocaceae	L	L	M	L	L	L	L	L
Chlorophyta (Green Algae)	Desmidiaceae	M	L	L	L	L	L	L	L
	Palmellaceae		L	L		L			
	Volvocaceae		L						
	Characiaceae	L	L	M					
	Phacotaceae	L	L		L	L	L		
	Oöcystaceae	M	L	L	L	M	M	M	L
	Scenedesmaceae	L		L	L	M	L	L	
	Tetrasporaceae			H					
	Micratiniaceae						L		
	Coccomyxaceae						L		
	Zygnemataceae	M		M		H		H	H
	Cladophoraceae	M	M	H	L	M		M	L
	Hydrodictyaceae	L		M				M	H
	Characeae	L							
Euglenophyta (Euglenoid Algae)	Euglenaceae			M	L				
Chrysophyta (Golden-brown Algae and Diatoms)	Mallomonadaceae					L			
	Ochromonadaceae						L		
	Coscinodiscaceae	H	L	L	M	M	H	M	M
	Rhizosoleniaceae				L				
	Tabellariaceae	L					L		
	Diatomaceae	L			L		L		
	Achnanthaceae	M	L	L	L	L	L		
	Naviculaceae	M	M	M	H	L	M	M	L
	Gomphonemataceae	L	M	M	L	L	L		
	Cymbellaceae	M	H	L	M	H	M	M	M
	Nitzschiaceae	L	L	L		L	L		
	Surirellaceae	L	L	L	L		L		
	Fragilariaceae	M	M	M	M	H	H	M	M
	Synuraceae				L				
Pyrrophyta (Dinoflagellates)	Gymnodiniaceae				L				
	Glenodiniaceae			L	M				
Cryptophyceae (Cryptomonades)	Cryptomonadaceae		M	M	M	L	M		

TABLE 2.11. Dominant Macrophytes Observed in Ponds and Streams on the Hanford Reservation. The letters L (low), M (medium) and H (high) indicate a qualitative spectrum of appearance of these plants.

Common Name	Genus	Gable Pond	B-Pond	U-Pond	West Pond	A-29 Ditch	B-3 Ditch	Z-19 Ditch	100-N Trench
Horsetail	*Equisetum*	L		M					
Pondweed	*Potamogeton*	H	M	H				H	
Hornwort	*Ceratophyllum*	M							
Water Milfoil	*Myriophyllum*	M							
Duckweed	*Lemna*			M					
Cattail	*Typha*	M	M	H	M		M	H	
Bulrush	*Scirpus*	M	M	M	M	M	H	M	L
Smartweed	*Polygonum*			L					
Speedwell	*Veronica*					L	M	L	L
Watercress	*Rorippa*			L				L	
Wild Lettuce	*Lactuca*			L				L	
Willow	*Salix*			L					
Cottonwood	*Populus*			L				L	

TABLE 2.12. Relative Abundance of Aquatic Fauna Observed in Ponds and Streams on the Hanford Reservation. The letters L (low), M (medium) and H (high) indicate a qualitative spectrum of appearance of these organisms.

Common	Taxon	Gable Pond	B-Pond	U-Pond	West Pond	A-29 Ditch	B-3 Ditch	Z-19 Ditch	100-N Trench
Flatworm	*Dugesia*	L	L	L		L	L		
Leech	Hirudinea		L	L					
Segmented Worm	Oligochaeta	M	L	L	H	L	L	L	L
Waterflea	*Daphnia*	H	L	H					
Seed Shrimp	Ostracoda			L					
Scud	*Hyallella*	H	H	M			L		
Watermite	Hydrocarina			M					
Mayfly	Baetidae	M	M	L					
Dragonfly	*Aeschna*	H	M	H			L		L
Dragonfly	*Libellula*	M	L	H					
Dragonfly	*Tramea*			M					
Dragonfly	*Erythemis*			L					
Dragonfly	*Anax*	L	L						
Damselfly	*Ischnura*	M	M	H	L		L	L	L
Waterstrider	Gerridae	L	L	L		L	L	L	
Backswimmer	Notonectidae	L	L	H	L			L	L
Creeping Water Bug	Naucoridae				L				
Water Scorpion	Nepidae	L		L					
Giant Water Bug	Belostomatidae	L		M					
Water Boatman	Corixidae	M	H	H	L			M	
Caddis fly	Tricoptera	L	L	L	M		M		
Predaceous Diving Beetle	Dytiscidae	L	L	L			L	L	
Water Scavenger Beetle	Hydrophilidae	L	L						
Crawling Water Beetle	Halipidae	L							
Beetle	Amphizoidae			L					
Beetle	Noteridae			L					
Beetle	Helodidae			L					
Midge	Chironomidae	M	L	M	M	H	H	M	H
Black fly	Simuliidae					L			
Shore fly	Ephydridae				M				
Aquatic Caterpiller	Nymphulidae		L				L		
Snail	*Physa*	L	M	M					
Snail	*Lymnaea*	H		H					H
Snail	Planorbidae	L	L						
Goldfish	*Carassius*	M	L	H			L		

TABLE 2.13. Quantitative Biological Characteristics for Ponds and Streams on the Hanford Reservation. Maximum productivity rates express highest rates observed for each pond. Means are shown with 95% confidence values (i.e., mean 95% C.V. = 95% confidence interval about the mean).

	Gable Pond	B-Pond	U-Pond	West Pond	A-29 Ditch	B-3 Ditch	Z-19 Ditch	100-N Trench
Pond-wide Primary Productivity (kg C/hectare per day)	—	—	42	—	—	—	—	—
Maximum Periphyton primary productivity(a) (μg chl α/cm^2 per day)	7.4	38.5	45.2	42.1	48.9	33.0	—	—
Maximum *Aufwuchs* Productivity(b) (μg C/cm^2 per mo)	156	105	69	180	607	657	—	—
Colonization Rate (No. organisms/m^2 per day)	1020 ± 1440	777 ± 982	417 ± 325	188 ± 176	2750 ± 2180	1610 ± 910	—	897 ± 1513
Goldfish productivity (kg/hectare per year)	—	—	37	—				
Diversity Indices:								
N (No. indiv. in 4 m^2)	245 ± 130	295 ± 260	—	168 ± 74	—	5964 ± 1779	—	1578 ± 4213
H (comm. div. per indiv.)	2.1 ± 0.5	1.7 ± 0.6	—	1.2 ± 0.5	—	0.1 ± 0.03		0.9 ± 0.6
J (evenness)	0.6 ± 0.2	0.6 ± 0.2	—	0.5 ± 0.2	—	0.04 ± 0.10	—	0.5 ± 0.3

(a) Highest rate of chl α accumulation occurring within a 4-wk test period
(b) Highest rate of *Aufwuchs* C accumulation occurring over several 1-3 mo test periods

on the burial grounds and control plot. Captured mice were given an individual identification number using a series of toe amputations. The animals were then anesthetized and small dosimeters were surgically implanted subcutaneously on the dorsal side of their necks.

To date there have been a total of 60 dosimeters recovered from these four plots. Deer mice had only six returns, all which came from the 300-6 and 7 burial ground. Results are presented in Table 2.14. Pocket mice captured on the burial grounds received the same doses as those on the control. The deer mice captured on the 300-6 and 7 burial ground showed doses above background ranging from 9.2 - 237 mR/wk. These higher doses may possibly be explained by the past presence of a "cave-in" on the burial ground. All six deer mice were captured in the vicinity of the "cave-in."

Future trapping sessions will be necessary to determine if the exposure potential has been alleviated. It appears that the protective backfill of gravel and cobblestone is an effective barrier to pocket mice keeping them from burrowing next to the buried waste.

Nesting Ecology of the Burrowing Owl on the Hanford Site

The burrowing owl is one of the most abundant birds of prey nesting on the Hanford Site. Little, however, is known about its life history in southeastern Washington.

In 1975, we began a study of the nesting ecology of the species. Data were collected on owl nesting densities, productivity, food habits, habitat utilization, and prey species abundance. Regurgitated castings (pellets)

TABLE 2.14. Mean Gamma Doses to Mice on 300 Area Burial Grounds in milliroentgens per Week, mR.

	Pocket Mouse				Deer Mouse			
	n	x	S.E.	Range	n	x	S.E.	Range
Control	16	1.2	±0.14	0.6 - 2.2				
WYE B.G.[a]	21	1.1	±0.10	0.4 - 1.8				
300 North B.G.[a]	8	1.8	±0.09	1.7 - 2.2				
300 6-7 B.G.[a]	9	1.8	±0.11	1.5 - 2.6	6	93.4	±33.5	9.2 - 237

[a]Burial ground

were collected weekly at each of four nest sites. Data on insect densities (major food of burrowing owls) were gathered simultaneously in areas adjacent to each owl nesting burrow.

We are presently analyzing samples and will be examining prey availability and its effects on prey utilization. Data on productivity and nest site selection have also been collected and will be correlated with habitats utilized by the owls and other parameters which could influence productivity.

- **Radioecology of ^{129}I and ^{99}Tc**

- **Influence of Soils and Aquatic Sediments on the Chemical Behavior, Transport, and Bioavailability of Pollutants**

Principal Investigators: T. R. Garland, R. G. Schreckhise, R. E. Wildung, D. A. Cataldo, K. M. McFadden, C. L. Wilkerson, J. F. Cline, D. W. Uresk and E. L. Klepper

The terrestrial aspects of these investigations, involving close cooperation between soil and plant scientists, are directed toward identification of soil physicochemical factors and plant kinetic and metabolic factors governing the availability to plants of nuclear and non-nuclear wastes. Principal recent emphasis has been placed on ^{99}Tc and ^{129}I. Laboratory and field studies have been initiated to define key processes governing their behavior.

The long-term environmental behavior and ultimate fate of ^{129}I cannot be inferred with confidence from the well-studied short-lived isotope ^{131}I. Furthermore ^{99}Tc, it now appears, does not resemble iodine in its soil/plant behavior, as has been assumed in the past. Therefore, its long-term behavior and ultimate fate remains uncertain. The quantitative measurement of ^{129}I and ^{99}Tc in environmental samples is also technically complex and costly. The ^{129}I can be measured in a variety of low-level environmental samples using neutron activation procedures developed at PNL. The ^{99}Tc still presents difficulties, but a sensitive mass spectrometric approach has been under development for environmental application.

In aquatic sediment studies, investigations have been directed toward measurement of the physicochemical properties of suspended and bottom sediments of the lower Columbia River watershed and radionuclide (^{99}Tc, ^{129}I) and trace metal speciation, transport and biological availability as influenced by sediment properties and seasonal and man-induced changes in watershed and river conditions. Limited efforts were directed toward these studies during the reporting period.

The soil and sediment studies closely interdigitate with several investigations dealing with the fate of radionuclides in associated terrestrial and aquatic studies.

Ingestion of Radioiodine by Range Cattle

Four steers were pastured on a native bluebunch wheatgrass pasture in spring 1976. Two steers had been fitted with esophageal fistulas and two with rumen fistulas. Fistulas permitted direct measurement of dietary composition and forage digestibility. Total forage intake, calculated from water consumption rates, was proportioned by weight into species to estimate iodine intake rates. Salt intake was determined from the average weight loss from salt blocks supplied to the steers. Iodine-127 and ^{129}I concentrations in the various dietary components were determined by neutron activation.

Table 2.15 gives the average dietary composition and amounts of ^{129}I and ^{127}I ingested by the three steers kept on the pasture for 46 days (one steer was kept only 20 days). Forage supplied over 97% of the ^{129}I ingested and only 28% of the ^{127}I. Salt was the major source of ^{127}I (66%). The ratio of ^{129}I to ^{127}I was higher in forage because of the ^{129}I present in surface soils as a result of airborne nuclear fuel effluents from the Hanford plant and from worldwide fallout.

TABLE 2.15. Dietary Composition and Iodine Intake for Range Steers on the ALE Pastures from April 30 to June 15, 1976.

Component	Consumption per Head	^{129}I Intake, pCi	^{127}I Intake, mg
Water	1208 Liters	0.06	7.5
Salt	1873 g	2.81	78.7
Bluebunch wheatgrass	878 kg	28.99	8.8
Cusick poa	88.5 kg	33.62	7.8
Stipa thurberiana	61.1 kg	28.72	6.3
Other grasses	12.5 kg	4.89	1.2
Crepis atrabarba	2.4 kg	0.38	0.3
Other forbs	74.3 kg	11.89	9.1
Total	—	111.35	119.7

Behavior of Tc in Soils

Soil and plant factors influencing Tc availability are currently under study. These include: 1) mobility and chemical form of Tc in soils, 2) possible conversion of Tc due to microbial activity, and 3) correlation of soil properties to Tc solubility and availability to plants. A 24-hr aerobic study of Tc mobility in 22 soils conducted in 1974 showed Tc to be highly mobile, sorbing significantly only in high organic, low pH soils. In contrast, a study of 11 soils under anaerobic conditions at the University of Minnesota showed nearly quantitative sorption of Tc, though a 2-5 week period was required for completion of the process. Since this raised the question of equilibrium attainment, further studies were conducted.

A set of 32 soils of varying soil properties was used. These soils encompass the ranges in soil properties of those soils used in the Minnesota study. The soils have been extensively characterized for elemental content, particle size distribution, mineralogy, and extractable components.

Soils were mixed in a 1:10 ratio with distilled water containing 0.01μ Ci ^{99m}Tc/ml, and sorption followed over a 6 week period under aerobic conditions. Soils generally showed an initial rapid sorption followed by a leveling or decrease in all but typically high organic content soils. After several weeks, a slight increase in sorption was sometimes observed, but in no case was the Tc quantitatively sorbed. Most of the apparent K_d values remained constant, but in one case showed continual increase in sorption with time during the 3-week interval. After 9 weeks of shaking, several flasks were sealed to promote anaerobic conditions. Only one of four soils showed significant increases in sorption after 3 weeks under these conditions, and this sorption was far from complete.

A more quantitative test consisted of soils (similar soil to solution ratio) analyzed after 3, 12 and 26 days of aerobic mixing. K_d values calculated from the 3-day equilibration were in the range of 1-4 ml/g with the exception of a peat which showed a K_d of 19. With increased time, about two-thirds of the soils showed some increase in sorption, while four showed decreased sorption. In the 24-hr study, K_d values generally were higher than, but correlated well with, the K_d values determined. In that study, the aqueous fraction was separated by centrifugation, which would not have fully separated the colloidal clay fractions if significant dispersion occurred, perhaps resulting in lower apparent adsorption.

Results indicate that equilibrium sorption was probably established in 1-3 days, after which processes other than true adsorption begin to participate. Microbial activity or reductive precipitation may be involved. Some of the statistically significant linear

correlations which have been calculated are given in Table 2.16. R-values listed indicate that correlation is expected, but the actual relationships may not be linear. In some instances, soils appeared to group into two linear patterns rather than a single linear correlation. Further analysis of the data is in progress.

TABLE 2.16. Correlations Between Tc Sorption and Soil Properties, Statistically Significant at the 0.01 Level

Soil Property	Number of Determinations	R-Values vs Kd, TcO_4	R-Values vs % TcO_4 Sorption
% Organic carbon	25	0.543	0.555
% Nitrogen	25	0.574	0.583
C/N Ratio	25	0.506	0.534
% Moisture	27	0.644	0.655
Soil pH	23	-0.543	-0.559
Cation exchg capacity	23	(No)	0.530
% Fine clay	25	0.513	0.539
% Coarse clay	25	0.706	0.704
% Very fine sand	25	-0.565	-0.574
Extractible Fe:			
Organic	27	0.678	0.648
Inorganic	27	0.631	0.592
Oxides	27	0.739	0.721
Extractible Al:			
Organic	27	0.642	0.606
Inorganic	27	0.668	0.639
Oxides	27	0.608	0.579

Future studies will include analysis of the effect of microbial activity on sorption; determination of the effect of an added carbon source on sorption, and research to establish specific sorption mechanisms. The uptake of Tc by plants grown on these soils will be studied for correlations with soil properties, in an attempt to predict uptake from other soils based on their physicochemical properties.

Influence of Soil Properties on Tc Uptake by Plants

As noted above, previous studies have shown that Tc is poorly sorbed by soils and significant sorption occurs only in soils that are high in organic matter and low in pH. Additional correlations with measured soil properties have indicated that sorption is closely related to the oxalate extractable Fe and Al. This information, in addition to the extremely high uptake of Tc by plants from most soils, has allowed studies to be initiated with a range of soils which have varying degrees of sorptive capacity for Tc. The results of these studies will allow a further definition between soil properties, Tc adsorption behavior, and plant availability. The properties of soils chosen for the study are listed in Table 2.17.

Previous studies at PNL have shown Tc to be readily accumulated by plants grown both in solution culture and in soils, with phytotoxicity being apparent at soil concentrations in excess of 0.1 ppm. Investigations have been continuing along several lines. These include: 1) evaluation of absorption kinetics, 2) effects of nutrilites on TcO_4^- absorption, and 3) mechanisms of TcO_4^- toxicity in plants.

Kinetic analysis of the absorption isotherm over a concentration of 0.1 to 0.8 μM (∿10-80 ppb) gives a K_m of 2.8 μM and a V_{max} of 18.3 μg Tc/g dry wt root • hr. Currently, $H_2PO_4^-$, Mn^{++}, $MoO_4^=$, $SO_4^=$, IO_4^-, and ReO_4^- can be shown to have an inhibitory effect on TcO_4^- absorption. Detailed competition kinetics have been hampered by a complex interaction of nutrient anions with root metabolism. This appears to be a problem inherent in analysis of the uptake kinetics of anions.

Technetium uptake by 40-day old soybean plants from either 0.5 mM $CaCl_2$ or complete nutrient solutions containing 0.03 ppt $^{95m}TcO_4^-$ was followed with time (Figure 2.6). In the presence of no competing ions, Tc was rapidly removed from solutions surrounding the root, with complete depletion after 72 hr. The presence of a complex mixture of nutrient ions had an inhibitory effect of Tc uptake, but depletion was still rapid and nearly complete by 72 hr. These data support competition data obtained for Tc and coions of higher concentrations and suggest that Tc accumulation occurs by virtue of its analog behavior to one or more nutrient species. Nonspecific adsorption at these low concentrations has been ruled out by previous studies and recent autoradiographic studies. The latter show detectable Tc in internal root tissues within 5 min and in stem and leaf tissue within 15 min.

TABLE 2.17. Properties of Soils Selected for Tc Uptake Studies

Characterization	Soils 1	2	3	4	5	6	7
Soil pH	4.7	4.2	4.9	5.7	5.4	5.8	6.2
% Organic C	4.11	2.35	2.93	4.73	1.93	0.76	0.50
% Fine Clay	12.8	7.70	5.70	8.7	12.9	4.8	5.7
Dithionite Fe μg/g	2740	1000	1970	730	1700	1570	1430
Pyrophosphate Fe μg/g	2040	760	690	360	540	70	80
Oxalate Fe μg/g	4370	2180	3230	1610	1300	606	548
Dithionite Al μg/g	500	290	1420	200	250	140	90
Pyrophosphate Al μg/g	1440	820	3440	500	660	180	100
Oxalate Al μg/g	1430	1200	6910	672	732	421	369
Hydroxylamine Mn	830	2320	540	1080	540	460	320
KOH Si_1O_2 μg/g	3150	2400	2650	1310	2080	2800	2420
KOH Al_2O_3 μg/g	1100	590	2060	250	660	220	250
S μg/g	393	304	371	619	444	446	130
F μg/g	37	66	28	127	218	253	216
Cl μg/g	89	66	111	55	29	54	51
I μg/g	2.2	1.0	3.2	1.6	6.1	1.7	0.56

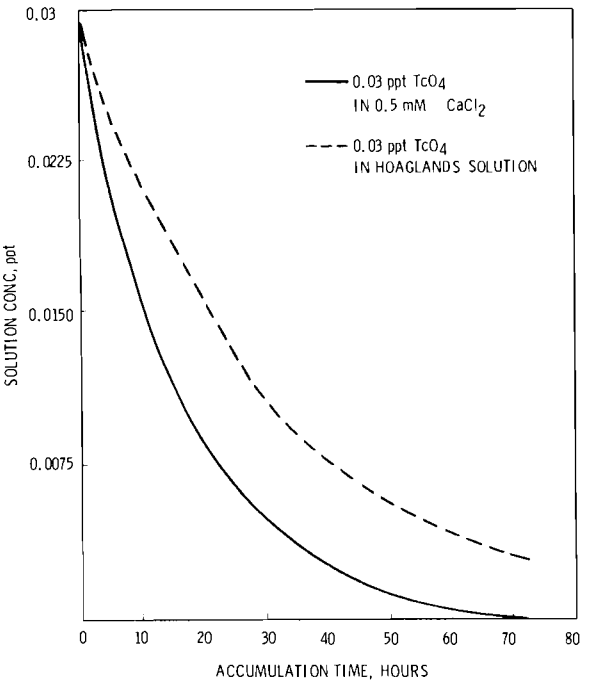


FIGURE 2.6. Depletion of TcO_4 from Hydroponic Solutions, in the Presence and Absence of Nutrient Ions, by Soybean Plants

A number of approaches have been attempted to resolve the observed toxicity of Tc. The first approach was to determine whether Tc toxicity could be a radiological effect since meristems were the first plant tissues to exhibit a toxicity response. Although the total tissue dose was insufficient to induce the observed effect, accumulation of Tc in proximity to genetic material could result in a localized increase in dose and therefore produce the observed toxicity. Autoradiography of meristem thin sections failed to show any disproportionate localization or concentration of Tc in nuclear material. Similarly, observation of mitotic behavior of root and shoot meristems failed to show chromosomal aberrations, bridges, or formation of micronuclei indicative of radiation damage. Since radiation damage can most likely be ruled out, investigations are continuing on the basis of chemical toxicity. These studies, which are partially completed, include analysis of dry matter production and gross metabolic processes such as respiration, photosynthesis, and nitrogen metabolism.

Field Studies

Parallel with development of a highly sensitive mass spectrometric method for Tc

determination in environmental samples by the Physical Sciences Department was the collection of samples in the vicinity of the Hanford reprocessing plant. Although active fuel reprocessing at Hanford halted in 1972, Tc might be expected around the aqueous waste ponds or downwind from reprocessing plant stacks.

Sampling was done within the Hanford 200 West Area boundaries. The first set of samples was collected northeast of U Pond, and consisted of tree, shrubs, grasses, and soils. Portions of a cottonwood tree growing on the banks of U Pond were collected, as were vegetation and soil from an undisturbed treeless field east of U Pond and downwind from the U plant stack.

Gamma-scan analysis of these samples revealed the presence of radionuclides, some of which are listed in Table 2.17. Aliquots of these samples will be analyzed for ^{129}I and natural I in addition to ^{99}Tc.

TABLE 2.18. Levels of Selected Nuclides in Samples in the Vicinity of a Reprocessing Plant (pCi/g dry wt)

	^{137}Cs	^{235}U	^{40}K	^{60}Co	^{226}Ra	^{228}Ra	^{238}Th	^{234}Th
NE of U Pond:								
Asparagus	24	0.10	18					
Canadian Thistle	89		39	0.96				
Willow Leaves	160	0.17	12	0.75				
Soil:								
Litter	1200	0.96	5.4	0.19				
0-5 cm	2200		23	0.11	0.95	2.2	1.3	
6-15 cm	19	0.23	20	0.017	1.0	1.0	0.98	0.96
16-25 cm	5.7	0.28	22		1.1	1.2	1.1	4.7
26-35 cm	8.2	0.19	19	0.011	0.95	0.92	0.98	
At U Pond:								
Cottonwood Leaves	160	0.11	14	0.77				
Bark	87	0.08			0.37			
Wood	39	0.083	2.8		0.21			
E of U Pond:								
Jim Hill Mustard	0.1		15		0.048			
Sagebrush Twigs	3.4	0.13	11		0.22			6.7
Leaves	2.0	0.43	17		0.22		0.079	
Soil:								
Litter	18	0.18	7.7	0.064	0.81	1.2	0.44	
0-5 cm	0.33	0.13	17		0.87	0.83	0.94	3.8
6-15 cm		0.14	16		0.96	1.0	0.92	4.4
16-25 cm		0.14	22		1.3	1.2	1.3	
26-35 cm	0.14	0.14	23		1.2	1.1	1.1	

The determination of ^{99}Tc in environmental samples using the mass spectrometric method is extremely time consuming. An alfalfa sample from a field 30 miles downwind from the reprocessing plant has been taken through the entire procedure, but final results await analysis of a reagent blanks and recovery values. As a means of providing the most realistic vegetation test sample for recovery values, hydroponically grown soybeans were exposed to ^{95}Tc as TcO_4^- in solution. Uptake was essentially complete in 3 days, with a shoot to root ratio of about 2:1. These samples are being used in recovery studies.

Additional studies aimed at shortening or bypassing long or difficult steps in the chemical separation procedure are planned. The feasibility of replacing a destructive fusion step by an extraction step is one possibility, particularly for vegetation samples. Use of beta counting techniques in conjunction with mass spectrometric measurement of higher level samples will also be investigated.

Aquatic Studies

Several Tc isotopes are produced in sizeable quantities during nuclear fission. Of these, ^{99}Tc has the longest half-life (2.1×10^5 yr) and is considered of utmost importance due to its potential for dose contribution to man. Entrance to the environment can occur during processing of spent nuclear fuels, or in disposal of aqueous wastes from nuclear, hospital or other facilities.

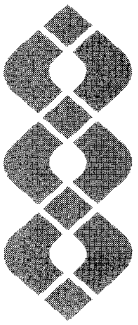
The Tc can exist in any valence state from -1 to +7, with +4 and +7 the most stable. The pertechnetate ion (TcO_4^-) is highly stable in aqueous systems and would be expected to migrate with the groundwater. Sorption of TcO_4^- by diverse soil types has been shown to be generally low.

If Tc retains its mobility, it would be expected to enter river systems and ultimately be carried to the oceans. Due to the difficulty of detecting ^{99}Tc at environmental levels, no value has been reported for the concentration of ^{99}Tc in ocean waters, although ^{99}Tc as pertechnetate has been shown to sorb from seawater onto bottom deposits. The sorbed Tc was easily and completely desorbed by washing with seawater. Tc uptake was marked in marine algae and seaweed, and showed a 60-day biological half-life in red abalone. Such studies would predict a high biological availability of Tc.

Based on a measured level of ^{137}Cs in ocean water of 0.021pg/kg, and assuming the contribution is due entirely to fallout, ^{99}Tc due to fallout would then be about 1.5×10^{-3} pg/ℓ. If a fuels reprocessing plant is assumed to contribute an order of magnitude higher level of ^{99}Tc, then a 100 ℓ sample size would be sufficient for detection by the mass spectrometric techniques under development.

A method for sampling such a volume of seawater is under study. Simple anion exchange using AGL-X4(NO_3^- form), 50-100 mesh, showed retention of TcO_4^- from about 750 mℓ seawater/mℓ resin before loss in the eluent reached 10% of the concentration in the seawater initially (or a total sample loss of about 5%). Replacing the resin with 100-200 mesh resulted in slightly increased retention. TcO_4^- equlibrated for 2 weeks in seawater showed no loss in recoverability using this technique. Optimum conditions of column dimensions at the flow rate required for practical sampling are being studied.

After refining the ion exchange method for analysis of Tc in seawater, surface seawater samples will be collected along the Washington and Oregon coasts, where the Tc contribution from fuel reprocessing at Hanford is postulated to be deposited via the Columbia River. In conjunction with the water study analyses of suspended matter, plankton, and seaweeds will be conducted. All these studies require the high sensitivity of the mass spectrometric method.



3.0 Nuclear Fusion

NUCLEAR FUSION

- **Sublethal Effects of Tritium on Aquatic Systems**
- **Effects of Low-Level Chronic Irradiation on Embryonic Development**
- **Effects of Beryllium and Lithium on Aquatic Systems**
- o **Teratogenic Effects of Low-Level Magnetic Fields**

It is conceivable, and possible, that with increasing applications of nuclear energy, greater quantities of potentially harmful radionuclides will be released to the environment. This may be particularly true for advanced reactor designs such as the fusion reactor where radionuclides, principally tritium, may be expected to escape from the plant both in gaseous and liquid effluents in quantities significantly greater than for present PWR or BWR designs. Foreseeing such contingencies, the research programs described herein are in response to the need to measure the potential radiation effects of tritium releases on individuals, and ultimately populations and biotic communities. As a first approach, our efforts are directed to determine effects of low-level chronic exposures on developing embryo and larval stages, clearly the most radiosensitive.

The anticipated increase in the release of beryllium and lithium from mining, refining, and fabrication of materials used during construction of fusion reactors has also caused concern as to potential adverse effects on the environment. Accordingly, FY-78 fusion related research will include efforts to study the metabolism of each metal in the living organism, and to determine at what levels toxicity may be expected. Again, the effects of beryllium and lithium will be assessed in embryo and larval stages of selected aquatic organisms, with ultimate interest in the response of populations and communities.

Our fusion related research in FY-78 will also include preliminary experiments on the effects of low-level magnetic fields. It is conceivable that magnetic fields of 70-450 gauss will be encountered by attendant personnel working in the transport and hot cell areas of fusion reactors. Also, those personnel assigned to the areas immediately surrounding the reactor may be exposed for substantial durations to

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o Work order agreement

field strengths of 1-70 gauss. The current PNL program is aimed at developing different measurement end-points for the early detection of developmental and adult chronic effects. Work involving mammalian tissue and cell systems is reported in PNL-2500, Part 1, Biomedical Sciences; work involving nonmammalian animal systems is reported here. Dr. Dennis Mahlum is coordinator for the comprehensive program. The approaches described here will utilize the embryo and larval stages of aquatic organisms which should provide meaningful data applicable to ultimate assessment of effects in life processes of higher vertebrates.

NUCLEAR FUSION

- **Sublethal Effects of Tritium on Aquatic Systems**
- **Effects of Low-Level Chronic Irradiation on Embryonic Development**
- **Effects of Beryllium and Lithium on Aquatic Systems**
- **Teratogenic Effects of Low-Level Magnetic Fields**

Principal Investigators: J. A. Strand, G. Roesijadi and R. M. Emery

Other Investigators: M. P. Fujihara and J. C. Montgomery

Technical Assistants: C. S. Abernethy, W. G. Woodfield and D. C. Klopfer

Continuing studies of the sublethal effects of tritium on freshwater species emphasize the potential for genetic transmission of suppressed immune competence in offspring of parental rainbow trout, Salmo gairdneri, exposed to tritium (0, 0.04, 0.4, 4.0, 40.0 rads) over embryogenesis. It is planned that the offspring of recently conducted test crosses will be assayed for agglutinating antibodies in response to the bacterial antigen, Flexibacter columnaris, and decreased levels of circulating antibody indicating the potential for transmission of altered genetic material mediating immune competence.

Continuing studies with marine species, Pinnixa occidentalis, Hemigrapsus nudus, and Cragnon sp., examine uptake and release of tritium over varying life stages and evaluate radiation effects on development and survival.

In another study to begin in FY-78, interest is focused on the potential effects of beryllium and lithium on aquatic systems. Increased levels of each metal are likely to be encountered in surface waters due to mining and refining subsequent to their use in the construction of fusion reactors. Planned studies include an evaluation of potential toxicity of beryllium and lithium on embryological life stages of rainbow trout, Salmo gairdneri, and an assessment of fate and effects in artificial stream habitats.

Studies to be initiated in FY-78 include an evaluation of the effects of low-level magnetic fields on embryologic development of rainbow trout, Salmo gairdneri. It is the objective of these studies to provide data which will have application to assessment of potentially harmful

effects of low-level magnetic fields encountered by attendant personnel working in the transport and hot cell areas of fusion reactors. This approach is less costly, provides large numbers of experimental organisms for meaningful statistical analysis, and permits examination of potential late effects in a representative vertebrate.

Sublethal Effects of Tritium on Aquatic Systems

Present studies are directed to determine the potential for genetic transmission of suppressed immune competence in offspring of parental rainbow trout, Salmo gairdneri, sublethally exposed to tritium as tritium oxide (HTO) (0, 0.04, 0.4, 4.0, and 40.0 rads) during embryogenesis. Previous studies demonstrated that the primary immune response of parental stocks at 5 and 17 months of age was significantly suppressed and permanently altered at doses as low as 4.0 rads.

As the race of experimental fish presently reared in the Hanford Hatchery did not mature until winter of their 3rd yr of growth, offspring of test crosses conducted in February-March 1977 will not be of sufficient size to assay for diminished immune competence until November-December 1977.

However, it is planned that the offspring of each test cross will be compared as parental stocks were in previous studies; that is, for agglutinating antibodies in response to the bacterial antigen, Flexibacter columnaris, as detected by agglutinin assay. Decreased levels of circulating antibody in the serum of offspring from tritium irradiated stocks will serve to indicate the potential for transmission of altered genetic material controlling or mediating immune competence.

Challenge techniques employing virulent strains of the pathogen, F. columnaris, will also be applied in this context. The procedure as adopted for such subsequent experiments will involve the injection of 0.1 cc of a standard suspension and tenfold dilutions of the pathogen into separate lots of 10 or 20 fish each from the respective test crosses. Mortality will be recorded for 5 consecutive days and the LD_{50}, or the dilution at which 50% mortality occurs will be calculated. If permanent genetic alteration has occurred, it may be expected that significantly lower LD_{50} endpoints will result when the offspring of tritium irradiated parental stocks are tested.

Effects of Low-Level Chronic Irradiation on Embryonic Development of Marine Fish and Invertebrates

During FY-77, efforts were centered on the determination of HTO turnover rates in young postlarval stages or eggs of three marine crustaceans. Additionally, larvae of two species of commercial shrimp were reared in low levels of tritiated seawater.

Measurements of HTO turnover rates were determined in the megalopa and crab stages I to V of Pinnixa occidentalis, a brachyuran crab. Mean values for rate constants and half-times were 3.27 ± 0.40 ($\bar{x}$ ± S.E.) hr and 0.28 ± 0.04 hr, respectively. Differences in rates of HTO turnover for different stages were not statistically significant. Therefore, body size or life stage apparently did not influence HTO turnover rates in this crab.

Rate constants and half-times for HTO turnover in the eggs of the crab Hemigrapsus nudus were 0.32 ± 0.02 hr and 2.20 ± 0.10 hr, respectively; values for the eggs of shrimp Crangon sp. were 7.58 ± 2.99 and 0.20 ± 0.06 for rate constants and half-times, respectively. These data indicate that HTO turnover was very rapid in Crangon sp. compared to H. nudus and were reflective of the habitats of the two species. H. nudus exists in the intertidal environment and is frequently exposed to the atmosphere. Crangon sp., on the other hand, is a subtidal species which inhabits a relatively stable marine environment.

Larvae of the shrimps Pandalus platyceros and P. danae were reared in seawater containing 3×10^{-4} and 3×10^{-3} μCi/ul tritium as HTO. Survival, molting frequency, and duration of larval development were similar in control and tritium-exposed individuals of both species.

Effects of Beryllium and Lithium on Aquatic Systems

The anticipated increase in the release of beryllium and lithium from mining, refining, and fabrication of materials used

during construction of fusion reactors has caused concern as to potential effects on the environment. In order to assess and predict with assurance the effects of beryllium and lithium released to the environment, it is necessary to know how readily each metal is metabolized by living organisms, and at what levels toxicity may be expected. Entry of these metals, particularly beryllium, into the food chain with biomagnification may present a serious hazard to man. However, few studies relevant to these needs are providing the results necessary to make an environmental assessment.

Beginning in FY-78, we plan to identify and measure the acute and chronic effects of lithium and beryllium compounds on specific aquatic organisms and associated freshwater communities. Since this study is designed to reflect actual perturbations caused by the accidental release of these metals into freshwater ecosystems, it differs from a standard toxic bioassay in that the observed and measured effects will be viewed under ecologically dynamic conditions. Thus, the expressions of response will be more relevant to the assessment of environmental impact associated with mining, refining, and fusion plant operation.

Summary of Study Plan

Task I. Toxicity. In initial experiments to establish acute (96 hr) and chronic (30 days) effects, eggs and embryos of rainbow trout will be exposed to varying concentrations of beryllium and lithium. Injury will be assessed by the proportion of embryos failing to reach larval stage, and by the proportions of morphologically abnormal embryos (relative to total reaching larval stage). For the latter, anomalies scored will include major malformations of head and body. Changes in rate of embryonic development will also be quantitated for definite, recognizable developmental stages. Metal accumulation will be studied by periodic removal and analysis of embryos and larvae. Turnover rates will be estimated by transfer of embryos and larvae to single-pass incubation systems where similar measurements will be conducted.

As time provides over the 1st yr, additional exposures will be conducted over different stages of development to determine relative sensitivities. As well, exposures of varying duration will be tested. The metals beryllium and lithium will be added to the exposure medium in their respective ore forms, or as oxides, hydroxides, fluorides, or other salts as appropriate.

Task II. Community Response. During the initial period of this study, artificial streams will be developed in hatchery troughs to resemble a low velocity habitat of the Columbia River. Flow rates of unfiltered Columbia River water passing through the troughs will be regulated to permit optimal growth for periphyton, benthic invertebrates, and certain fishes. These experiments will be performed outside, under natural thermal and solar regimes, during spring, summer, and fall seasons. These streams will be arranged in a parallel fashion to permit nearly identical conditions in experimental and control systems. Artificial streams will undergo natural colonization, however, fishes will be added to enhance the trophic structure of the community. By late spring, sufficient colonization will be attained to begin experimental additions of lithium and beryllium compounds. The effects of these compounds in the microcosms will be detected and measured in terms of significant changes in:

- community diversity,
- biomass,
- primary production of periphyton communities,
- photosynthesis (respiration ratios),
- rates of recolonization on bare substrates,
- survival and growth of specific organisms,
- seasonal periodicity of species occurrences as reflected by the control streams.

The compounds of greatest interest in this work will be the fluorides, oxides, and hydroxides of lithium and beryllium. Concentrations of these compounds will be measured and maintained in experimental systems by direct sampling and continuous delivery procedures. Ranges of concentrations for testing will be specifically determined by preliminary toxicity bioassays and toxicological information gathered from the literature.

Teratogenic Effects of Low-Level Magnetic Fields

Development of magnetic fusion reactors will result in occupational exposure of personnel to varying strengths and geometries. It is conceivable that magnetic fields of 70-450 gauss will be encountered by attendant personnel working in the transport and hot cell areas of the reactor. Also, those personnel assigned to the areas immediately

surrounding the reactor may be exposed for substantial durations to field strengths of 1-70 gauss.

Working in cooperation with the Biology Department, our study objective is to determine the potential effects of low-level magnetic fields on sensitive life stages of lower vertebrates. Our initial approach is to study the teratogenic effects of magnetic fields on the embryonic life stages of rainbow trout, Salmo gairdneri. Several advantages may be gained by examining magnetic effects on a lower vertebrate. This approach is less costly and will provide large numbers of experimental organisms for meaningful statistical analysis. It also permits study of potential latent effects resulting from exposure during embryogenesis. In addition, the data obtained will be applicable to processes occurring in higher vertebrates.

A literature review began in August 1977 to initiate this program. Although a significant number of investigations of the biological effects of magnetic fields on both plants and animals have been conducted, most studies have employed high field strengths over short periods of exposure, with little attention given to the effects of low-level or chronic exposures. Many of these studies were deficient in using inadequate numbers of test organisms for statistical evaluation, and the data obtained were of high variability and were often not reproducible. Since August 1977, we have obtained both permanent magnets and a large Varian electromagnet. Field strengths of 4 kilogauss may be obtained with available permanent magnetic sources; while field strengths of 10 kilogauss are achievable with the electromagnet. Incubation chambers for specimen exposure to magnetic fields were designed and are being tested for their ability to sustain normal development of trout embryos. Experimental protocals for both types of magnets were also completed.

Initial tests are scheduled for November 1977 and will determine effects of magnetic fields on rainbow trout eggs during and after incubation. Trout embryos will be exposed to both homogeneous and nonhomogeneous sources. Mortality, abnormality, delayed development and hatching will be critically monitored. If preliminary tests demonstrate significant differences between control and treatment groups, subsequent tests will be designed to quantify critical maximum field strengths, as well as the relative sensitivity of different developmental life stages.

4.0 Oil and Gas

OIL AND GAS

- **Fate and Effects of Petroleum Hydrocarbons in Marine Coastal Ecosystems**
- **Effects of Refinery Wastes and Oil From Transfer Facilities**
- **Long-Term Effects of Hydrocarbons on Selected Ecosystems and Associated Organisms**

The major objective of these programs, in support of Oil and Gas Technology development, is to establish classes and levels of petroleum hydrocarbons which do not significantly disrupt aquatic ecosystems. This knowledge will provide a strong technical rationale for specifying degree of environmental control technology needed to be applied at production sites, i.e., offshore well head and at the refinery. Additionally, this research provides data for predicting the potential ecological impact of petroleum hydrocarbons resulting from transportation spills and accidents.

Four symposia providing major contributions regarding the fate and effects of petroleum hydrocarbons in the marine environment were held from August 1976 to October 1977. The proceedings of these, containing contributions from our laboratory, are either available or "in press." Much of the recent literature was also reviewed in a two-volume publication edited by D. C. Malins in 1977. Data are also available on the short-term lethal concentrations of various oils and specific petroleum hydrocarbons for a variety of marine organisms. These data were reviewed by Rice et al., 1977. Tissue accumulation (biomagnification) of specific hydrocarbon components was reviewed by Malins and Varanasi (1977) and Anderson (1977), who attempted to relate tissue uptake and release kinetics to the presence of abnormal physiological responses in marine organisms.

It is now time to address the most pertinent questions regarding the fate and effects of petroleum hydrocarbons in the natural environment. Current research programs at the Marine Research Laboratory (Sequim) and PNL (Richland) are designed to answer these questions using integrated approaches and our expertise in ecology, environmental physiology and analytical chemistry. Hypotheses formulated from results of either field or laboratory investigations are verified by testing in both types of systems, so that the significant variables are defined and the rates and magnitudes of effects are evaluated under natural conditions.

The framework of experimentation is designed to provide answers to major problems associated with oil and gas technologies:

- To assess the effects of petroleum hydrocarbons in the natural environment at the population and community level. This is achieved by establishing the statistical validity of sampling methods for populations and by exposure of representative communities to chronic low-level contamination.

- To define the threshold levels of petroleum hydrocarbon contamination in seawater, above which the growth and reproduction (thus, survival of the population) of sensitive marine organisms cannot occur.

- To describe the fate and effects of petroleum hydrocarbons bound to marine sediments. Phases of this large topic regard the recovery (depuration) rate of oiled sediments in the natural environment, the transport of specific components of oil from sediment to benthic organisms, and the effects of contaminated sediment on the health of individual species as well as populations settling (recruited) on the substrate.

OIL AND GAS

- **Fate and Effects of Petroleum Hydrocarbons in Marine Coastal Ecosystems**
- **Effects of Refinery Wastes and Oil From Transfer Facilities**

Principal Investigator: J. R. Vanderhorst

The objective in these studies is to assess effects and establish fates of petroleum hydrocarbons at the population and community levels of biological organization in marine coastal ecosystems. The two major tasks are: 1) identify potential effects and fates of petroleum hydrocarbons in laboratory studies, and 2) measure actual fate and effects of petroleum hydrocarbons resulting from coastal oil and gas technologies.

Identification of Potential Effects - Laboratory Studies

Up to the current year we demonstrated a need for, and developed, apparatus for the continuous exposure of populations and communities to petroleum. A description of effects of continuous flow exposure on a representative Pacific Coast species was published and manuscripts describing the apparatus and initial effects on intertidal colonies were prepared.

The manuscripts describing apparatus and effects were published in FY-77. Data arising from the initial long-term exposure of intertidal colonies were further analyzed and a second manuscript was accepted for publication. In the initial exposure we found that ambient light conditions provided a more fruitful environment for the colonies that rely on natural photosynthetic production for maintenance. Ambient light apparatus was constructed and tested using amphipods and clams.

The colony studies have: 1) allowed tentative identification of the ecological mechanisms (recruitment) resulting in effects on species composition at very low concentrations of petroleum hydrocarbons from No. 2 fuel oil, and 2) identified criteria useful in selection of parameters for evaluation of fate and effects in field studies. Figure 4.1 shows that in control colonies, total numbers of species and numbers of species on pretest colonies (Table 4.1) did not show significant change (P=0.05) during the course

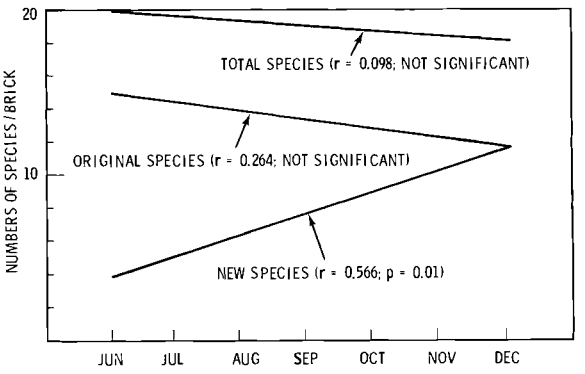


FIGURE 4.1. Relationship of Numbers of Species/Brick (Combined Controls) to Month of Experiment

TABLE 4.1 Numbers of Individuals per Brick Representing Species on Pretest Colonies Used for No. 2 Fuel Oil Experiment

Species per Group	Average Numbers per Brick
Platynereis bicanaliculata	0.33
Lepidonotus caelorus	0.50
Prionospio sp.	0.33
Armandia bioculata	0.16
Lumbrinereis sp.	0.16
Leptocheila savignyi	1.00
Emplectonema gracile	0.16
Exosphaeroma amplicauda	1.83
Aoroides columbiae	1.16
Parallorchestes ochotensis	16.33
Ampithoe simulans	1.00
Ampithoe sp.	0.16
Amphipods (undet.)	9.50
Pagurus hirsutiusculus	0.16
Pagurus sp.	0.16
Pinnixia faba	0.16
Pinnixia occidentalis	0.33
Pugettia gracilis	0.16
Paguristes sp.	0.16
Mytilus edulis	0.16
Alvinia sp.	43.66
Lacuna sp.	9.33
Odostomia sp.	0.33
Bittium sp.	0.33
Nassarius sp.	0.16
Margarites sp.	2.16
Acmaea pelta	0.16
Acmaea persona	0.50
Acmaea digitalis	0.50
Acmaea scutum	0.33
Cooperella subdiaphana	0.50
Mysella tumida	4.17
Hiatella arctica	0.16
Mopalia lignosa	0.33
Lepidozona sp.	0.16
Cyanoplax hartwegii	0.16
Katharina tunicata	0.16

of exposure, while numbers of new species (recruits) increased dramatically (P=0.01) reaching 63% of the total. In contrast, for No. 2 fuel oil treated colonies (Figure 4.2), both the numbers of pretest species and total numbers of species significantly declined (P=0.05). However, the numbers of recruits showed no significant change. The effects on recruitment in the laboratory system provide an estimate of the magnitude of this important factor applicable to certain field conditions. Since species availability among treatments was randomized, these differences in recruitment can best be explained by impairment of the substrate suitability for settling or by larval mortality.

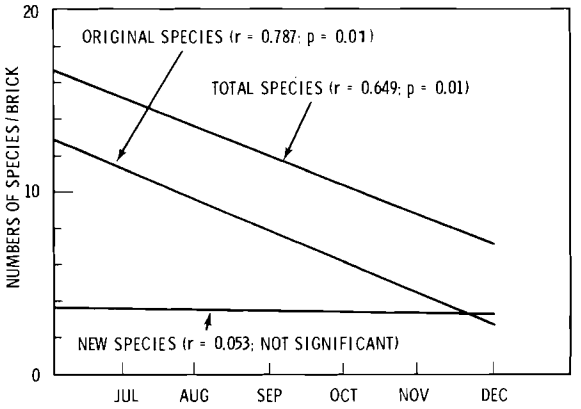


FIGURE 4.2. Relationship of Numbers of Species/Brick (Combined Treatments) to Month of Exposure to No. 2 Fuel Oil

These studies identified a lack of sensitivity of the relative abundance component of species diversity to rather large shifts in dominant composition, the very factor which this index is reported to detect. From this we concluded that the use of such an index would require an assumption of compositional stability for meaningful interpretation. That assumption is seldom warranted. The general index of diversity (H') was sensitive to treatment by No. 2 fuel oil (P=0.01) but did not detect the compositional shift described in Figure 4.1. Our recommended criterion, i.e., a pairwise comparison of occurrence based on chi square contingency, has a built-in measure of species importance since very frequent occurrence in controls is a prerequisite to assigning high probability of effect from contaminant. Yet to be evaluated are: 1) the role of alternate substrate availability in the recruitment process, and 2) the effect of a parent source of petroleum hydrocarbons. We anticipate an initial evaluation of these factors in the coming year.

Measurement of Actual Effects on Populations and Communities from Oil and Gas Technologies

Up to the current year, we designed and implemented a field program to test for differences in biological and chemical parameters at four sites in the north Puget Sound region. Two of the sites were designated as contaminated; one adjacent to an operating oil refinery which received crude oil spillage in 1972; and one in proximity to a proposed superport development. Two sites were

designated as controls and had no known oil spillage. They are in areas with a lower risk of future spillage and have no planned oil or gas technology development. The program includes collection of data on both community and population parameters at sites and a search for quantifiable and petroleum-sensitive elements. For reasons set forth in last year's report, emphasis was given to populations of the native little neck clam (Protothaca staminea) at two of the sites. One primary parameter (stock density) was quantitatively evaluated, and a manuscript prepared for publication. A second primary parameter (individual growth rate) was quantitatively evaluated and a manuscript is being prepared for publication.

The data on Table 4.2 constitute the primary set upon which quantitative evaluation of stock density was based. The data were evaluated in a hypothesis testing framework in terms of hours of effort required to detect given percentages of difference in stock density. Our most effective approach required 100 man-hr of effort/site to detect a 10% difference. That approach had the following characteristics: 1) restricted interest to marketable size stock, 2) used a smaller quadrat size (1/16 compared to 1/4 m^2), 3) used a spatial stratification of sampling based on cover by the green alga, Ulva, and 4) gave equal probability to Type I and Type II statistical errors. The contribution to reduced effort by each of these factors can be seen on Figure 4.3. The computations were based on Type I error probabilities of 0.05 and Type II probabilities of 0.20 except where equal probabilities of 0.20 were used (Bar 4, Figure 4.3). Important implications of our data on density are: 1) that detection of changes smaller than 10% would result in manpower costs greater than the value of the stock, and 2) detection of changes as small as 5% would decimate the little neck clam stock. From the standpoint of resource managers or stewards, the difference in effort for estimates with Type I

TABLE 4.2. Mean Numbers of Little Neck Clams (*Protothaca staminea*) per Sample at the Study Sites

Site	Month/ Year	Quadrat Size, m^2	Clam Size Classification	Number of Samples, n	Mean Number of Clams per Sample	Coefficient of Variation, %
Sequim Bay[a]	5/76	1/4	Sub-marketable	33	2.94	101.6
			Marketable	33	7.88	84.2
			Total	33	10.82	74.2
Sequim Bay[b]	5/76	1/4	Sub-marketable	24	3.62	85.2
			Marketable	24	9.04	73.3
			Total	24	12.66	61.9
Sequim Bay[a]	7/76	1/4	Sub-marketable	38	2.68	86.2
			Marketable	38	9.50	71.1
			Total	38	12.18	63.7
Sequim Bay[b]	7/76	1/4	Sub-marketable	27	2.85	78.2
			Marketable	27	11.15	62.5
			Total	27	14.00	55.4
Sequim Bay[c]	5/77	1/4	Sub-marketable	50	2.10	102.0
			Marketable	50	9.70	52.0
			Total	50	11.80	50.0
Sequim Bay[c]	5/77	1/16	Sub-marketable	50	0.50	149.0
			Marketable	50	2.40	93.0
			Total	50	2.90	79.0
Sequim Bay[c]	7/77	1/4	Sub-marketable	36	2.33	102.6
			Marketable	36	11.03	47.7
			Total	36	13.36	48.9
Sequim Bay[c]	7/77	1/16	Sub-marketable	36	0.55	140.0
			Marketable	36	3.14	57.6
			Total	36	3.69	57.2
Cherry Point[c]	5/77	1/4	Sub-marketable	39	2.90	125.0
			Marketable	39	6.30	85.0
			Total	39	9.20	85.0
Cherry Point[c]	7/77	1/4	Sub-marketable	39	1.80	136.0
			Marketable	39	4.80	107.0
			Total	39	6.60	105.0

[a] Strictly randomized coordinates.
[b] After the fact stratification to remove coordinates outside cover of *Ulva*.
[c] True stratified random sampling with cover of *Ulva* forming strata.

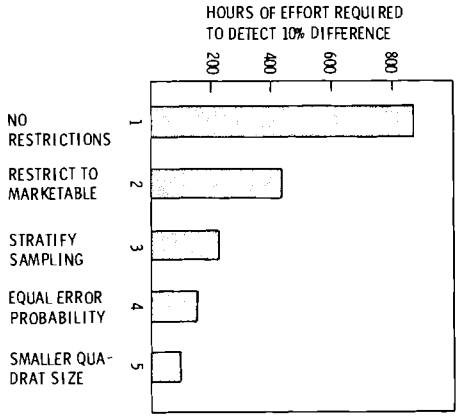


FIGURE 4.3. Contribution to Efforts in Detecting a Ten Percent Change in Density of Little Neck Clams for Major Factors

error probability at 0.05 and 0.20 (70 hr/plot) is one which should be borne directly by potential polluters since the lower probability of error is in their interest.

A similar type of framework was adopted for measurement of clam growth, waterborne oil concentration, and concentration of petroleum hydrocarbons in clam tissues. When analysis of these data are completed, it is expected that the search for sensitive, yet environmentally stable, parameters of populations and communities will continue.

- **Long-Term Effects of Hydrocarbons on Selected Ecosystems and Associated Organisms**

Principal Investigator: J. W. Anderson

The objectives of this program are to determine safe (threshold) levels of hydrocarbon contamination in water for the survival of populations of sensitive marine organisms, and the fate and effects of sediment-bound petroleum hydrocarbons in the marine ecosystem. This research includes: 1) effects of soluble hydrocarbons on growth and reproduction, 2) transfer and effects of hydrocarbons from sediments, and 3) retention time of hydrocarbons in sediments and recruitment rate of benthic organsims.

Effects of Soluble Hydrocarbons on Growth and Reproduction

The Flowing Seawater Exposure System is being used for studies of soluble hydrocarbons from Prudhoe Bay Crude oil. The components of this contaminated seawater have been characterized by utilyzing several analytical methods. Total oil concentration has been monitored by absorbing the oil from the exposure water on XAD resins and quantifying the oil absorbed using infrared spectrometry. The concentrations of light monocyclic aromatic hydrocarbons have been determined using a helium equilibration gas chromatograph technique (Bean and Blaylock, 1977). Concentrations of saturate, diaromatic and triaromatic hydrocarbons have been determined from analysis of XAD-2 resin column extracts by capillary gas chromatography. The results of the analytical procedures using Prudhoe Bay Crude oil are given in Table 4.3. Results indicate that the hydrocarbon type found at the highest concentration is the monocyclic aromatics. This observation is consistent with those made on other oil/water systems (Anderson et al., 1977; Bean and Blaylock, 1977).

Comparisons were made between the compound type distribution in the continuous flow apparatus and the original crude oil. Naphthalene has been enriched more than sevenfold over its original concentration in Prudhoe Bay Crude. The methyl- and dimethylnaphthalenes also represent a greater contribution to the total hydrocarbon composition of the exposure water, than in the original crude. Tricyclicaromatics, with the possible exception of phenanthrene also appear to be enriched, but n-alkanes contributed substantially less to the total hydrocarbon composition of the water than they did to the original oil. These observations are consistent with the contention that the crude oil is existing as a dispersion of fine droplets suspended in seawater with the more water-soluble aromatic hydrocarbons existing in truly aqueous solution. Those hydrocarbons, with intermediate water solubility, are apparently present in both the oil and water phases at equilibrium concentrations. This basic physical distribution was postulated earlier from studies of dispersions of No. 2 fuel oil in the same seawater delivery system (Vanderhorst et al., 1977).

Two organisms (Neomysis, mysid and Anonyx, amphipod) have been used in extensive studies utilizing the Flowing Seawater Exposure System. One manuscript is in press (Anderson and Kiesser, 1978) concerning the amphipods, and one paper is in preparation on each of these species. They have both been maintained for an entire generation under both control conditions and constant exposure to 0.1 to 0.2 ppm total hydrocarbons. Findings include: 1) the affects of short-term exposures on

TABLE 4.3. Concentrations of Hydrocarbons in Continuous Flow Bioassay Apparatus as Determined by IR, Helium Equilibration, and Capillary GC Techniques. Concentrations are in parts-per-billion (ppb)

Total oil (IR analysis) 141.6 ± 14.4 ppb

Monoaromatics (helium equilibration analysis)

Compound	Concentration, ppb
Benzene	36.1 ± 5.2
Toluene	55.5 ± 4.5
Ethylbenzene	5.2 ± 0.8
Meta and Para Xylenes	20.0 ± 11.1
Ortho Xylene	12.5 ± 0.6
Total Trimethylbenzenes	9.2 ± 1.0
Total Monoaromatics	138.0 ± 23.2

Saturates, diaromatic, and triaromatic hydrocarbons (capillary GC)

Compound	Concentration, ppb
C-11	0.145 ± 0.012
C-12	0.153 ± 0.008
C-13	0.150 ± 0.005
C-14	0.163 ± 0.021
C-15	0.186 ± 0.012
C-16	0.181 ± 0.026
C-17	0.203 ± 0.022
Pristane	0.134 ± 0.009
C-18	0.204 ± 0.021
Phytane	0.093 ± 0.009
C-19	0.212 ± 0.010
C-20	0.195 ± 0.014
C-21	0.163 ± 0.002
C-22	0.181 ± 0.001
C-23	0.154 ± 0.008
C-24	0.140 ± 0.003
C-25	0.113 ± 0.004
C-26	0.101 ± 0.001
Total Saturates	2.871 ± 0.188
Naphthalene	1.604 ± 0.158
2-MN	1.338 ± 0.051
1-MN	1.057 ± 0.039
Total MN	2.395 ± 0.090
1-Ethyl + 2-Ethyl Naphthalene	0.159 ± 0.009
2,6 + 2,7-DMN	0.248 ± 0.040
1,3 + 1,6-DMN	0.403 ± 0.024
1,7-DMN	0.288 ± 0.021
1,4 + 2,3 + 1,5-DMN	0.258 ± 0.011
1,2-DMN	0.078 ± 0.008
Total DMN	1.387 ± 0.064
TMN-1	0.085 ± 0.009
TMN-2	0.058 ± 0.006
TMN-3	0.074 ± 0.019
TMN-4	0.086 ± 0.009
2,3,6-TMN	0.045 ± 0.003
TMN-5	0.092 ± 0.012
Total TMN	0.572 ± 0.041
Phenanthrene	0.097 ± 0.008
MP-1	0.037 ± 0.009
MP-2	0.047 ± 0.018
MP-3	0.058 ± 0.015
MP-4	0.042 ± 0.009
Total MP	0.184 ± 0.021
DMP-1	0.054 ± 0.003
DMP-2	0.031 ± 0.004
DMP-3	0.019 ± 0.002
Total DMP	0.104 ± 0.007
Total Diaromatics + Triaromatics	6.501 ± 0.379

MP = methylphenanthrene
DMP = dimethylphenanthrene
MN = methylnaphthalene
DMN = dimethylnaphthalene
TMN = trimethylnaphthalene

survival, 2) affects of exposure to contaminated sediment (for the burrowing amphipods), and 3) the affects of long-term exposure on growth and reproduction. Each aspect of these findings is correlated with exposure water concentrations and tissue accumulation of hydrocarbons. One phase of these studies is summarized in Figure 4.4, which illustrates the effects of a 0.24 ppm concentration of total hydrocarbons on the reproductive capability of Anonyx. These animals were exposed a total of 21 weeks and the differences between the reproductive parameters of control and exposed organisms were determined.

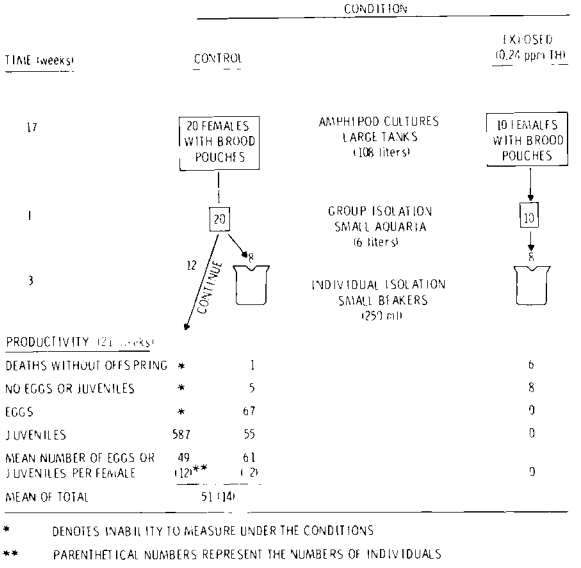


FIGURE 4.4. Exposure Conditions of Female Amphipods and the Resulting Production of Offspring. Animals were Exposed to 0.24 ppm Total Hydrocarbons for a Total of 21 Weeks.

Growth and tissue contamination were measured at various intervals during the exposure and it was observed that maximum size was not reduced in exposed animals, while tissue contamination reached a stable level. These results demonstrate that long-term studies of this type are required to assess actual deleterious effects. If size were used as the measure of hydrocarbon effect, the conclusion would have been little or no response to exposure. However, reproduction was shown to be reduced such that no viable offspring were produced by exposed organisms.

Studies are presently underway, using both amphipods and mysids, which represent sensitive marine organisms. There is little doubt that the ability to reproduce, and thus assure survival of the species, is a significant parameter to correlate with the level of hydrocarbon exposure. Our findings will

provide accurate evaluation of the levels of petroleum hydrocarbon contamination in marine and estuarine waters that are hazardous to marine organisms, populations and communities. Regulations on oil spillage and effluent releases can then be determined from a sound scientific data base.

Effects of Transfer of Hydrocarbons From Sediments to Organisms

In the last Annual Report, our findings regarding the uptake and depuration of naphthalenes from oil by the deposit-feeder, Phascolosoma agassizii was reported and these results have been published (Anderson et al., 1977). As noted earlier in this discussion, the amphipod, Anonyx, was exposed not only to waterborne hydrocarbons but also sediment-bound oil. The results of these studies (Anderson and Kiesser, 1978) indicate that no significant mortality was produced by an 18 day exposure to sediment hydrocarbon concentrations of about 300 ppm total hydrocarbons and 2 ppm total naphthalenes. Animals exposed directly to oiled sediment accumulated 8 ppm total naphthalenes, while those suspended above the substrate in the same tanks contained 4 ppm (Figure 4.5). These data indicate that accumulation of these diaromatic compounds was via water, which received contamination leaching from sediment to interstitial water. As in several earlier studies, the compounds present in the tissues during the later phases of exposure to both water and sediment were the alkyl-naphthalenes and the parent compound (naphthalene) had been lost from both sediment and tissue.

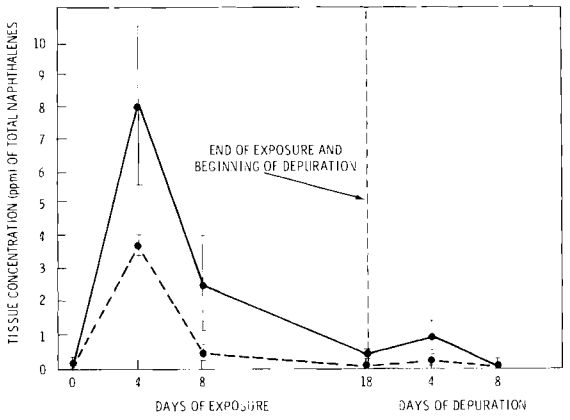


FIGURE 4.5. Uptake and Release of Naphthalenes by *Anonyx* Exposed to Oiled Sediment (solid line) or Hydrocarbons Released from Sediment (dashed line). Means and Standard Deviations (bars) were Derived from 3 Samples of 2 Animals Each.

In a recent presentation (Roesijadi, Anderson and Blaylock, 1978), the uptake of aliphatic and diaromatic petroleum hydrocarbons from contaminated sediment by two deposit-feeding organisms (Macoma inquinata and Phascolosoma agassizii) was compared to that of a filter-feeding bivalve (Protothaca staminea). The highest uptake was exhibited by the deposit-feeding clam, Macoma inquinata, which accumulated both aliphatics and diaromatics during the 60 day field exposure to oiled sediment. The total uptake amounted to about 2.5 ppm, while the sediment declined from 887 ppm to 420 ppm over the 60 days. These data again demonstrate relatively low uptake rates from highly contaminated sediment, which may indicate rather low bioavailability of hydrocarbons bound to sediment. In these field exposures, it is not possible to separate uptake from ingestion of sediment particles from the possible contribution of hydrocarbons present in interstitial water. Since uptake from water generally results in high accumulation factors in tissue, a low concentration in the surrounding water could have produced the observed contamination levels in these benthic organisms. In other studies with Macoma differences were shown in the uptake and release rates of hydrocarbons with two to five aromatic rings and it appeared that the higher molecular weight compounds are taken up from sediment over a longer period and retained longer in the tissues (Roesijadi et al., 1978).

Relatively few effects of oil-contaminated substrate on marine organisms have been observed in these studies and those few reports in the literature. As will be discussed later, benthic recruitment was not significantly suppressed by oiling of substrate (Anderson et al., 1978). However, Roesijadi and Anderson (1978) recently demonstrated effects of oiled sediment on the condition index and free amino acid content of Macoma inquinata exposed in a field experiment for 38 days. Both laboratory and field exposures indicated that these parameters may be sensitive indices of the effects of hydrocarbons on these bivalves.

In general, the data obtained on bioavailability of sediment-bound hydrocarbons all show similar trends. The extent of uptake is never large as compared to concentrations in the sediment. Uptake may well be via interstitial water rather than from ingestion of particles. Results of effects studies also indicate that sediment-bound hydrocarbons may be two orders of magnitude less toxic than the same concentration present in water. These studies are most helpful in assessing the retention time of hydrocarbons in sediments and the effects of these sinks on marine organisms.

Retention Time of Hydrocarbons in Sediments and Recruitment Rate of Benthic Organisms

Three separate field installations, consisting of clean and oiled sediment in fiberglass trays, were placed in the intertidal zone of Sequim Bay, Washington, to determine rates of hydrocarbon depuration and recruitment of benthic organisms. Detailed chemical analysis, using glass capillary gas chromatography, and gas chromatography/mass spectroscopy were conducted such that individual compounds and hydrocarbon classes associated with the sediment after varying periods of field depuration could be quantitated. Depuration rates of hydrocarbon types in sediment receiving oil on the surface (installations I and II) decreased in the order of saturates, methylnaphthalenes and methylphenanthrenes (Figure 4.6). Rates of specific compound and hydrocarbon class depuration followed the general pattern exhibited by total hydrocarbons (infrared analyses). In a period of 100 days, total hydrocarbons in surface oiled, coarse sediments (I and II) decreased by 82 to 88%, while the amount in the finer substrate with mixed-in oil (III), only decreased by about 21% and remained quite stable up to 290 days (Figures 4.6 and 4.7).

The ratios of nC_{17}/pristane and nC_{18}/phytane were monitored for installations I and III (oil mixed with sediment). The ratios remained constant for a period of 4 to 6 months and then dropped sharply, suggesting the presence of oil biodegradation.

The differences observed in the rates of depuration of hydrocarbons from the two types of oil-sediment systems can be attributed to many physical, chemical and biological mechanisms. Tidal and wave action, surface volatility, photochemical and biodegradative processes are likely the major contributors to the depuration rates. All of these are probably more active at the surface than at lower layers of the sediment, thus explaining the slower depuration rates observed when oil is mixed into sediment.

Initial concentrations of oil in sediments upon field emplacement were about 5,000 to 6,000 ppm in installations I and II, and approximately 700 ppm in installation III. After 100 days in the field, concentrations of toxic aromatics in the sediments of all three installations were relatively low (2-4 ppm methylnaphthalenes and 1-3 ppm methylphenanthrenes) and quite similar. This may help to explain the similar results obtained in all three installations regarding benthic organism recruitment. There was no clear indication that the contamination resulted in the suppression of populations of two species of bivalves (Mysella tumida and Psephidia lordi) or three species of polychaetes (Ophiodromus pugettensis, Platynereis bicanaliculata, and Armandia bioculata). Samples

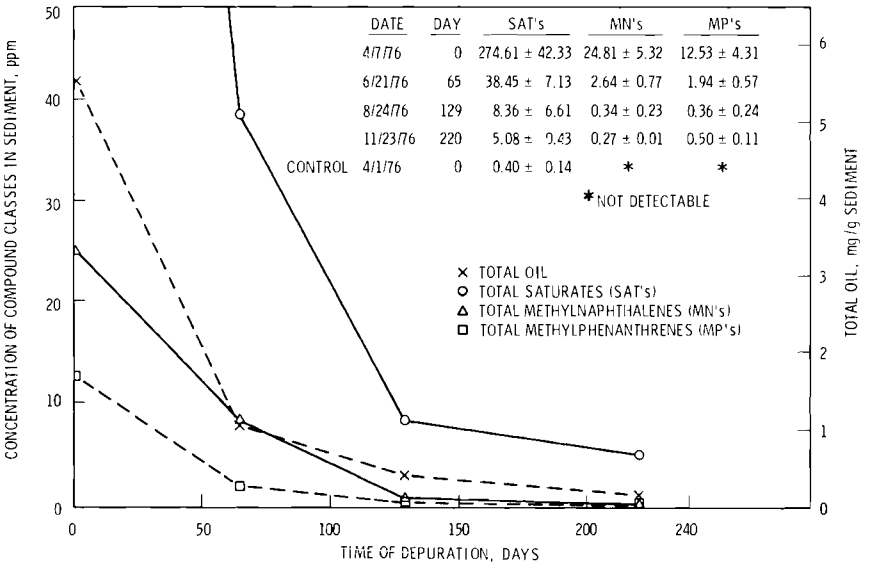


FIGURE 4.6. Concentrations of Compound Classes in Sediments of Installation II, as a Function of Depuration Time

have now been analyzed from sediments in the field for 467 days, and there are still no clear indications that oiling has reduced the abundance of these five species. The details of this research have been submitted for publication (Anderson et al., 1978) and presented at a symposium. Further analysis of our findings is in progress and additional studies will be conducted to examine other aspects of the problem. At present it appears that hydrocarbons are rather rapidly depurated in this environment and affects on benthic recruitment are minimal.

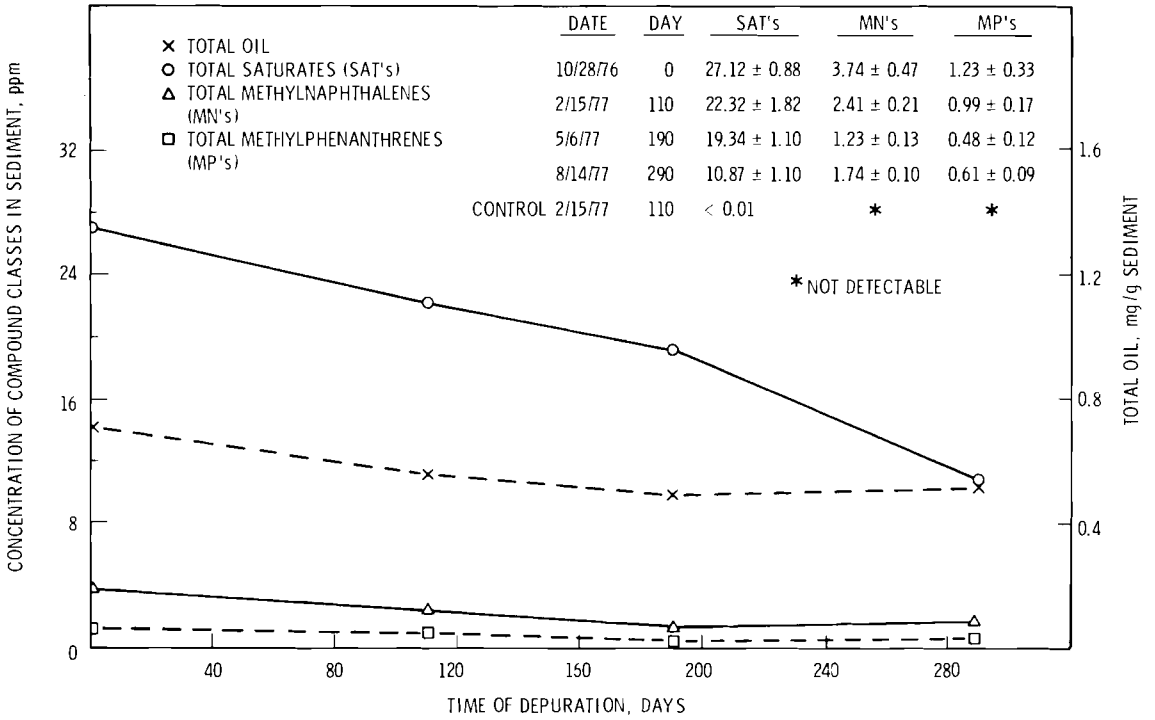
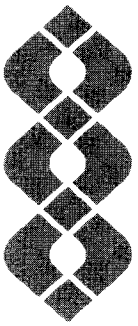


FIGURE 4.7. Concentrations of Compound Classes in Sediments of Installation III, as a Function of Depuration Time



5.0 Hydroelectric Generation/Pumped Storage

HYDROELECTRIC GENERATION/PUMPED STORAGE

• Effects of Hydroelectric Generation on Riverine Ecology

Nearly all available sites for development of conventional hydroelectric generation in the Pacific Northwest have been utilized. Pumped storage hydro is an alternative which would aid in meeting peak power needs and in conservation of fossil and nuclear fuels. The Corps of Engineers has identified several hundred potential sites in the region which could support pumped storage development. Present information indicates that pumped hydro storage is the least costly of all currently practicable storage means.

This is a small ecological project designed to develop data for measuring aquatic environmental impact at hydroelectric and pumped storage sites. Dissolved gas supersaturation due to air entrainment, modification of ambient temperature regimes, fluctuation in river flow caused by the use of hydroelectric facilities, and loss of appropriate wildlife habitat caused by the impoundment of large quantities of water, all create certain problems.

One effect of pumped storage operation will be due to daily water level fluctuations in reservoirs and downstream from impoundments. Fluctuations in water level due to pumped storage or changing river flows may result in reduced primary production, losses of fish food organisms, and mortalities to eggs, fry, and adult fishes due to desiccation, entrapment, stranding, or enhanced predation. Impacts on the commercially important salmonids and other game fish populations are of particular interest.

Effects of impoundment also include alterations in sediment load, temperature, drift organisms, and dissolved gas concentrations below the dam in addition to the major change from a stream to a reservoir behind the dam. Each of these factors potentially impacts productivity of aquatic systems. It is our goal to define significant impacts at potential and developed sites and to develop a systematic data base of hydro energy resources. Engineering developments to minimize detrimental effect on biological resources will require a substantial body of ecological data. Alternatives to present or planned operations and facilities are being considered and evaluated as appropriate.

- **Effects of Hydroelectric Generation on Riverine Ecology**

Principal Investigators: D. H. Fickeisen and J. C. Montgomery

Other Investigators: C. D. Becker

Technical Assistants: D. D. Dauble

Norcus Appointees: R. T. Cordo, D. Crass, S. W. Cubberly and J. M. Haynes

Experimental phases of gas bubble disease work were completed. Studies on effects of water level fluctuations due to peaking operation of conventional hydroelectric facilities continued, with a substantial effort devoted to radiofrequency telemetry of smallmouth bass movements. The extremely low runoff this year aided observations of adult and juvenile fish and their food supply that were entrapped in ponds when the water level fell. Determining the impacts of daily fluctuations in water level is of regional and general importance as use of hydroelectric facilities (conventional and pumped-storage) increases worldwide. A special studies project was also undertaken to determine research needs related to pumped storage development.

River Level Fluctuations

Changes in turbine loading at dams to meet variable power demands result in daily and weekly fluctuations in discharge volumes and, hence, in downstream water levels. Shoreline organisms are, therefore, subjected to stranding, increased exposure to predators, entrapment, or desiccation. The most important effects on riverine biota in the Hanford Reach of the Columbia River apparently are entrapment of adult and juveniles of important fish species and subsequent mortality due to desiccation, predation, solar heating, and possible dewatering of chinook salmon redds under extreme low flows during winter. Loss of food organisms occurs and may be an ecologically significant factor.

Earlier work indicated detrimental impact on smallmouth bass spawning and behavior in the Hanford Reach of the Columbia River. We used radiofrequency telemetry of some 40 adult bass to monitor their location. Figure 5.1 shows a tagged fish. Transmitter life was up to 120 days, permitting the position of fish to be tracked throughout the summer of 1977. Tracking has provided positive correlations of tagged bass with predation on entrapped salmonids. Data are presently being analyzed from this intensive study.

Observations on entrapment and subsequent mortality of fish continued to provide background information needed to design a qualitative study to be initiated next year. These included "walks" at frequent intervals over three main study areas while observations of effects were recorded and documented.

Drought conditions in the Pacific Northwest during FY-77 facilitated study of several shoreline ponds in which fish were entrapped. Normally the ponds are connected to the main-stem river during the spring run-off season. One pond of several acres was found to contain adult bass and coarse fish as well as bass fry. However, the food supply was very limited and quickly depleted, resulting in poor condition factors and stunting of bass. Smaller ponds were commonly subject to high summer temperatures, causing mortalities to several species of small fish entrapped in them.

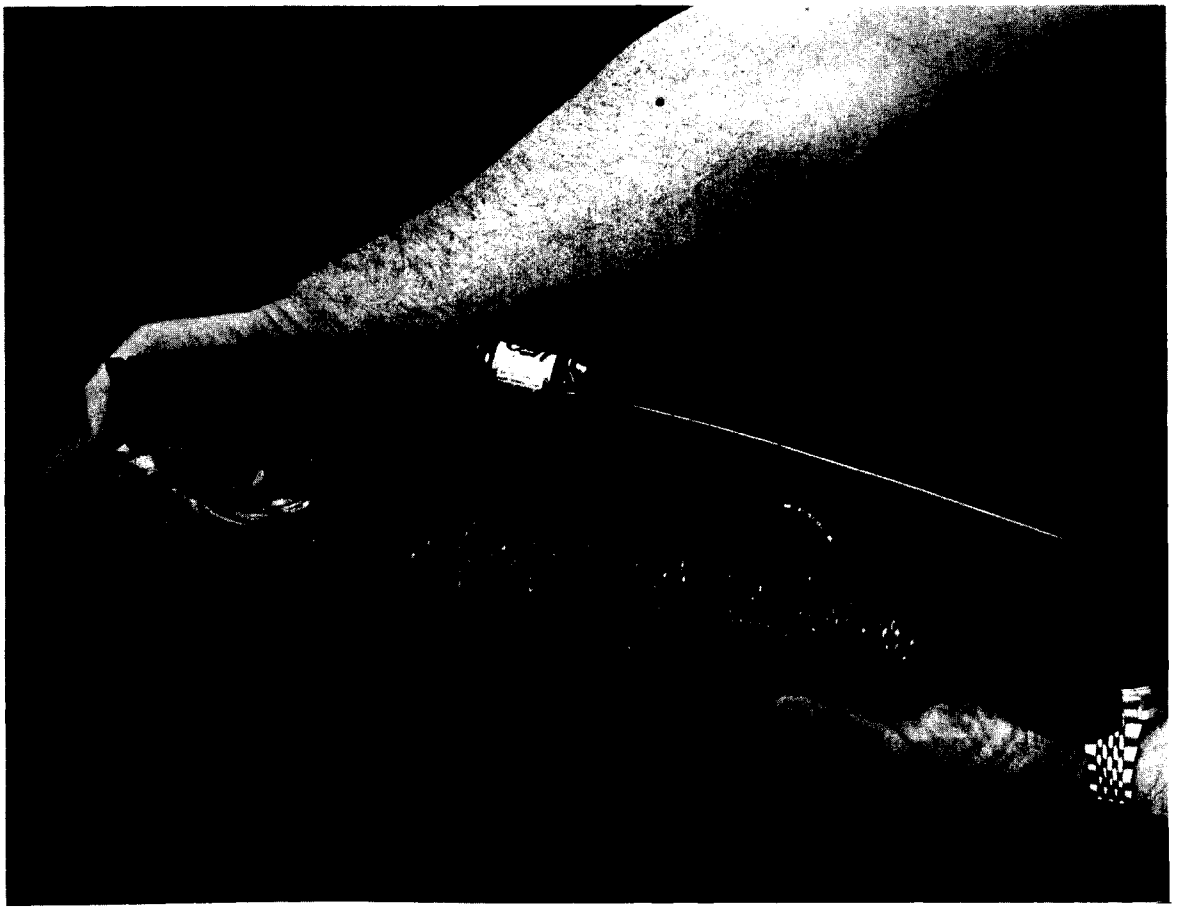


FIGURE 5.1. Smallmouth Bass with Radiofrequency and Dart Tags, Either of Which is Applied for Monitoring Fish Movement Under Fluctuating Water Conditions

Preliminary results are presently being analyzed and will be prepared for publication. Desiccation losses have been confirmed for 12 species of Columbia River fish, and 18 species have been found entrapped.

Gas Bubble Disease

The final phase of our laboratory tests on gas bubble disease was completed by exposing rainbow trout, black bullhead, and pumpkinseed sunfish to the combined stressors of temperature and dissolved gas supersaturation. The results indicate that severe (but sublethal) thermal shock did not significantly alter tolerance of fish to gas supersaturation. This result is important in extending previous work on tolerance to gas supersaturation in situations such as exposure to power plant cooling water discharges where both gas supersaturation and thermal increases may simultaneously stress fish.

These results and those of our previous gas bubble disease studies are presently being modeled to provide predictive algorithms for effects of gas supersaturation and temperature.

6.0 Shale Oil

SHALE OIL

- **Terrestrial Effects of Oil Shale Development**
- **Development of Chemical Reactions, Stability and Transport Model of Oil Shale Process Wastes in Soil**

The search for domestic energy supplies will lead to increased exploitation of domestic fossil fuels. Vast domestic resources of oil shale may provide a long-term alternative to petroleum-derived fuel products. Practical processing experience with shale oil is limited in comparison to other fossil fuel production since serious industry consideration of shale oil production has not been concerted. Methods proposed for winning shale oil from its deposits range from conventional mining and aboveground retorting technologies to in situ combustion methodology. Although the technical and economic aspects of this wide range of options have been generally studied, the environmental impacts must be assessed, as questions of siting, control technology, licensing, and regulation can strongly affect the best production strategy.

Three major segments of the aboveground shale oil production cycle have potential for environmental consequences. These include mining of the shale, extraction of the shale oil and disposal and reclamation of processed shale. In the mining process, aquifer disruption, runoff and movement through the soil profile to groundwaters of process residuals may result in impact on streams and rivers. During the extraction cycle (crushing, retorting and refining), the major impact will result from gaseous emissions and liquid effluents containing organic, saline, and trace metal residues. Disposal of retorted shale presents problems in loss of recreation land, in reclamation of land occupied by retorted shale, and in environmental and human exposure resulting from resuspension of particulates and leaching of pollutants to ground and surface waters. The use of pyrolytic methods, together with a relatively high concentration of organic N in oil shale, can lead to the formation of organic components which are unique, thus producing environmental problems not ordinarily encountered in other energy industries.

The current PNL program focuses on the chemical characterization of effluent residuals (see PNL 2500, Part IV, Physical and Technological Programs), the potential toxicity of effluent residuals (see PNL 2500, Part I, Biomedical Sciences), and the soil processes affecting plant uptake toxicity and vegetational events (this report). Supporting research on the feasibility of revegetation in arid climates and application research on a water harvesting approach are reported in the preceding section of this report, where they are of primary importance to surface mining of coal (see Section 1.0). Dr. Sidney Marks is coordinator for the comprehensive program. The specific studies reported in this section concern soil biochemical transformations impacting human food chains and the viability of green plants. This effort represents a multitask interdisciplinary research approach which takes unique advantage of PNL capabilities in hydrocarbon chemistry, soil science, microbiology, and plant physiology. Principal objectives of the project are to: 1) develop a basic knowledge of important extraction and environmental processes influencing the form, mobility, chemical, and microbiological stability, and plant availability and toxicity of organic residuals in retort waters and retorted shales, 2) model movement of stable organic residuals in soil and retorted shale, and 3) develop a rational basis for selection and subsequent laboratory and field testing of proper soil conditioners, irrigation waters and native or agricultural plant species to revegetate retorted shales for permanent, safe restoration of shale lands. The studies are being closely integrated with field investigations by other Federal agencies and universities. Together, these programs should provide a basis for assessment of the potential terrestrial effects of waste disposal and for viable restoration of shale lands.

- **Terrestrial Effects of Oil Shale Development**
- **Development of Chemical Reactions, Stability and Transport Model of Oil Shale Process Wastes in Soil**

Principal Investigators: R. E. Wildung, T. R. Garland, R. G. Riley, D. J. Silviera, J. E. Rogers, R. M. Bean, and S. W. Li

Initial program emphasis has been directed toward 1) development of sampling protocols with industry, collection of samples from a range of retort processes, and physicochemical characterization of retort waters, retorted shales, retorted shale leachates, and soils of the region, 2) measurement of the sorption properties of retort water components in retorted shale and soils, 3) initiation of studies to evaluate the role of microbes in influencing the stability of organic residuals in retorted shales and retort waters, and 4) initial investigation of the plant availability and effects of organic and inorganic residuals in retorted shale and retort waters.

Field studies of the microbial stability and movement of trace metals and organic residuals in retorted shale have been initiated at the Paraho Oil Shale Demonstration Project, operated by Development Engineering Incorporated, Anvil Points, Colorado and in the Piceance Creek Basin, Colorado, in conjunction with Colorado State University. The results of these investigations will be used to validate laboratory models of the mobility of residuals in the oil shale region.

The program has served as a basis for technical support of current programs in integrated regional assessment of oil shale development and for assessment by the National Academy of Sciences of redistribution of accessory minerals as a result of mining and extraction of oil shale.

CHEMICAL STUDIES - RETORT WATER

Characterization of Retort Water for Major and Trace Inorganic Ions

Water is a valuable and scarce commodity in areas that have the potential for large-scale production of oil from shale. During the retorting of oil shale, sizable quantities of water are produced along with the oil. This retort water has a high dissolved organic carbon content along with various inorganic components. The ability to use this water directly for dust control, and compacting retorted shales or after some cleanup would be beneficial. Studies of the retort water, both laboratory and field, require that the major and trace elements be characterized and that the potential variation in specific elements be known prior to the initiation of experiments.

Three retort waters, one sampled from the Laramie simulated in situ retort in December 1974 and two from the Paraho process sampled in May 1977 and August 1977 were analyzed.

After sampling, the retort waters were kept at 4°C, and clarified by filtration through acid, base and water-washed quartz glass wool. After clarification, the individual retort waters were filtered through 0.4 and 0.01 μ filters and analyzed by either plasma atomic spectroscopy, graphite furnace atomic spectroscopy, or atomic spectroscopy. With the exception of Hg, filtration through the 0.4 and 0.01 μ filters did not significantly change the quartz wool-clarified results. The results obtained for the three samples of retort water (Table 6.1) show a marked difference between the Laramie and Paraho processes (or shale source) for most of the elements. The two samples of the retort water from the Paraho process are quite similar in elemental composition at the two sampling times with the notable exceptions of Cu, Cd, Zn, and As. For comparison purposes, the trace element content of the approximately one-third column volume leachate of a soil near the Anvil Points plant is shown. This soil contains quantities of raw shale derived from the cliffs which contain the source of oil shale for the Paraho process. With the exception of C, F, and As, the Laramie retort water has comparable or lower concentrations of most of the trace elements than the soil leach sample. At the pH of the retort water and the level of the organic C, it would not be unreasonable to have most of the ions of +2 or +3 valence in a complexed form. The concentration of soluble Fe suggests it would be complexed. Complexation may markedly influence the behavior of metals in soils and retorted shales and subsequent availability to plants.

TABLE 6.1. Concentration of Trace Elements in Three Samples of Retort Water[a]

Element	Laramie Simulated In Situ (Dec. 1974)	Paraho (May 1977)	Paraho (Aug. 1977)	Anvil Points Soil Leachate (1/3 Col. Vol.)
	μg/ml			
pH	8.45	8.45	8.45	7.90
C (organic)	15,000	37,000	42,000	73
C (inorganic)	670	1,700	2,000	69
$SO_4^=$	2,550	18,900	22,300	2,022
Cl^-	110	1,350	1,750	520
P (as $PO_4^=$)	2.1	9.4	11.3	<2.0
F^-	18.8	46.0	49.5	1.5
As	6.8	6.8	16.0	0.01
Ba	<0.03	0.27	0.24	0.077
Be	<0.005	<0.005	<0.005	<0.005
B	0.69	32.2	37.6	1.7
Cd	<0.001	0.001	0.10	<0.001
Ca	10.2	23.1	32.1	131.
Cr	0.030	0.090	0.090	0.006
Co	0.04	0.28	0.27	<0.01
Cu	0.11	0.019	22.6	0.021
Fe	4.96	1.44	1.09	<0.01
Pb	0.007	0.015	0.016	<0.005
Mg	19.5	767.	951.	252.
Mn	<0.01	0.28	0.16	0.024
Hg	0.001	<0.001	0.001	<0.001
Mo	0.20	0.59	0.63	0.38
Ni	0.18	0.65	0.41	<0.01
K	15.7	37.8	61.8	51.3
Se	0.19	4.00	3.25	<0.02
SiO_2	38.7	41.0	32.1	26.9
Ag	<0.001	<0.001	0.002	<0.001
Na	70.0	259.	395.	837.
Sr	0.059	0.078	1.18	2.93
V	0.60	0.15	0.15	<0.10
Zn	0.22	0.23	5.10	0.04

[a] Results are on a <0.01μ filtered sample

Molecular Weight Distribution Analysis of Organic Components of Retort Water by High Pressure Gel Filtration Chromatography

Water derived from an oil shale retorting process consists of a complex mixture of polar organic components of varying molecular weights. Until recently, the ability to directly and rapidly monitor the molecular weight distribution or changes in distribution of these types of aqueous complex mixtures has been hampered by the lack of modern analytical techniques. An initial solution to this problem has been the development of a high-pressure liquid chromatographic technique employing columns of porous silica microspheres.

A description of the application of this technique to the analysis of Paraho retort water is as follows. A sample of the retort water (100 μl, adjusted to pH 7.0) was chromatographed on μ-Bondagel (125Å, capable of separating components 50,000 MW or less) employing water as the mobile phase. The results of the 9 min analysis are shown in Figure 6.1. Dextran standards were used to calibrate the column and a select set of amino acids and other organic acids were also used in the calibration to compensate for adsorption effects. Based on retention volume and the calibration curve (Figure 6.1), apparent molecular weights were assigned to refractive index (RI) and ultraviolet (UV-254 mm) peaks. These results are shown in Table 6.2. Assuming that response from the refractive index is a qualitative measure of organic mass, a major portion of the mass appears to be in the range of MW ∿ 1000 or less.

A future study will correlate the total organic carbon content of isolated fractions to the molecular weight distribution profile of retort water. Also, the technique is being further developed to provide information on the fate of organic components of retort water in microbial, soil, and plant studies.

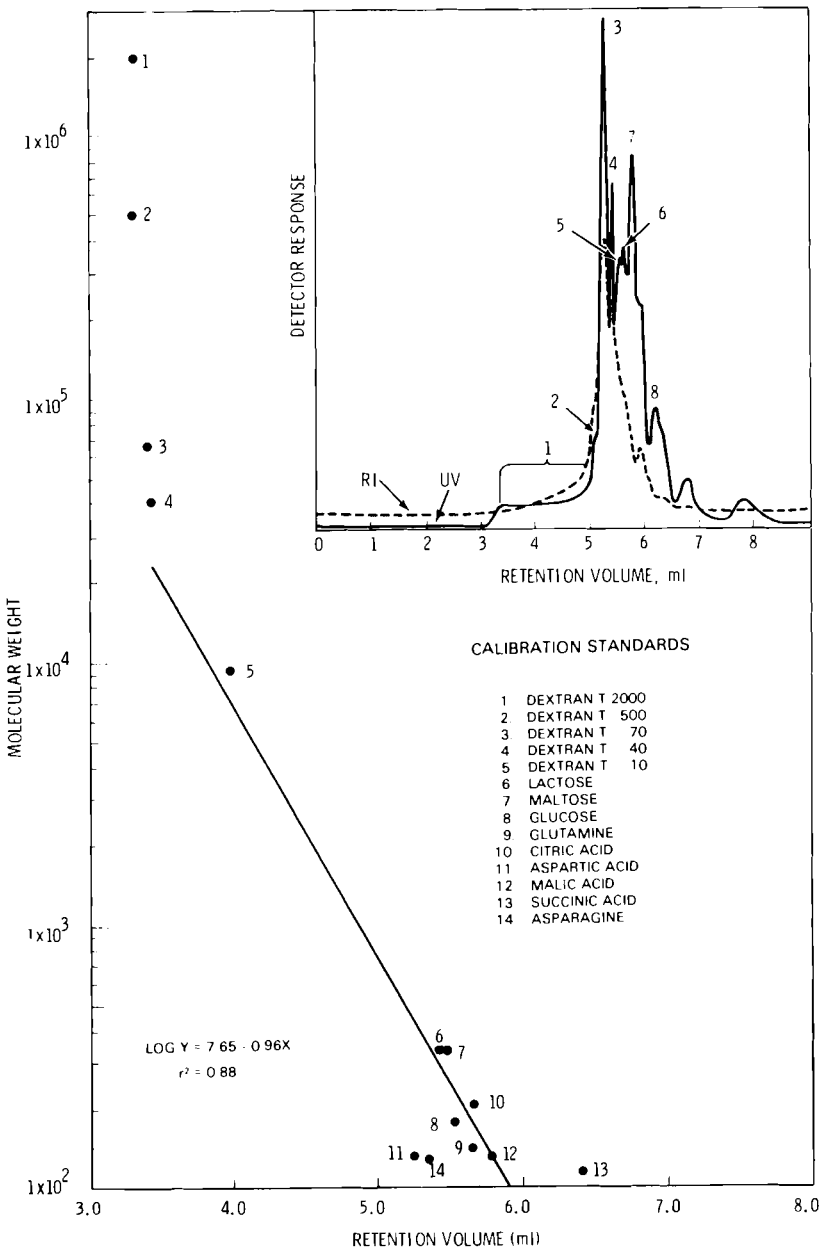


FIGURE 6.1. Calibration Curve for μ Bondagel Column. Inset shows gel filtration chromatogram of retort water.

TABLE 6.2. Apparent Molecular Weights (MW) of RI and UV Peaks Derived from the Fractionation of Paraho Retort Water on μ-Bondagel (See Fig. 6.1 inset).

Peak or Range	MW (RI)	MW (UV)
1	$1.2 \times 10^6 \rightarrow 600$	$1.2 \times 10^6 \rightarrow 630$
2	~630	~630
3	~410	~410
4	~280	~280
5	~210	~210
6		~170
7		~110
8		~ 40

Identification of Saturate and Aromatic Hydrocarbons in Shale Oil by Capillary Gas Chromatography

In order to be able to establish relationships to component origin in the studies of mobility and toxicity of retort water and retorted shale wastes, characterization of the parent shale oil is necessary. In this study, some saturate and aromatic hydrocarbons in shale oil derived from the Paraho process were identified and quantitated and

the results were compared to the composition and concentrations of the same components in Prudhoe Bay Crude oil (PBC).

Shale oil and PBC were separated into saturate and aromatic fractions by silica gel chromatography and each fraction was analyzed by temperature programmed glass capillary gas chromatography. The quantitative results of triplicate analysis of both oils are shown in Table 6.3. Total concentrations of saturate hydrocarbons were 4 times as high in shale oil as compared to PBC. Concentrations of individual naphthalenes were similar in both oils with the exception of 1,7-dimethylnaphthalene and several of the trimethylnaphthalenes which were present in higher concentrations in shale oil. The most striking difference occurred with the triaromatics where no phenanthrene or alkyl-substituted phenanthrenes were detectable in shale oil. Although quantitative information is not available at this time, considerably greater concentrations of aliphatic olefins were indicated in shale oil from qualitative examination of the chromatogram of the saturate fraction. In one analysis, olefins were found to amount to 18% of the hydrocarbon fraction of an in situ crude shale oil. Characterization of components of shale oil will continue with emphasis being placed on the identification and quantitation of more polar constituents, such as organonitrogen compounds. Examination of these compounds in light of compounds identified in retort water will be made.

Organic Nitrogen Compounds in Retort Water

Only a few percent of the water-soluble organic material in Paraho retort water are soluble in organic solvents. Nevertheless, the organic-soluble fractions contain volatile, lower molecular weight organic components which may be of importance due to their soil mobility and biological activity. Studies are underway to isolate and identify these lower molecular weight components, with particular emphasis on the nitrogen-containing constituents, using nitrogen-specific capillary gas chromatography.

Several extractions of a retort water sample using solvents of increasing polarity at increasing pH values were performed in order to isolate fractions rich in nitrogen components. Extractions were performed successively with isooctane, cyclohexane, and benzene at pH 2, 5, 8, and 11. Isooctane at pH 8 was shown by gas chromatographic analysis to produce a particularly clean fraction which was relatively rich in organonitrogen components.

TABLE 6.3. Concentrations (mg/gram oil) of Saturate and Aromatic Hydrocarbons in Shale Oil and Prudhoe Bay Crude (PBC).

Compound	Concentration in Shale Oil (mg/gram oil)	Concentration in PBC (mg/gram oil)
C_{11}	17.70 ± 0.19	4.09 ± 0.49
C_{12}	18.06 ± 0.05	3.95 ± 0.07
C_{13}	16.24 ± 0.32	3.70 ± 0.08
C_{14}	13.54 ± 0.55	3.64 ± 0.12
C_{15}	12.94 ± 0.30	3.69 ± 0.08
C_{16}	11.66 ± 0.42	3.55 ± 0.06
C_{17}	13.19 ± 0.18	3.39 ± 0.04
pristane	9.77 ± 0.56	2.15 ± 0.01
C_{18}	10.55 ± 0.44	3.00 ± 0.05
phytane	12.00 ± 0.34	1.34 ± 0.00
C_{19}	11.88 ± 0.76	3.24 ± 0.02
C_{20}	8.95 ± 0.12	2.73 ± 0.07
C_{21}	9.36 ± 0.44	2.57 ± 0.08
C_{22}	8.24 ± 0.42	2.57 ± 0.07
C_{23}	8.04 ± 0.90	2.28 ± 0.03
C_{24}	5.93 ± 0.80	2.19 ± 0.09
C_{25}	8.09 ± 1.60	1.96 ± 0.05
C_{26}	5.87 ± 0.98	1.80 ± 0.05
Total Saturates	201.99 ± 5.07	51.84 ± 0.90
Naphthalene	2.86 ± 0.07	1.55 ± 0.16
2-MN	3.33 ± 0.52	2.49 ± 0.69
1-MN	2.42 ± 0.78	2.14 ± 0.95
Total MN	5.75 ± 1.15	4.63 ± 1.59
1-Ethyl + 2-Ethyl-naphthalene	*	0.91 ± 0.23
2,6 + 2,7-DMN	2.28 ± 0.20	1.75 ± 0.19
1,3 + 1,6-DMN	1.46 ± 0.19	1.84 ± 0.02
1,7-DMN	4.50 ± 0.15	1.43 ± 0.05
1,4+2,3+1,5-DMN	1.55 ± 0.15	1.69 ± 0.23
1,2-DMN	0.75 ± 0.17	0.68 ± 0.06
Other DMN	**	0.59 ± 0.55
Total DMN	**	7.97 ± 0.95
TMN-1	0.80 ± 0.10	0.72 ± 0.05
TMN-2	*	0.76 ± 0.07
TMN-3	3.47 ± 0.12	0.52 ± 0.36
TMN-4	1.33 ± 0.55	0.68 ± 0.19
2,3,6-TMN	0.89 ± 0.05	0.33 ± 0.08
TMN-5	2.31 ± 0.05	0.82 ± 0.09
Other TMN	**	0.98 ± 0.35
Total TMN	**	4.81 ± 0.94
phenanthrene	*	0.58 ± 0.06
MP-1	*	0.41 ± 0.05
MP-2	*	0.41 ± 0.01
MP-3	*	0.47 ± 0.07
MP-4	*	0.37 ± 0.10
Total MP	*	1.67 ± 0.20
DMP-1	*	1.00 ± 0.04
DMP-2	*	0.30 ± 0.02
DMP-3	*	0.37 ± 0.03
DMP-4	*	0.29 ± 0.03
Total DMP	*	1.96 ± 0.03

MP = methylphenanthrene, DMP = dimethylphenanthrene
MN = methylnaphthalene, DMN = dimethylnaphthalene
TMN = trimethylnaphthalene
*not detectable
**not determined

In addition to the solvent extraction studies, sample preparation using high-pressure liquid chromatography (HPLC) was investigated. Ether extracts of retort water were fractionated according to molecular weight using three series coupled μ-Styragel columns (1000 Å, 500 Å, and 100 Å). A volatile fraction enriched in organonitrogen components was collected based on a calibration curve developed partially from standard organonitrogen compounds constituting five compound classes.

Samples were analyzed by temperature-programmed, glass capillary chromatography employing a nitrogen/phosphorus detector. Several nitrogen compounds have been tentatively identified by retention time matching with known standards and employing the use of a computer library containing the retention indices of 85 organonitrogen compounds.

The gas chromatograms from application of the above method to the pH 8.0 isooctane extract and the HPLC prepared sample are shown in Figure 6.2 and a list of those compounds tentatively identified from these two fractions are listed in Table 6.4. A continued research effort will be directed toward positive identification of compounds tentatively identified in the extraction processes through the use of capillary gas chromatography/mass spectrometry as well as other compounds in the complex mixture for which standards are not commercially available.

CHEMICAL STUDIES - RETORTED SHALE

Cooperative Study with Colorado State University

A field study designed to examine problems associated with revegetation of a retorted shale disposal site and provide a field validation of predictive models for movement of trace metals and organic residuals is being conducted in cooperation with the Range Science Department of Colorado State University. The field plots are located in the Piceance Creek Basin of Colorado and were prepared by removing the topsoil and subsoil to various depths down to a maximum of 60 in. The trenches formed were then backfilled with layers of retorted shale and covered with a soil overburden to depths ranging from 0 to 36 in.

The objectives of the PNL phase of the Piceance Creek Basin Study are to determine: 1) the physicochemical properties and homogeneity of the retorted shale and soil overburdens after placement and 2) the change in solubility and the overall movement of salts, trace metals and organic components within the soil-shale profile.

Sampling and instrumentation began immediately after the fertilizing and seeding operations were completed this fall. Soil cores of the plots were taken using either a 2 or 3 in. coring probe. Porous ceramic cups were installed at various depths in the soil-shale profile. The access tubes will remain capped until the spring of 1978 when they will be used to extract some of the moisture present at various depths in the soil-shale profile.

The initial analyses performed on the core samples will include pH, electrical conductivity, cation exchange capacity, organic carbon and texture. All analyses will be run on both the replaced soil and retorted shale at all sampling depths. After the variability of the plots is determined, subplots will be selected on the basis of soil properties and transport model parameters will be measured as a function of depth. These will include extractable macroelements (exchangeable and soluble) trace metals, organic carbon, cation exchange capacity and $CaCO_3$ content.

At some later time (1 to 3 yr), depending upon moisture movement, these parameters will again be measured and correlated to moisture movement through the profile. Measured and predicted values will then be compared.

In addition to the core samples taken for determining the homogeneity of the test site, intact cores of the profile will also be obtained for the purpose of modeling, in the lab, the transport behavior of soluble components.

Physicochemical Characterization of Retorted Shale

The retorted shale used in these studies was obtained from the Paraho Oil Shale Demonstration Project. It was shipped to PNL in two steel barrels, each containing approximately 180 kg of spent shale. The shale was screened through a 6.3 mm sieve with 29.6% of the material passing through the screen. Approximately 1000 g of the < 6.3 mm and > 6.3 mm material was ball-milled and passed through a 100 mesh (.149 mm) sieve.

Samples (25 g) of both the > 6.3 mm and < 6.3 mm size retorted shale, which had passed through a 100 mesh screen, were weighed into 500 ml Erlenmeyer flasks and allowed to equilibrate with 250 ml of distilled water for 3 days while stirring on a gyratory shaker at 200 rpm. The shale-water suspensions were then filtered through a .45μ filter and stored in glass bottles at 4°C. Preliminary analysis of the filtrate showed the electrical conductivity and pH of the

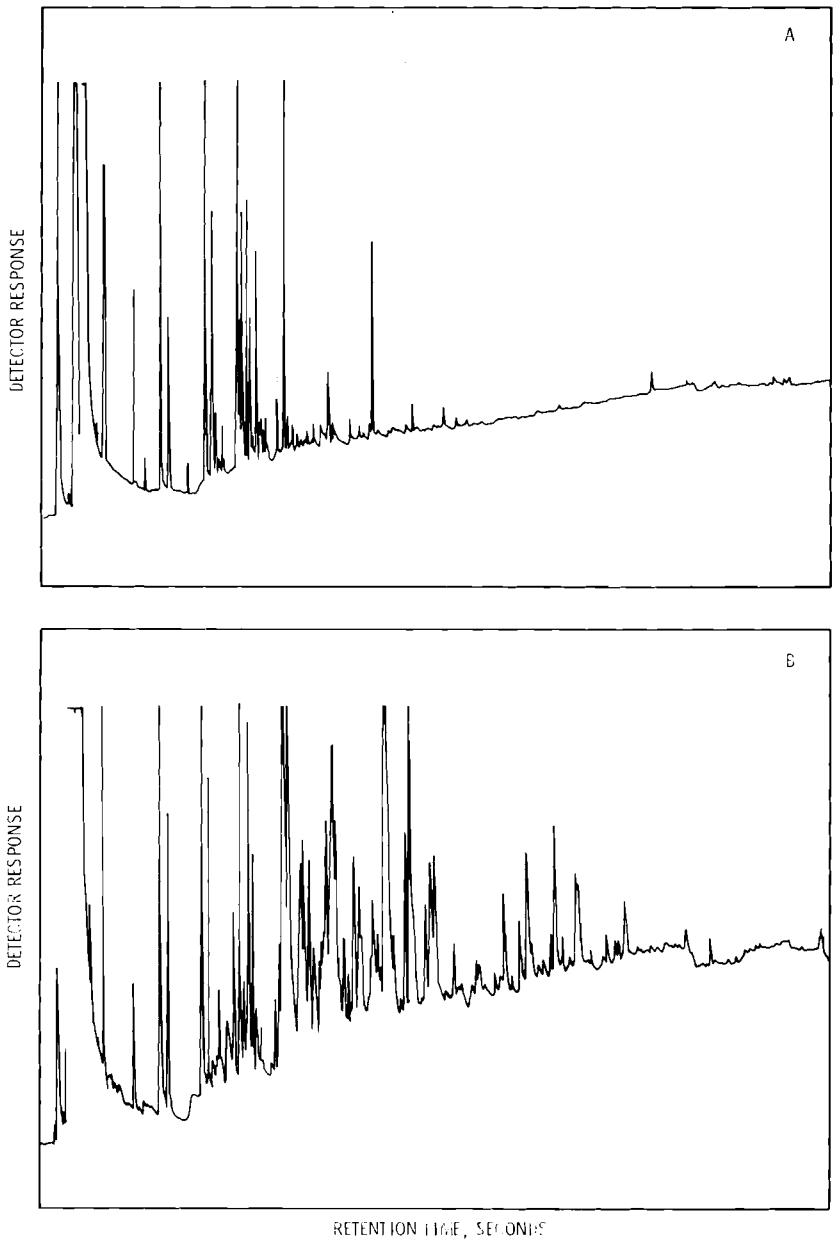


FIGURE 6.2. Gas Capillary Chromatograms of Isooctane pH 8.0 Extract (A) and Prepared HPLC Extract (B) of Retort Water Employing Nitrogen/Phosphorus Detector

< 6.3 mm material to be 3400 µmhos/cm and 12.4, respectively. In contrast, the filtrate from the > 6.3 mm size had an electrical conductivity of 1500 µmhos/cm and a pH of 12.2. This suggests that the soluble salts may be associated with the surface of the retorted shale and that the percentage of fines in a given batch of shale, because of its high surface area, may influence the soluble salt content in leachate.

These results were supported by a second experiment that involved packing pyrex tubing having an I.D. of 1.32 cm with 100 g of air-dry shale (< 100 mesh) which had been mixed with 29 ml of distilled water. The columns were packed to a density of 1.4 g/cm^3. The moisture in the shale was then displaced with ethanol by modifying a procedure used for displacing soil solutions from soil columns.

TABLE 6.4. Organonitrogen Compounds Tentatively Identified[a] in the Isooctane pH 8.0 Extract and HPLC Prepared Extract of Retort Water

Isooctane pH 8.0 Extract:		HPLC Prepared Extract:	
Retention Time	Compound	Retention Time	Compound
6.53	2,4-lutidine	8.34	1,2,5-trimethylpyrrole
7.27	3-ethylpyridine	8.34	2,4,6-trimethylpyridine
7.27	2-isopropylpyridine	8.34	3,4-lutidine
8.34	1,2,5-trimethylpyrrole	13.88	N-ethylaniline
8.34	2,4,6-trimethylpyridine	16.40	2,4-dimethyl aniline
8.34	3,4-lutidine	16.66	P-ethylaniline
11.96	N,N-dimethyl aniline	18.69	2,3-dimethtlaniline
11.96	M-toluidine	18.78	2,3-dimethtlaniline
13.71	N-ethylaniline	19.50	tributylamine
13.93	N-ethylaniline	19.58	tributylamine
16.54	2,4-dimethyl aniline	19.76	tributylamine
16.54	P-ethylaniline	19.87	tributylamine
20.15	N,N-diethylaniline	19.93	tributylamine
23.15	8-methylquinoline	20.28	N,N-diethylaniline
		20.39	N,N-diethylaniline
		21.12	Isoquinoline
		21.48	1-methylindole
		23.18	8-methylquinoline
		26.85	2,6-dimethylquinoline
		26.85	3-methylindole
		26.85	2-methylindole
		36.37	diphenylamine

[a]In some instances, different compounds have been assigned more than one retention time. Final structure and retention time assignment will be based on GC/MS analysis.

The shale solutions displaced by the ethanol were collected in 5 ml fractions and analyzed for the presence of ethanol. In columns packed with shale that were originally < 6.3 mm in size, the electrical conductivity (e.c.) was 12,500 μmhos/cm while the material > 6.3 mm had a value of 9000 μmhos/cm.

A second series of columns were prepared in the same manner as the first; however, water was used as the displacing solution instead of ethanol. The values for pH and e.c. for the first 10 ml were essentially the same as the ethanol displaced solutions.

Work is in progress using dry-packed shale columns through which water is being pumped to approximate percolation; then, the effluent solutions will be collected and analyzed for chemical composition.

Variables being examined include density of shale, size of shale particles, residence time of the percolate, rate of depletion of soluble components, changes in pH, and salt content of percolate. After passing through the column, the percolate will be analyzed for pH, electrical conductivity, organic carbon, and heavy metals in addition to organic chemical characterization of the leachate.

Prior to receiving the retorted shale from Paraho a glass column was packed with < 2 mm size retorted shale from the simulated in situ 10-ton retort at the Laramie Energy Research Center. The height of the shale column was 34 cm with an I.D. of 4.05 cm and packed to a density of 1.06 g/cm^3. Single hole rubber stoppers were used for end plugs and short pieces of glass tubing protruding from the stoppers were used to connect the influent and effluent lines. A layer of glass wool was added to the bottom of the column to prevent soil from plugging the influent line and to facilitate uniform flow away from the source. Upward flow of the influent was utilized to minimize air pockets.

The influent solution (0.01 M, $CaCl_2$, pH 7) was pumped into the column at a rate of 0.168 ml/min with the effluent being collected in approximately 15 ml fractions by a sealed fraction collector to minimize evaporation losses.

Analysis of the effluent (Table 6.5) indicates that an equilibrium of soluble Na and K in the shale effluent is reached when approximately 0.25 to 0.5 of the column volume has been displaced.

It is also interesting to note that for Na, Cu, Cr, As, and total and organic carbon there is a 10- to 12-fold decrease in concentration between the first and fifth fraction collected which involves only approximately 0.1 column volumes. Since the influent contains 0.01 M $CaCl_2$ (400 ppm Ca) the concentration of Ca does increase with each fraction but appears to have reached an equilibrium at approximately 1200 mg/l. The Ca equilibrium value may be the result of the dissolution of $CaSO_4$ believed to be present in the retorted shale.

The information obtained from the columns will be used to provide input for computer models currently being developed to help predict transport behavior in the field. These models will evaluate Langmuir and Freundlich adsorption isotherms in an attempt to evaluate adsorption behavior of shale components. In addition, the nitrogen cycle as it relates to the shale-soil profile will also be studied for incorporation into the model.

Organics and Organic-Metal Complexes in Leachates from Retorted Shale Piles

The development of a viable shale oil industry will have, as a byproduct, large quantities of retorted shale to dispose. Many studies have shown elevated salt

TABLE 6.5. Properties of the Effluent from a Column Packed with < 2 mm Size Retorted Shale from Simulated In Situ Retort and Leached with 0.01M, $CaCl_2$, pH7

Sample	(Col. Vol.)	Na	K	Ca	Mg	Cu	Cr	As	Se	Organic C	Total Carbon
	V/Vo	mg/l									
1	(0.015)	23,925	2,980	565	<0.5	1.5	0.15	0.33	-	1430	1460
3	(0.056)	8,140	1,240	728	<0.5	0.6	0.03	0.15	-	349	360
5	(0.114)	1,980	501	670	<0.5	0.15	0.01	0.03	0.14	120	124
10	(0.282)	200	91	816	<0.5	-	0			20	22
15	(0.442)	180	55	902	<0.5	-	-			10	11
50	(1.57)	171	55	1,338	<0.5	0.02	0.01	0.01	0.01	4	8
99	(3.10 0	207	52	1,127	<0.5	0.02	0.01	0.01	0.01	5	5

contents in leachates from spent shales, but few studies have addressed the potential mobility of organic or organic-metal complexes originating from shale residues or the application of retort waters to the shales.

The studies underway include both laboratory and field investigations. All of the initial studies will emphasize retorted shales and other waste products originating from the Paraho process. Leachates from simulated field plots at Anvil Points were supplied to PNL through the cooperation of the Colorado State University Agronomy Department.

The Colorado State University lysimeters at Anvil Points have several experimental variables included in their research design (water application rate, slope, and varying depths of soil cover). The data from the first year were insufficient to measure the effect of these design variables on leachability of either the organics or organic-metal complexes. Preliminary information which has been obtained from the leachates includes organic C, inorganic C, pH, conductivity, and trace element concentration as a function of collected leachate volume. The organic C contents of leachates from treatments of retorted shale only, retorted shale plus 8 in. of soil overburden, and a soil only control are shown in Figure 6.3. In the retorted shale lysimeters, the organic carbon droped sharply and then increased to approximately 170 μg/ml, remained relatively constant and then droped sharply at 2500-3000 gal or approximately one column volume. One explanation of the initially higher levels and the discontinuity at approximately 500-1000 gal, involves the use of water during compaction of the bottom 3 ft of shale. Increased contact time of the water and compacted retorted shale may have increased the initial levels in the leachate. Further insight into this and other factors involved with trace element complexation will be obtained during the next year and from current laboratory studies.

Development of Chemical Reactions, Stability, and Transport Model of Oil Shale Process Wastes in Soil

A literature review entitled "Ground Disposal of Oil Shale Wastes: A Review with Indexed Annotated Bibliography" has been prepared as a U.S. DOE report. The annotated bibliography, containing over 350 references, includes a general introduction to oil shale technology as well as a detailed discussion of the environmental problems associated with ground disposal of both liquid and solid oil shale wastes. The review should be particularly useful to environmental scientists and engineers, since ground disposal has been proposed for nearly all the shale oil wastes produced. Retorted shales, soluble salts, mining wastes, offgas scrubber, and prerefinery wastes are discussed in terms of quantity produced, proposed disposal methodologies, and potential environmental problems. A manuscript entitled "Oil Shale Technology and the Environmental Impact of Ground Disposal of Oil Shale Wastes: A Review" has been submitted for publication in the open literature. This paper summarizes the bibliographic information and discusses future environmental research needs regarding oil shale ground disposal.

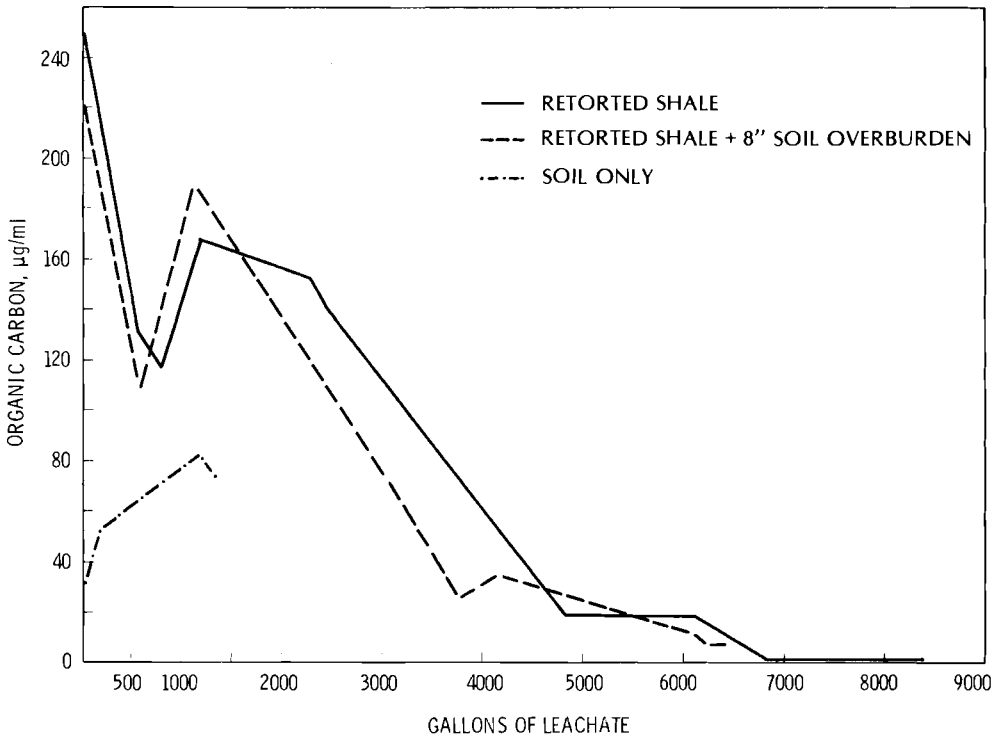


FIGURE 6.3. Organic C From Treatments of Retorted Shale Plus 8 in. of Soil Overburden and a Soil Only Control

SOIL MICROBIAL STUDIES

Microbial Colonization of Retorted Shale

Microbial activity within retorted shale deposits will be one of the principal factors influencing the stability and possible toxicity of organic residues. The initial studies concerning the microbiological aspects of retorted shale have centered around the bacterial colonization of retorted shale and the bacterial utilization of the organic residues in retorted shale as the sole source of carbon and energy.

The Colorado State University lysimeter at Anvil Points (previous section) has been designed to yield information on a number of physical, chemical, and biological questions concerning the disposal of retorted shale. After completion, Colorado River water was applied to the various plots at a rate of 0.5 in./day from June 1 to August 20, 1977. On June 12, samples for microbiological studies were removed from plots containing retorted shale only and an 8 in. soil overburden. Samples of retorted shale and overburden soil were removed from the top 3 cm of each plot and aseptically transported to PNL. Inoculated plates were incubated at 30°C for 7 days before enumeration of bacterial colonies. The results from these studies are listed in Table 6.6. It is apparent from the results that the total heterotrophic bacterial populations of the two samples were essentially equal (1.1×10^7 bacteria/g retorted shale and 1.3×10^7 bacteria/g soil overburden). However, it is of interest that 30% of the colonies observed with the retorted shale

TABLE 6.6. Total Heterotrophic Bacteria in Retorted Shale and Soil Samples

	Retorted Shale		Soil	
Dilution[a]	T.H.[b]	R.C.[c]	T.H.	R.C.
10^{-6}	4	2	4	0
10^{-5}	26	9	33	0
10^{-4}	183	66	204	0

[a] Samples of 0.10 ml were applied to agar-plates
[b] Total heterotrophic bacterial colonies observed (average of 4 plates)
[c] Total colony count of a bacteria which formed characteristically red colonies on the agar medium used

sample resulted from a bacterium which could not be observed on plates from the soil sample.

Results similar to those found in the field studies have been observed in laboratory studies. In these studies, a mixture of 10 g sterile retorted shale, 1 g of Anvil clay soil and 1ℓ of a sterile salts solution was incubated with shaking at 25°C for 7 days. A control mixture which did not contain sterile retorted shale was treated in the same manner. The total heterotrophic population was determined before incubation (0 time) and after the 7-day incubation period. No red colonies representative of the organism observed in retorted shale samples from the lysimeter studies were found associated with the 0 time retorted shale plus soil, 0 time soil-only or the 7-day soil-only nutrient-agar plates used for bacterial enumeration. However, this organism was found at a level of 7.3×10^6 bacteria/ml of mixture when the retorted shale plus soil was examined after 7 days. These results would strongly suggest that this organism is present in Anvil clay soil, though not at a detectable level and that it can utilize retorted shale as a sole source of carbon and energy.

These results emphasize the importance microorganisms have on the stability and possible toxicity of organic residues in retorted shale. This work is continuing and will be expanded to determine the types and numbers of additional microorganisms which develop on retorted shale with environmental aging. Studies are also being initiated to measure the rate and extent of microbial decomposition in retorted shale of total carbon and representative water-soluble residual organic compounds.

Microbiology of Retort Water

Use of retort waters in the compaction of retorted shale at disposal sites has been proposed. Retort waters from the Paraho process have been shown to contain 16% (w/v) soluble organics. The organic components of the retort waters appear to be a unique and complex mixture. Preliminary results indicate that the organics are extremely polar, hydrophylic, poorly extracted into less polar solvents immersible with water.

One of the principal factors influencing the stability and possible toxicity of retort waters within retorted shale disposal sites will be the activity of the indigenous microorganisms. In this regard, studies have been initiated to determine the effects of retort waters on microorganisms and the effects of microorganisms on retort water.

The initial studies have primarily been concerned with the bacterial component of Anvil clay soil, a soil found in the area of proposed retorted shale disposal sites. Bacterial inocula for growth studies consisted of either soil or a washed bacterial suspension prepared from soil. Growth cultures contained in a final volume of 5 ml; salt solution, fungizone (30 µg/mℓ), retort water (0-10%, v/v) and either 100 mg soil or 0.5 ml of washed bacterial suspension which contained the bacterial population that could be prepared from 100 mg of soil. The cultures were prepared in 16 x 150 mm test tubes and were incubated at 25°C as standing cultures. Growth was followed by measuring the increase in optical density at 595 nm with a Baush and Lomb Spectronic 88®.

Bacterial cultures were observed to utilize retort waters as the sole source of C and N. This was especially evident with cultures inoculated with washed bacterial suspensions. Very similar growth patterns were observed with growth cultures containing complete salt solutions and a salts solution minus KNO_3. The presence of phosphates, however, was essential for growth. Cultures containing no phosphate supported only minimal growth.

An initial lag period of no growth was observed with both types of inocula. In both cases, the length of lag increased with increasing concentration of retort water. When growth cultures were inoculated with soil, the correlation between retort water concentration and length of lag was erratic. However, when a washed bacterial suspension was the inoculum, a linear correlation was observed (Figure 6.4). An effect of retort water concentration on maximal growth was also observed. Similar results were again observed with both types of inocula. Maximal growth occurred between 2 and 4% (v/v) retort water. Concentrations of retort water greater than 4% resulted in a substantial decrease in maximal growth. These results suggest that, although bacteria grow on retort water, the retort water has certain toxic and inhibitory properties which can affect growth.

An initial effort was also made to determine which molecular weight fractions of the organics in retort water were being utilized for bacterial growth. Stationary growth cultures which were prepared with 4% retort waters were filtered through 0.4µ filters and the filtrate examined for molecular weight

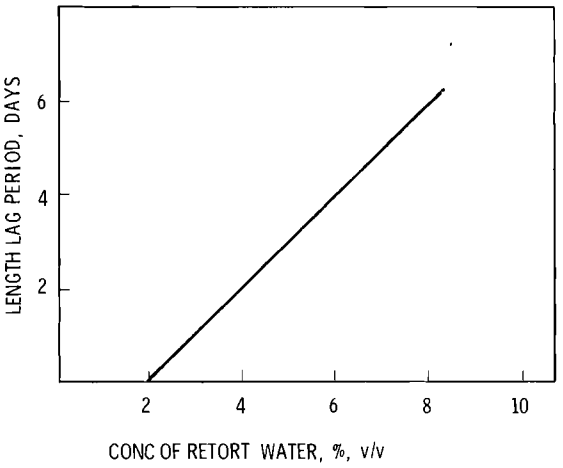


FIGURE 6.4. Concentration Dependence of Retort Water on Bacterial Lag Phase

distribution of the organics by HPLC. A μ-Bondagel column was used and the elution of organics was monitored by U.V. spectroscopy. When the elution patterns derived from uninoculated controls were compared with stationary cultures, the only observable differences occurred below an apparent molecular weight of 400.

These studies are currently being continued and will be expanded to include similar studies on the fungal components of soil, the microbiological colonization of retort water-retorted shale mixtures, the microbiological aspects of various retort water fractions (molecular weight, ionic and organic solvent soluble), and the degradation of important soluble organic compounds identified from retort waters.

PLANT STUDIES

Physiological Effects of Oil Shale Retort Water on Plants

Initial plant studies were designed to evaluate the effects of Paraho retort water on plant processes and therefore enable definitive interpretation of the environmental parameters affecting both physical transport and bioavailability of pollutants associated with oil shale processing. Since an extensive data base is available describing the behavior of metabolic processes in agronomically important species, soybeans were selected for preliminary studies, thus providing a basis for future studies using species indigenous to the Piceance Basin.

Hydroponically grown soybean seedlings (25 day old) were grown in the presence of 0.05, 0.1, 0.5, 1.0, and 5.0% (V:V) retort water pH adjusted to 5.5, to determine overall toxicity response. Plants grown in the presence of > 0.5% retort water exhibited an apparent loss in turgor within 12 hr of treatment, chlorosis occurred within 48 hr. This was followed by death of plants after 3 days for the 1.0 and 5.0% treatments and 7 days for the 0.5% treatment. The apparent toxicity of retort water to plants may result from any number of phenomena. The observed wilting and chlorosis in treated plants would suggest an alteration of membrane permeability and/or mineral imbalance.

To elucidate the mechanism of toxicity in soybean, 34 day-old plants were grown in 0, 0.05, 0.1, and 0.5% retort water for 18 days and the time course of water usage, pH of culture solution, ion uptake, and dry matter production determined. Water usage by plants containing 0, 0.05, and 0.1% retort water exhibited water use rates of 140 and 320 ml/day following 1 and 18 days of treatment, respectively. While water usage by plants on 0.5% retort water was reduced to < 100 ml/day over the 18 day period. Similarily, pH of nutrient solution (measured daily) exhibited a similar response for the 0, 0.05, and 0.1% retort water treatments. However, at 0.5% retort water, instead of showing a characteristic increase in pH from 5.8 to ∿ 7.0 due to preferential cation uptake, pH decreased daily from 5.8 to < 4.0. Dry matter production by shoots was relatively unaffected at 0.05 and 0.1% retort water, while roots showed a 10-20% reduction in accumulated mass. Plants grown in the presence of 0.5% retort water exhibited a 60% reduction in accumulated mass in both shoots and roots. Since the observed effects of retort water appear to have a threshold concentration (in both root medium and tissues) which result in phenotypic changes in the plant (reduced water transport, apparent element toxicity or deficiency, and root absorptive processes), the rate of ion uptake was followed for plants grown in the absence and presence of various concentrations of retort water. These data, when available, should indicate whether the observed phenotypic effects of retort water result from an overall interference with metabolism and absorption processes of the root or the root absorption of some toxic component present in retort water which elicit its effect on accumulation in shoot tissues.

These preliminary studies are designed to determine readily measurable plant parameters

which are affected by components present in retort water. This will provide a basis for screening specific fractions and components in retort water for their bioavailability, and would include both potentially toxic organics and organic ligands capable of complexing and altering the availability of trace elements.

Bioavailability of Organic Constituents Present in Retort Water

Retort water represents a complex mixture of polar organic compounds, which may either be accumulated by plants and/or have a pronounced metabolic effect on the root tissue. Studies have been initiated to determine readily measureable plant properties which can be used in evaluating effects of retort water, retort water fractions, and other oil shale effluents. The objectives are to correlate observed changes in plant pathology and metabolism with the physical properties and chemical composition of shale wastes in order to determine mechanisms of biological transport and toxic action.

Methods developed for determining the bioavailability of trace metal complexes in plants are currently being employed to evaluate the bioavailability of organic components of retort water and eventually specific compounds. Hydroponically grown, 40-day old soybean plants were placed on fresh nutrient solution containing 0 or 0.5% retort water. After 1 hr, plants were decapitated and xylem exudate collected at 1 hr intervals for 6 hr. Xylem exudate represents a transport solution containing not only inorganic ions absorbed by the root, but also a pathway for movement of organic compounds produced in the root. Therefore, analysis of exudate components should provide an indication of changes in the metabolic status of the root resulting from the presence of retort water and/or evidence for the uptake and transport of specific components in retort water.

Molecular weight distributions of organic components in exudates of control and treated plants were determined by high pressure liquid chromatography (HPLC) using a μ-Bondagel column (125 Å). Qualitative changes in concentrations of molecular weight fractions were monitored with a differential refractometer (RI) and UV detector set at 254 nm.

Figure 6.5 shows the molecular weight distributions for both control and treated exudates at 1 and 6 hr intervals. The exudate fractions were normalized relative to the content of the original 0.5% retort water substrate and therefore represent the relative masses of constituents moving in the xylem stream. Although a number of subtle changes were observed in the 12 sample series, one was noteworthy. Fraction A (Figure 6.5) representing a major mass containing component of ∿200 MW, was rapidly lost in control plants, but only slightly reduced in treated plants. Since most of the mass in xylem exudate of legumes is due to organic acids, amides and amino acids, these may be represented by fraction A. And, indeed, analysis of glutamine, citric acid, aspartic acid, malic acid, succinic acid and asparagine showed that these compounds had retention volumes in the range of fraction A. The continued presence of organic acids, amides and amino acids in exudates of treated plants may represent a metabolic disruption by retort water. In fact, a major effect of retort water has been shown to be a significant reduction in dry matter production by roots.

Analytical procedures are currently being extended to characterize compounds present in fraction A and determine the overall effect of retort water on plant metabolism. This will include changes in endogenous compounds, and identification of compounds present in retort water which may be absorbed by roots and transported in the xylem.

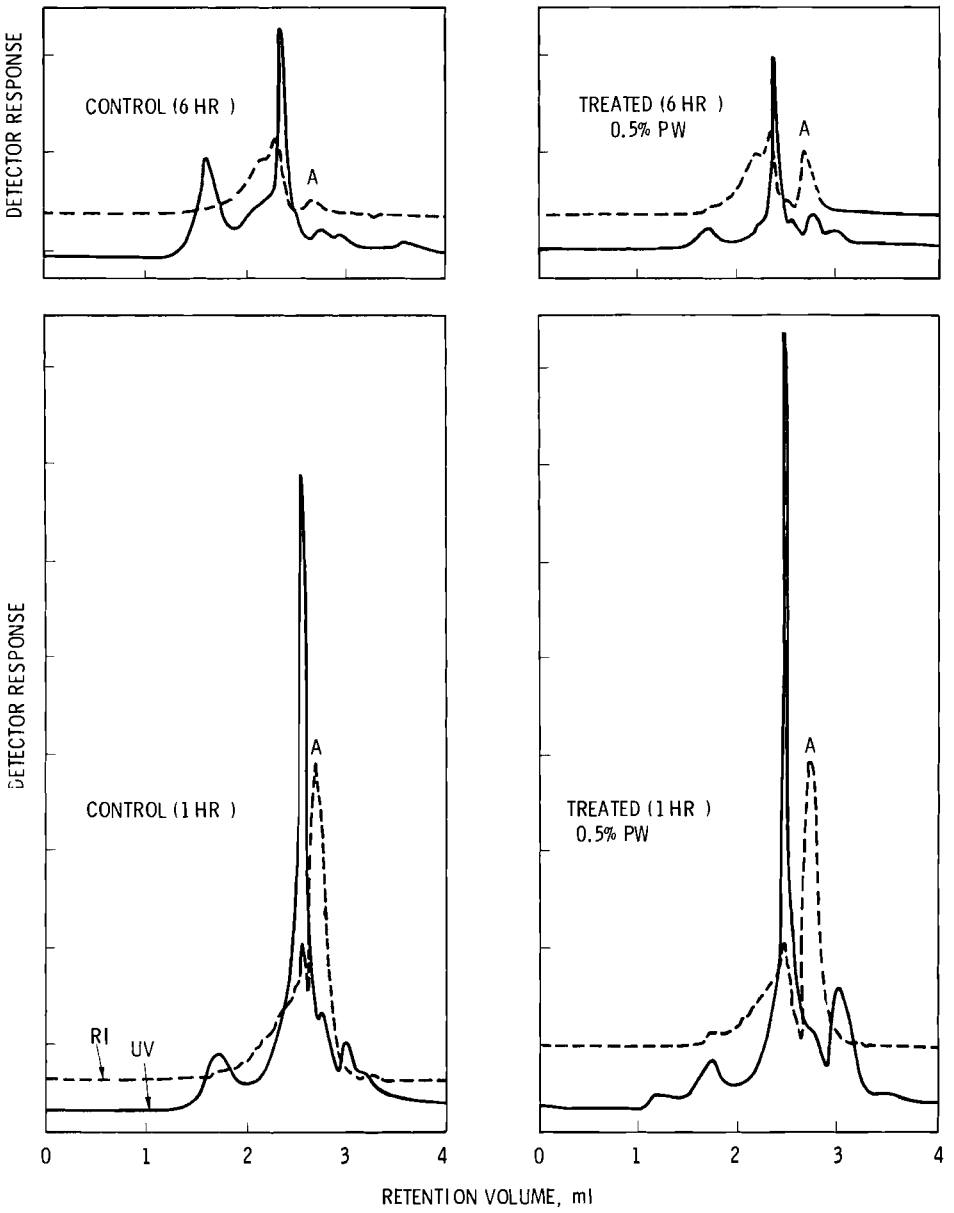


FIGURE 6.5. High Pressure Gel Filtration Chromatograms of Xylem Exudate from Control and Treated Plants at 1 Hour and 6 Hour Intervals

7.0 Multitechnology and Supporting Research Programs

MULTITECHNOLOGY AND SUPPORTING RESEARCH PROGRAMS

- **Analysis of Natural Systems**
- **Quantitative Ecology of Impact Evaluation**
- **Effects of Water Quality Alterations on Fish Behavior**
- **Ecological Effects of Combined Aquatic Stressors**
- **Effects of Energy Systems Effluents on Coastal Ecosystems**
- **Bioavailability of Energy Effluent Materials in Coastal Ecosystems**
- **Marine Chemistry of Energy-Generated Pollutants**
- **In Situ Measurement of Pollutants**

This section includes research efforts that provide information applicable to several presently operating technologies as well as those being investigated for the future. In these technologies the nature of the environmental problem is equally applicable to any one technology; e.g., thermal and chemical pollution of water due to operation of steam electric plants, whether nuclear, fossil fuel, or gas fired; or, the statistical design needed for differentiating a general background of industrial pollution from the contributions, if any, arising from operation of an energy facility.

The two main groups of projects in this category include biomathematical effort represented by the first two projects indicated above and aquatic ecological studies represented by the remaining 5 projects.

Biomathematical Projects: These represent important effort in applying statistical theory to real-world problems such that appropriate error limits may be set when quantitative descriptions of ecological relationships are attempted. Specifically four areas of investigation are underway: 1) the application of sampling theory for the reliable measurement of environmental contamination, 2) methods for the statistical estimation of wildlife populations, 3) the evaluation of simulation models, and 4) statistical approaches for supplying an overview of large environmental data sets (e.g., pre- and post-operational monitoring data from steam-electric plants). In

addition to those projects covered in this section, an additional biomathematical project, described in Section 2, concerns the estimation of pollutant inventory at contaminated sites. The latter project concerns plutonium estimation at the NTS field site, but the approaches are applicable to other pollutants at heavily contaminated sites.

Aquatic Projects: These include both marine and freshwater studies and concern the bulk of Multitechnology Support effort. Generally, this research is directed at understanding the basic processes involved in the functioning of aquatic ecosystems. Several important developments in these programs will more clearly pinpoint the likelihood of actual biological impact from aquatic effluents. One development is the use of behavioral end-points for determining what organisms do in the natural environment. Isopleths of concentration for heat, gas saturation, or chemicals vary with depth and with location from an industrial plant. *In vivo* tagging procedures show that fish and other organisms may, in fact, remain outside the area of impact in certain situations. Another development is the move away from the single component bioassay, which is quite unrealistic, to multiple stressor ("synergistic effects") studies involving response surfaces. For example, we may now pinpoint a certain combination of heat, chlorine, and nickel concentrations, which when acting separately cause little or no mortality, but when acting together, produce 95-100% mortality. In the marine area, emphasis has been placed on establishing the extent of chemical speciation, target tissues, and the extent to which pollutants sorbed to sediments become biologically available. Pertinent related work will be found, also, in Section 2.

- **Analysis of Natural Systems**
- **Quantitative Ecology of Impact Evaluation**

Principal Investigators: L. L. Eberhardt and J. M. Thomas

Technician: M. I. Cochran

These projects have continued to be devoted to developing quantitative methods for broad use in applied and theoretical ecology. The wide array of technologies that must be dealt with in the present energy programs all have impacts on natural populations and man's environment. These impacts cannot effectively be evaluated without considerable improvements in quantitative methodology. We have thus continued an emphasis on sampling technology as it concerns both contaminants and animal populations. Census methods have also been emphasized, since they are essential to environmental studies and a considerable effort has been directed to the evaluation of mechanisms for animal population regulation, since this is a key item in "impacts." Direct evaluation of environmental effects of energy technology has been attacked this year by a detailed review of actual pre- and post-operational data on a number of sites. We thus hope to facilitate the development of efficient and effective field methodology.

Sampling for Contaminants

Our work on sampling for contaminants continues to emphasize issues in sampling methodology. As we have pointed out in several previous reports, it is possible to consider sampling methodology in four broad categories: 1) descriptive sampling, 2) analytical sampling, 3) sampling for spatial pattern, and 4) sampling for modeling. The available textbooks are mainly concerned with the first two categories. The first is the subject of the books with "sampling" in the title, while the second, although mentioned in the recent sampling books, falls between sampling methodology and the domain of "hypothesis-testing" statistical methods, e.g., the analysis of variance.

A technology of sampling for pattern has been evolving in the areas of mineral and hydrocarbon exploration, and has only very recently reached the stage where textbooks are available (Agterberg, 1974). Sampling for modeling has evolved largely in the areas of industrial research and experimentation, with statistical results largely available in journals like Technometrics. We have attempted to discuss some of the problems in applying sampling for pattern to radionuclides and other contaminants in two recent papers, and have given an illustration of the utility of sampling for modeling in two current publications (Eberhardt, 1977a, b).

Our further efforts along the lines of sampling for contaminants have included work on designing improved sampling plans for use at the Nevada Test Site and in DBER-sponsored studies of transuranic elements. This work is described in greater detail elsewhere in

this Annual Report. We prepared a paper concerning the sampling problems associated with the transuranic area (Eberhardt and Gilbert, 1978) and participated in a workshop held at Woods Hole in April 1977. We were involved in a second workshop dealing with sampling issues associated with contamination problems in "microcosm" research, in June 1977, sponsored by Oregon State University, NSF, and EPA.

We expect to continue our current emphasis on sampling for pattern in an effort to improve present methods used in appraising contamination as needed in determining current levels and planning remedial measures, if required, in various field situations. We also plan efforts to provide further designs for the application of the methodology of sampling for modeling to areas of DBER concern, inasmuch as it continues to be apparent that the evident advantages of "optimal" sampling schemes for designing, e.g., uptake and retention studies, seem not to be known to most researchers concerned with such problems.

Mechanisms for Animal Population Regulation

One of the key issues in evaluating the ecological effects of any human activity (such as energy development) is the ability of natural populations to compensate for various stresses. The important element in the ecosystem response is the population of a given species. The ability of populations to respond successfully to man-made insults is undoubtedly a consequence of the self-regulatory mechanism of the particular species. Since there has been a considerable argument as to the nature of these mechanisms for 30 yr or more ("density-dependence" vs "density-independence"), it will be apparent that the exact mechanisms are not known in precise detail. Enough is known, however, to make it evident that a current tendency to discuss energy effects in terms of the "health" of ecosystems is, generally, rather misleading. Prevalence of disease, or other evidence of the well-being of individuals, may be only one facet of the overall complex.

The major applied use of density-dependence as an operational expression of population self-regulation has been in fisheries research and management. In managing certain fisheries, empirical data on abundance are used in the form of a "stock-recruitment" curve. Two models for such curves have been generally in use, and are known as the "Ricker" and "Beverton and Holt" curves. We have reviewed the basis for these two models and found a connection that facilitates the choice of a model for a particular species-population (Eberhardt, 1977c).

Use of stock-recruitment curves depends on the accumulation of observational data extending over intervals measured in decades. When such data are not available, other approaches must be utilized. A number of criteria as to relative condition of a particular population may then be helpful, along with models for population dynamics. We have reviewed this approach for one group of mammalian species (Eberhardt and Siniff, 1977) and find that it may be useful in dealing with populations at or near maximal levels.

The ultimate goal in research on population regulation, and the consequent understanding of the ability of populations to compensate for human impacts, is necessarily a much-improved understanding of the basic regulatory processes. Full understanding will require many years, if progress along these lines in the last 40 or 50 yr is any guide. Much of the available data have been derived in the applied fields of entomology and fisheries management. Most of the species dealt with in those fields exhibit very high reproductive rates. It has seemed worthwhile to consider the accumulated evidence for species with much lower reproductive rates, i.e., the larger mammals. As a starting-place, we have formulated a generalized model for self-regulation in long-lived species. Starting with a population at a very low level of abundance (e.g., after introduction into a new, but suitable, habitat), the model predicts the following sequence of events as regulation develops:

$$\left\{\begin{matrix}\text{Immature}\\ \text{mortality}\\ \text{rates}\\ \text{change}\end{matrix}\right\} > \left\{\begin{matrix}\text{Age of}\\ \text{first}\\ \text{reproduction}\\ \text{shifts}\end{matrix}\right\} >$$

$$\left\{\begin{matrix}\text{Reproductive}\\ \text{rate of}\\ \text{adult females}\\ \text{changes}\end{matrix}\right\} > \left\{\begin{matrix}\text{Adult}\\ \text{mortality}\\ \text{rate}\\ \text{changes}\end{matrix}\right\}$$

Many details of the model remain to be evaluated. A particularly important feature in practical applications is whether or not the full sequence of events is required for regulation of a particular species-population. Our current views appear in Eberhardt (1977d).

Perhaps the most important outcome of our investigation is the emphasis on relative

position of the Maximum Sustainable Yield (MSY) point for long-lived species. The traditional model has been that of the logistic growth curve, for which MSY is at 50% of maximal levels. There is evidence that MSY may be appreciably below 50% for species with high reproductive rates (many species of fish and most insects). However, it now seems likely that many of the large mammals possess MSY points well above 50%. The most obvious implications of such a finding are: 1) optimal harvests are achieved near maximal populations, and 2) these species probably do not possess the same degree of "resilience" to environmental impacts shown by many other populations.

Transect Methods for Population Studies

Our work on transect methods has been continued this year with the preparation of a publication reporting findings subsequent to an earlier review. The focus of the earlier work was on line transects, in which all records of animals seen in the course of following a transect line are used, with auxiliary information on distances and directions, to estimate abundance. We have since broadened the scope of inquiry to include line intersects, for which the basis is interception of an object by a straight line, and modified strip transects, in which only animals or objects within a strip of predetermined width are utilized for census purposes.

The major current problems appear to be those associated with the line transect method as applied to censuses of inanimate objects, or to animals that do not exhibit a characteristic response to the observer's approach (e.g., by "flushing"). As mentioned in last year's Annual Report, we have been recommending a distribution-free method for application in such circumstances. For animals that flush, or otherwise definitely respond to the observer's approach, there is a "robust" method due to D. W. Hayne, with an appreciably smaller variance. Hayne's method is also nonparametric, in the sense that it does not depend on assumption of a particular curve of flushing-distances or sighting-distances. It is based on radial or sighting-distances. There is, however, a good deal of recent evidence that the method does not "work" for inanimate objects or animals that adopt a passive attitude to the observer. In these circumstances, it very likely will be necessary to use right-angle distances and to postulate a sighting curve or visibility curve. The distribution-free method mentioned above does not assume a specific curve, but instead uses, in effect, two "percentiles" of the observed distribution of right-angle distances.

In 1976, we cooperated in the design of a very large-scale line transect survey of porpoise conducted by the National Marine Fisheries Service (NOAA, Department of Commerce). Some five million nautical square miles were subsequently surveyed by two long-range aircraft and two NOAA research vessels. Initial examination of the data (currently under detailed analysis) suggests a further complication in that sighting probabilities appear to be strongly correlated with size of the porpoise school (ranging from single individuals to well over 1,000 animals). There is, thus, a need for further research on this feature, which makes nonparametric estimation questionable due to the small sample sizes available for many of the school sizes. Undoubtedly this phenomenon holds in most of the circumstances where groups of animals or objects are to be tallied. It has been studied for quadrat sampling but not for line transects (Cook and Martin, 1974).

Methods and Sample Sizes for Population Studies

One of the most frequently-asked questions concerning the design of ecological surveys is about sample size. Our efforts to provide some partial answers to that question have been summarized in a manuscript (Eberhardt, 1977e) and a summary of one segment of the data appeared in last year's Annual Report. In those instances where a specific objective for the study can be assumed, and a particular mathematical model postulated for the census method, a fairly specific answer can be provided for the sample-size question. The chief difficulty in these circumstances is usually that of justifying the assumptions on which the mathematical model rests. The weak point here is almost invariably the assumption that each animal in the population being censused has the same probability of inclusion in a sampling made at some particular stage in the census. Evidence that this assumption is often not tenable has been available for some time (cf. Seber, 1973).

In many circumstances, no effort is made to estimate the actual number of animals present on a study area. Instead, some relative measure of abundance is utilized. A variety of such indices are in regular use, based on diverse sources (visual and auditory contact, signs of various kinds, etc.). As a general rule, the process of

converting such indices to measures of absolute abundance ("censuses"), depends on the incorporation of some additional assumptions into a formal model, and perhaps on securing some additional observational data. Although rather widely used in practical management, indices tend to have a somewhat uncertain scientific reputation. Whether this is deserved or not very likely depends on the methods used in obtaining a particular index. When the data are carefully collected according to a well thought out and standardized sampling scheme, it may well be that the only significant difference between an index and a population estimate is that the investigator has been reluctant to invoke an assumption (such as the equal-probability-of-capture assumption mentioned above) that he knows to be faulty. In other cases, however, an index may simply represent haphazardly collected data that are not otherwise usable.

Since the chief problem in population estimation is closely associated with a widely collected and utilized, but seldom analyzed, kind of measurement of abundance, it has seemed to us that further research on the intersection of the two sets is in order. In the paper referenced above (Eberhardt, 1977e), we have assembled some models for indices and data on the relative variability (coefficients of variation) from a wide range of sources. However, time available and length of the present ms (80 typed pages) made it impossible to include an adequate treatment of indices per se. We have begun further research devoted to the analysis of indices and the question of the validity of the assumptions required for absolute measures of abundance.

Biostatistical Aspects of Impact Analysis

We have nearly completed our review of statistical methods that can be applied to actual data sets obtained at nuclear power plants. Our evaluation was designed to assess the advantages, disadvantages, and limitations of various quantitative techniques which we applied *a posteriori* to data sets (actual data analysis was funded by the Nuclear Regulatory Commission). We plan to use this experience so that we can offer methods applicable to impact assessment *a priori*. For such methods to be useful considerable attention to the conduct and design of field studies will be necessary.

We discussed (Annual Report, 1975) our experiences with the statistical analysis of data where the ratio of measurements (usually abundance) of concern at "impacted" sites relative to control sites was the variable. In addition the use of nested unbalanced and one-way analyses of variance were covered. Thomas (1977) discussed the limitations of analysis of covariance, regression, nonparametric methods, arithmetic differencing and simulation models. In Thomas et al. (ms submitted) actual examples of the applications of these methods to data collected at three nuclear power plants is presented.

Thus, we have investigated the various quantitative and qualitative methods we believe to be useful in assessing data from monitoring studies. From this experience we have been able to devise some estimates of sample size needed to detect statistically significant changes for lower trophic level organisms. Using those sample size estimates and one or more of the statistical analyses schemes we have suggested, as well as a field design based on the probable analysis procedure, it is very likely that changes in these biota, attributable to a power plant could be detected (if they exist). Thus, what has been done in environmental monitoring can apparently be done in a much more scientific manner resulting in statistically defensible assertions about lower trophic level biotic changes. These kinds of studies are not enough for most commercially important species because litigation has resulted in a requirement for an assessment of the affect of current insults on future populations. While such an assessment via simulation models may be possible, a quantitative assessment seems beyond the current state-of-the-art in ecology.

Since questions about effects on future (fish, shellfish, etc.) populations are being asked about projects ranging from nuclear power plants to dams and coal mines, and because monitoring data must be collected in an attempt to answer these questions, then a more useful plan for conducting environmental studies needs to be devised. We suggest such a procedure in Figure 7.1.

Projected population effects, about an order of magnitude below those which are considered ecologically detrimental, found year after year (for an agreed period of years) can form a stopping rule for monitoring studies for commercially important species. Thus, after T-years of model refinement and field studies, an impact estimate that is consistently an order of magnitude below an assumed ecologically detrimental level could constitute a point where field studies could be terminated. However, the estimates of key parameters used in the model must be obtained from well-designed field

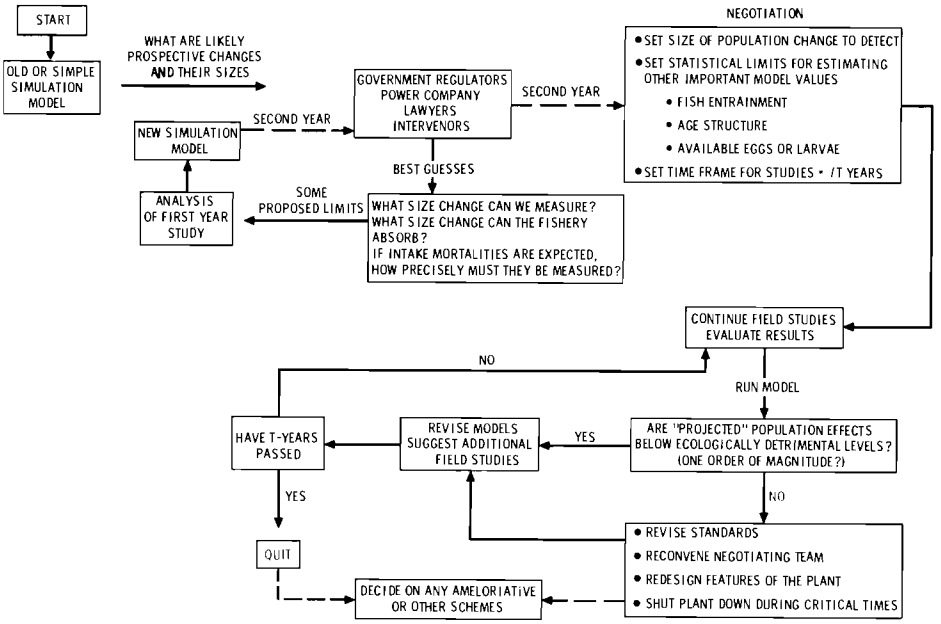


FIGURE 7.1. Plan for Conducting Environmental Studies of a Fishery at a Construction Site

studies conducted so that appropriate error estimates are obtained. These latter studies should be carefully designed and subsequently evaluated in terms of statistical precision (i.e., a fraction of available larvae entrained should be accompanied by a confidence interval and the study repeated several times during the part of the year when the particular species reproduces). Other parameters which are incorporated in models may include age structure and associated age specific mortalities, as well as estimated egg and larval losses from natural mortality. These estimates, as well as others, should be obtained as carefully and accurately as the state-of-the-art allows. Those field studies designed to estimate parameters that, when slightly changed, have a large effect on population "projections," should receive extra statistical design and analysis emphasis. Field studies conducted to determine some of the parameters above will be exceedingly difficult and the level of precision they can be expected to attain may be very low.

It should be stressed that answers to the question (What is the effect on future populations?) probably will not emanate from the program suggested in Figure 7.1 any more than from just sampling fish and shellfish, but some increased understanding both of fundamental ecology and better and more accurate simulation models should result in more accurate projections. Some benefits of the proposal outlined in Figure 7.1 include: 1) a stopping rule where after T-years of support of a good quality field monitoring program the applicant is allowed to terminate the studies, 2) a chance for some of the imaginative scientists currently doing routine monitoring to design and carry out field studies which address key questions in population ecology, and 3) the opportunity for both statisticians and simulation modelers to institute and/or direct (or at least suggest directions for) portions of field work.

Since field monitoring programs were instituted under the mandate to assess impact called for by NEPA in 1969 little, if any, progress has been made toward addressing the crucial population effect issue. The proposal outlined in Figure 7.1 should bring us closer to that goal in the future than the continuation of monitoring studies as now conducted.

Similarity Analyses

With the availability of additional data we have again focused our attention on this research topic (1975 Annual Report). When we incorporated this additional data, we found that the linear relationships we formerly proposed may, in fact, be nonlinear (residual MS of linear and logistic fits are 39 and 8,

respectively). However, if the analysis is conducted by "class of element" (e.g., transition metals, alkaline earths, etc.) it appears that more than one linear relationship may be present.

In a separate development it was brought to our attention that the expression for average half-time is mathematically related to biological equilibrium level (Dr. Charles Mays, personal communication).

This latter variable is calculated using the entire exponential retention function (calculated from whole body counting data after a single oral ingestion of a radionuclide). Preliminary research using average biological half-time (which also is based on the entire exponential retention function) gave results similar to those obtained using biological equilibrium level. We have previously been discouraged by the poor similarity ratios obtained using long-component biological half-time (mostly because interspecies predictions of half times for ecosystem models was the original impetus for the research). Our current results using average half-time, while not definitive, are encouraging, and we are preparing a ms for journal submission.

We believe we have shown that the ideas of similarity analysis appear to be promising both as a method to extrapolate metabolic data among nonruminant species (including man) but also as a device to determine biological half-times for ecosystem analysis. While some complex statistical questions are as yet unresolved, we are led to speculate that similarity ratios for other classes of compounds may be useful.

Marine Mammal Studies

As remarked in last year's Annual Report, we have been cooperating in a variety of marine mammal studies. Origins of our involvement are varied, including field work with Antarctic species some years ago and, more recently, appointment of L. L. Eberhardt to the Scientific Advisory Committee of the U.S. Marine Mammal Commission. Relevance to energy development activities is both direct (oil and mineral exploration, development, and transport; "bioconversion" techniques, etc.) and indirect (models for impacts of human activities on natural populations).

An important aspect of the Marine Mammal Commission work stems from the fact that the enabling legislation, the Marine Mammal Protection Act of 1972, constitutes a pioneering attempt to institute ecosystem management by legislative mandate. In the Act, it is made clear that the intent of Congress is to institute management policies directed towards maintaining "optimum sustainable population" (OSP) levels. Unfortunately, a precise definition of OSP and related terms is not available, so a good deal of effort has been devoted to attempts to achieve working definitions. As time goes on, such definitions attain legal status (through publication in the Federal Register), and become precedents for further legislation. There are, thus, strong reasons for careful attention in these formative stages. Some further details of the problems involved for marine mammals appear in Eberhardt (1977f) and, somewhat more generally, in Eberhardt (1977g).

Our work with Antarctic seals has been summarized in a recent publication Siniff et al. (1977a) and has been extended to a broader consideration of problems in the conservation of polar marine mammals Siniff et al. (1977b). The latter publication includes consideration of research, management, and impacts of economic development. An important distinction exists between the northern polar regions, in which resource exploitation of various kinds has been underway for some time, and the southern regions. In the south, the only past exploitation has been directed towards some of the marine mammals (whales, fur seals, elephant seals), but the prospect of mineral exploitation is now surfacing.

Nonlinear Regression for Ecological Data

As a part of a project to evaluate simulation models we have begun to collect available computer programs which estimate parameters in nonlinear regression models. Several new programs have been received and an extensive bibliography collected. In addition, we have extensively reviewed a code written at Hanford (Learn-Likely) with the aid of Dr. P. Doctor of the Energy Systems Department. Because most such codes are run in a batch mode (usually several times before a satisfactory fit or failure to fit results) and each run is fairly expensive, we have developed an interactive BASIC code (COMP) which runs on both a PDP 11/70 or PDP 11/34 computer. This circumvents both the considerable turnaround time for batch systems and costs/run are extremely low on such minicomputers. Resulting parameter estimates can be used as guesstimates for more precise (double precision accuracy) maxi-computer

codes and to obtain a full suite of statistical results and any diagnostic statements which are printed during the fitting process (suggesting erroneous fits).

The program uses the linearizing or Taylor Series expansion of partial derivatives. Therefore, partial derivatives must be supplied (as well as the function) by the user for any new models not currently contained in the programs. It should be noted that when a linearizing method is used to estimate parameters in a nonlinear model, all the usual procedures of linear regression theory can be applied. However, the results so obtained are only valid insofar as the linearized form approximates the true model.

The output of COMP includes the variance-covariance matrix, t-tests for parameters, Von Neumann's ratio, observed, predicted and residual values, the error mean square, and an optional procedure to evaluate heteroscedasticity. In the latter procedure the absolute values of the residuals are fit using a linear regression model. Thus, deviations which are significantly larger as X increases (usually X is time) may be detected using the linear regression. One explanation for such behavior may be multiplicative rather than additive errors.

The key to the system is the reenterant nature of the curve fitting routine (allowed only with an interpreted language such as BASIC). In general, the user supplies estimates of the parameters for selected model (18 are currently available). The program calculates a requested number of iterative refinements (hopefully improvements) to the parameter estimates in an attempt to minimize the squared deviations between the values predicted by the model and the observed data. During program execution the user can observe whether the results are logical. If not, the process may be stopped, new parameter estimates tried, the current fit examined, the process of iteration started again, or a new model selected. A detailed user's guide for running COMP on the PDP 11/34 and 11/70 has been prepared (Thomas et al., 1977). In addition, we will present our results at the fall DECUS U.S. Symposium to be held in San Diego, California, 29 November - 1 December 1977.

Other Activities

As in the past, a substantial amount of effort has been devoted to consulting and similar technical services to other projects and to a variety of program and project reviews. These include several proposal reviews for DBER, reviews of a number of manuscripts as requested by journal editors, and two published book reviews.

Participation on an ICRP panel (Thomas) continued this year, and a substantial effort was devoted to reviewing a proposed EPA document on plutonium contamination in soils, as well as to consulting on various soil sampling problems locally and at other sites. Several conferences and workshops were attended, both as part of our own program efforts, and to assist other projects in Ecosystems Department.

Our involvement in academic affairs has included various kinds of cooperation with several universities. Dr. Thomas has been serving as a coordinator for a Master's program in biology at the Joint Center for Graduate Study (Richland) and has been appointed as an Adjunct Associate Professor at Washington State University. Dr. Eberhardt continues as Affiliate Professor at the University of Washington.

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• Effects of Water Quality Alterations on Fish Behavior

Principal Investigators: R. H. Gray and J. M. Haynes

Objectives of this project were to study behavioral patterns of ecologically or economically important fish. To minimize potential effects, data on fish behavior are essential for proper design and operation of thermal and chemical outfall and water intake structures. The work is applicable to nuclear, fossil fuel and hydroelectric generating plants.

Pressure-sensitive radiofrequency transmitters were used to monitor behavior of returning adult chinook salmon in relation to gas supersaturated water on the lower Snake River. During fall and spring 1977, 40 fish were monitored between the river mouth and the fourth upstream dam. Equal numbers of fish served as controls. Results indicated swimming depths were significantly shallower in normally saturated water than the previous year in water supersaturated with atmospheric gases.

White sturgeon previously tagged with radio transmitters were monitored to study location, seasonal movement, and behavior of a valued species in a section of the Columbia River influenced by two hydroelectric dams and thermal loading from a nuclear power plant. Additional sturgeon were tagged with temperature-sensitive transmitters and monitored to evaluate thermal preference. Results indicated all size classes preferred free-flowing areas of the river and were relatively inactive in winter. Most movement occurred in summer. Diurnal variations in environmental temperatures and positions of sturgeon tagged with temperature-sensitive transmitters indicated movement into shallow shoreline areas at night.

Data on the instantaneous response of juvenile chinook salmon encountering a simulated river thermal plume interface were published in the open literature and presented and discussed at two symposia.

Depth Distribution of Chinook Salmon in Relation to Gas Supersaturated Water

Studies to monitor behavior of returning adult chinook salmon (Oncorhynchus tshawytscha) in relation to gas supersatured water in the Snake River continued in FY-77. Procedures and rational for the study were detailed in previous Annual Reports. Previous results had indicated fish spent more than 85% of their time below the critical supersaturation zone during a high runoff year.

FY-77 efforts evaluated swimming depths of chinook salmon in the absence of supersaturated water. During fall 1976, with gas saturation values of 101%, 9 fish carried internal pressure-sensitive radio transmitters and metal core anchor tags. Ten control

fish carried only metal core anchor tags. Comparison of travel times and percent passage at upstream dams supported previous conclusions that internal tagging affected upstream movement. Thus, external tags were used in spring 1977. In spring 1977, with gas saturation values of 107%, 30 fish had external pressure transmitters and anchor tags. Additionally, an equal number of fish were anchor tagged only as controls.

Swimming depths of fall 1976 and spring 1977 chinook salmon were shallower (Table 7.3) than those of spring 1976 fish, and the differences were significant ($p<0.05$). Our in situ studies showed adult chinook salmon swam deeper in supersaturated water than in normally saturated water and, thus, avoided potentially lethal conditions. In all cases, mean depth of travel was below the critical zone.

TABLE 7.3. Depth Distribution of Chinook Salmon Tagged Externally or Internally with Pressure Sensitive Radio Transmitters in the Snake River

	Percent Time at Depth			
	Spring 1976[a]		Fall 1976[b]	Spring 1977[c]
Depth, m	External Tags	Internal Tags	Internal Tags	External Tags
0-1	2.9	2.4	22.5	10.9
1-2	7.4	9.4	13.2	19.8
2-3	12.0	17.1	21.2	18.3
3-4	7.1	12.3	9.9	13.4
4-5	21.1	10.8	13.9	9.8
5-6	10.2	11.3	7.9	8.3
6-7	6.5	4.3	4.0	6.0
7-8	4.9	4.2	4.6	4.0
8-9	9.5	7.6	2.0	3.0
9-10	7.1	2.3	0.7	2.8
10-11	5.1	2.8	0.0	1.3
11-12	4.3	4.6	0.0	0.8
12-13	0.6	8.9	0.0	0.8
>13	1.1	1.9	0.0	0.9
Number of Fish	12	14	9	30
Number of Depth Recordings	1444	1246	151	2761
Mean Depth	5.7	5.8	3.1	3.8

[a] Atmospheric Gas Saturation in the Snake River ranged from 124-129%

[b] Atmospheric Gas Saturation in the Snake River was about 101%

[c] Atmospheric Gas Saturation in the Snake River ranged from 106-107%

Results of this work have been published in the open literature, submitted for publication, reported at scientific meetings and will appear in a doctoral dissertation. Additional manuscripts are in preparation.

Seasonal Location, Distribution and Movement of White Sturgeon in the Hanford Reach of the Columbia River

Studies to monitor seasonal location, distribution and movement of white sturgeon (Acipenser transmontanus) in the Hanford Reach of the Columbia River continued in FY-77. Procedures and rationale for this study were detailed in last year's Annual Report. This year, additional fish were instrumented with radio transmitters yielding information on position and 12 sturgeon were tagged with temperature sensitive transmitters to evaluate thermal preference in this section of the Columbia River. Monitoring of these and previously instrumented sturgeon occurred in FY-77 and will continue for the duration of transmitter life. Results indicate all size classes prefer free-flowing areas of the river and are relatively inactive in winter. Most movement occurs in summer. Preliminary temperature data show a diurnal cycle and suggest sturgeon spend the evening in shallow, warmer shoreline areas possibly to feed.

Results of this work have been submitted for open literature publication and will appear in a doctoral dissertation. Additional manuscripts are in preparation.

Initial Response of Juvenile Chinook Salmon to a Simulated River Thermal Plume Interface

Experiments to evaluate initial responses of juvenile chinook salmon (Oncorhynchus tshawytscha) to simulated thermal effluent were terminated at the end of FY-76 and results given in last year's Annual Report. Efforts continued in FY-77 to publish and discuss the implications of project results in the open literature and two symposia.

- **Ecological Effects of Combined Aquatic Stressors**

Principal Investigators: D. R. Anderson, C. D. Becker and M. J. Schneider

Technical Assistance: S. A. Barraclough and M. L. Wolford

The title of this program represents two major research areas formally separate projects, now combined under one title. This combination represents a recognition of the close relationship of the information each is generating. The program titles formerly used were: "Effects of Thermal Discharges on Aquatic Biota" and "Combined Effects of Waste Heat and Environmental Factors Acting in Concert."

The former was designed to measure thermal injury, latent and manifest, in fish and other organisms important in aquatic food chains. The latter makes use of such thermal data to determine when and how multiple pollutants, including thermal increments, affect fish. A key concept emerging from these studies is that of multiple stressors; i.e., where several pollutants each, at "sublethal" concentrations, act together to cause significant abnormality or death. This is probably the typical situation where environmental impact becomes manifest.

The major effort in the former project area has been the publication of information gathered to date. One paper has been published this year, one submitted, and a third is undergoing review. A hypothesis which could delineate the stamina limitation of fish under various sublethal stress is currently being tested.

In the latter project, the studies of combined effects of multiple chemical and thermal parameters, in a single bioassay, represent a relatively new area of research. One of the very interesting findings in this research is a heightened toxicity of Ni caused by rather low concentrations of chlorine. Work is based on flow-through bioassays, using multiple temperatures and chemical species. Rainbow trout, Salmo gairdneri, and coho salmon, Oncorhynchus kisutch, were assayed. During the next phase of the program, sublethal parameters such as growth, bioaccumulation and tissue destruction will be measured in response to the low level, sublethal toxicant dosages.

Effects of Water Temperature on Excercise, Blood Glucose and Lactate of Rainbow Trout

A variety of environmental factors are known to influence the metabolic activity of fish and, therefore, their response to stress. Two of the more important factors are water temperature and the level of fish activity. The purpose of this study is to investigate the interrelationships of fatigue and water temperature on fish tolerance to stress. This project assesses the combined effect of induced swimming and exposure to thermal stress on the performance ability of fish and physiological parameters, blood lactate and glucose.

The major effort of the past year has been the publication of the large volume of information developed by the research efforts to date. A paper entitled "Trap Tank for Nontraumatic Serial Sampling of Fish Stocks in Physiological Studies" was published in the July 1977 issue of the Progressive Fish Culturist.

The construction details of a device which allows unbiased sampling of fish for physiological studies are described. The device consists of two rectangular tanks, connected by a tunnel, which contain a trap door in the floor. A single fish swimming through the tunnel can be removed by opening the trap door; the tunnel is isolated from the two tanks by flap-valve doors when the trap door opens. Blood glucose and lactate levels of fish serially removed from the tank remain stable throughout the sampling day.

A second paper entitled "Effects of Water Temperature and Exercise of Fish Energy Metabolism" has been submitted for publication and is currently undergoing review. This paper discusses the combined effect of acute sublethal heat stress and enforced swimming acting in concert. Rainbow trout, Salmo gairdneri, acclimated to 12°C were exposed to 12, 17, 22 and 27°C while simultaneously responding to two levels of exercise, static (no swimming) and swimming at cruising speed. Measurements of blood glucose and lactate levels were taken at the end of a 30 min swimming period. The blood glucose levels at 12 and 17°C appear unaffected by temperature; at 22 and 27°C the glucose elevation is significant. A similar pattern was seen in the lactate determinations with significant elevation occuring only at 27°C. An interaction of exercise and temperature was found in the blood glucose and in the performance of the trout. It is hypothesized that the failure of trout to complete the exercise-temperature regimen may be due to the development of tissue hypoxia rather than the buildup of blood lactate.

The affect of temperature acclimation on the blood chemistry parameters, glucose and lactate, are discussed in a third paper. This manuscript has been drafted and is currently undergoing review by appropriate PNL staff. We anticipate submittal of the manuscript before the end of the calendar year.

An important hypothesis was developed during the analysis of the data discussed in the second paper (abstracted above). The development of this hypothesis is a significant point of progress because if verified, it will explain the physiological basis for failure of fish stamina under combined stress of swimming and elevated temperature. This information would provide the necessary data base to substantiate engineering decisions regarding allowable discharge temperatures in flowing waters. It is the testing of this hypothesis that we will next address. Should the hypothesis prove correct, it will be useful to the understanding of fish performance capability under physiological stress, particularly stressors other than temperature and exercise which interfere with oxygen uptake and transfer, e.g., chemical toxicants.

The Combined Effects of Nickel, Chlorine and Temperature on Rainbow Trout and Coho Salmon

The combined effects program is designed to quantify the combined action of thermal stress and two chemical pollutants, nickel and chlorine, on the physiology of rainbow trout, Salmo gairdneri, and coho salmon, Oncorhynchus kisutch. These chemicals were selected because of chlorine's use as a biocide and the use of nickel in a large percentage of the types of condenser tubing in steam electric stations. The affect of temperature on the toxicity of these two components is being studied due to the thermal component normally associated with effluents from steam electric stations.

The use of the combined effects approach is estimating the toxicity of multiple chemical and thermal parameters in a more realistic approach than a single factor approach in estimating the actual toxicity of effluents which contain multiple toxicants. The objective of the program is to study the interactions and mechanism of toxic action of nickel and chlorine at several water temperatures on coho salmon and rainbow trout. Acute flow-through bioassays are underway in a modified Mount-Brungs proportional dilluter.

Sublethal parameters of growth, bioaccumulation and tissue destruction are being measured in response to low-level toxicant dosage. A factorial experimental design including three levels of chlorine and three levels of nickel at different temperatures is being used. This enables statistical evaluation of each effect and the interaction of the combined effect of the toxicants and temperature. Both 96-hr (4 day) and 336-hr (14 day) bioassays are being conducted. Samples of gill, liver, and muscle tissues are being taken to monitor bioaccumulation of nickel tissue destruction.

A synergistic effect of chlorine and nickel toxicity on rainbow trout has been demonstrated. Concentrations of 0.05 ppm, Total Residual Chlorine (TRCl) result in fish mortality of 5%. Nickel concentrations ranging from 4 to 8.5 ppm result in 0% mortality. When these levels are combined the resultant mortality ranges from 95-100% of the fish exposed during a 96-hr bioassay. This is the first incidence reported in the literature of a chlorine-nickel synergistic toxic effect.

Evaluation of the "Critical Thermal Maximum"

Experiments were completed in FY-77 to critically evaluate the effects of increasing water temperatures at different rates on Critical Thermal Maximum (CTM) determinations by using representative warmwater and coldwater species of fish.

Rationale and results with pumpkinseed sunfish (Lepomis gibbosus), were presented in the previous Annual Report. Results with fingerling coho salmon (Oncorhynchus kisutch), data analysis, and potential applications of CTM data are summed here.

The CTM is a parameter used to determine the thermal resistance of cold blooded organisms by increasing (or decreasing) the temperature at a constant rate. The point where "locomotor" activity becomes disorganized and the animal loses its ability to escape from conditions that promptly lead to its death is the CTM.

Since various investigators have used different rates of temperature change to determine CTM's of fish, results are not strictly comparable. Standardization of methods is particularly important when the CTM is used to quantify sublethal effects on fish exposed to environmental stresses. We are concerned, in other phases of our program, with potential chemical contaminants derived from development and utilization of coal, tars, and oil shale as energy sources.

In our studies with juvenile coho salmon (a coldwater species), acclimations were at 5 and 15°C rather than at 10 and 20°C as used for pumpkinseed sunfish (a warmwater species). Other conditions were identical. Controlled temperature increases were 1, 6, 18, 30 and 60°C/hr. The data obtained were time and temperature of loss of equilibrium (LE) and death (D). Multiple comparisons of the derived CTM means were made to aid in evaluating temperature increase rates, Table 7.4.

TABLE 7.4. Comparison of CTM Means for Juvenile Coho Salmon by Duncan's New Multiple Range Test[a]

Acclimation Temperature	Endpoint	CTM Means at Indicated Heating Rate				
		1°C/hr	6°C/hr	18°C/hr	30°C/hr	60°C/hr
5°C	Loss of Equilibrium	25.01	24.84	25.32	25.87	25.80
	Death	25.65	26.09	27.51	28.18	29.16
15°C	Loss of Equilibrium	27.22	28.13	28.70	29.16	29.63
	Death	27.56	28.75	29.72	30.05	31.15

[a]Any means not underscored by the same line are significantly different at the 0.05 level of significance

LE temperature means for fish acclimated at 5°C were not significantly different at heating rates of 1, 6 and 18°C/hr, nor were the 30 and 60°C/hr rates significantly different from each other. But the 1, 6 and 18°C/hr heating rates differed significantly from the 30 and 60°C/hr rates. LE temperature means for fish acclimated at 15°C were all significantly different from each other.

D temperature means for fish acclimated at 5°C were all significantly different from each other irrespective of heating rate; the same is true for fish acclimated at 15°C. This assessment confirms that standardization of methods used in CTM determinations is required.

• Effects of Energy Systems Effluents on Coastal Ecosystems

Principal Investigators: G. Roesijadi and J. S. Young

The primary concern of this program is to determine the effects of potential toxicants resulting from energy technologies. This information is needed to: 1) assess the need for further study on the bioavailability and cycling of the contaminant, 2) provide a defined endpoint by which bioavailability can be measured, 3) determine the lowest concentrations that can be safely released into the environment to allow for minimal impact and maximum use of our resources, and 4) describe the effects that are manifested and how they contribute to an ecosystem. During the past year, work has centered on the two major chemical contaminants, chlorine and copper, released by the operation of cooling systems.

Chlorine

The studies on the effects of seawater chlorination on marine animals to date have primarily been concerned with the relative sensitivities of a number of Pacific Northwest marine species to total residual oxidants (TRO). The species we have tested had a wide range of sensitivity, with 96-hr LC_{50} values ranging from 0.032-1.418 mg/ℓ TRO (Table 7.5). Ontogenetic effects have also been identified in related studies. For example, morphological deformities were observed in juvenile shiner perch exposed to TRO, and exposure to TRO delayed hatching of coon stripe shrimp eggs.

In order to understand the reason for this wide variation in sensitivity and the mechanisms involved in producing adverse effects, research has been initiated to examine the physiological responses of organisms exposed to chlorinated seawater. Integrated with these research studies are the identification of biologically active compounds produced by seawater chlorination.

Existing data indicate that the presence of TRO is disruptive to the normal function of major physiological systems, particularly those associated with gas and ion transport such as gills and blood. To date, most of

TABLE 7.5. The Relative Sensitivity of 15 Fishes and Invertebrates to Chlorinated Seawater as Indicated by 96-hr LC50 Values

Name [a]	Number of Valid Tests	96-hr LC50, mg/ℓ TRO	95% Fiducial Limits, mg/ℓ TRO	Probit Regression Line Slope
Coho salmon, j [b] *Oncorhynchus kisutch*	3	0.032	0.026-0.038	21.2
Chinook salmon, j *Oncorhynchus tshawytscha*	2	- [c]	-	-
Pink salmon, j *Oncorhynchus gorbuscha*	3	-	-	-
Pacific Herring, j *Clupea harengus*	2	0.065	0.033-0.097	14.7
Shiner perch, j & a *Cymatogaster aggregata*	5	0.071	0.045-0.098	23.5
English sole, j *Parophyrys vetulus*	3	0.073	0.044-0.103	10.3
Pacific sand lance, j & a *Ammodytes hexapterus*	6	0.082	0.062-0.102	21.9
Shrimp, a *Pandalus goniurus*	3	0.090	0.063-0.119	27.8
Shrimp, a *Crangon nigricauda*	6	0.134	0.118-0.151	17.0
Amphipod, a *Anonyx* sp.	4	0.145	0.118-0.173	12.8
Nysid, a *Neomysis* sp.	4	0.162	0.150-0.175	14.1
Threespine stickleback, j & a *Gasterosteus aculeatus*	3	0.167	0.141-0.193	21.6
Coon stripe shrimp, j & a *Pandalus danae*	4	0.178	0.159-0.199	19.0
Amphipod, j *Pontogeneia* sp.	5	0.687	0.583-0.864	27.0
Shore crab, j & a *Hemigrapsus nudus* and *H. Oregonensis*	5	1.418	1.240-1.530	14.2

[a] Identified using keys in Hart and Kozloff.
[b] j = juvenile, a = adults
[c] The low number of data points between 0 and 100% mortality prevented calculating LC50 values for chinook and pink salmon. The mortality data obtained indicated sensitivity close to the coho and greater than the Pacific herring.

the research along these lines has been confined to freshwater systems. These studies examine the effects of seawater chlorination and specific compounds resulting from chlorination activities. Studies have been initiated on the affects of chlorinated seawater on ionic/osmoregulatory and respiratory physiology of selected marine invertebrates.

Currently, the relationship between ammonia excretion and exposure to chlorinated seawater in a marine crustacean, Cancer productus, the red rock crab is being investigated. Other work has demonstrated that some compounds created from the chlorination process (particularly chloramine and bromamines) can be harmful to marine organisms. These latter compounds result from the reaction of chlorine and bromine with ammonia. Since ammonia is an excretory product of aquatic animals, it is possible that microenvironments may exist in the region of ammonia excreting organs, such as gills, in which halomines (formed by reactions with ammonia and chlorinated seawater) may be present in relatively high concentrations. At the present time, the relationship between ammonia excretion and exposure to chlorinated seawater is being studied. Studies to identify halogenated organic compounds formed as a result of seawater chlorination, under sponsorship of the Nuclear Regulatory Commission, are being closely integrated with these effects studies.

Copper

The primary concern about a bioavailable contaminant is whether it may cause an adverse effect to a marine organism. If it does, then the next step is to assess the potential population and community level response that will result from the individual effect. At this time, these studies are concentrated on the individual response to elevated copper levels with two organisms, Eudistylia vancouveri and Pandalus danae. These organisms have been chosen as representatives of two physiological types; those that do not have a copper based respiratory pigment, and those that have Cu^{++} in the transport of oxygen.

Eudistylia vancouveri, a sedentary tube dweller with an anterior display of pinnate gills, is found in coarse sediments or attached to local pilings. To date, we have determined the following: 1) background tissue levels of copper range from 3 to 9 µg/g in the body and 5 to 18 µg/g in the gills; 2) copper begins to accumulate in tissues upon exposure to concentrations between 3 and 6 µg/ℓ Cu^{++} (Figure 7.2); 3) gills accumulate more copper than does the body; 4) at increasing concentrations of copper, chemical injury is manifested by corresponding increases in gill necrosis and autotomy; and 5) E. vancouveri withdraws into its tube more frequently upon exposure to copper.

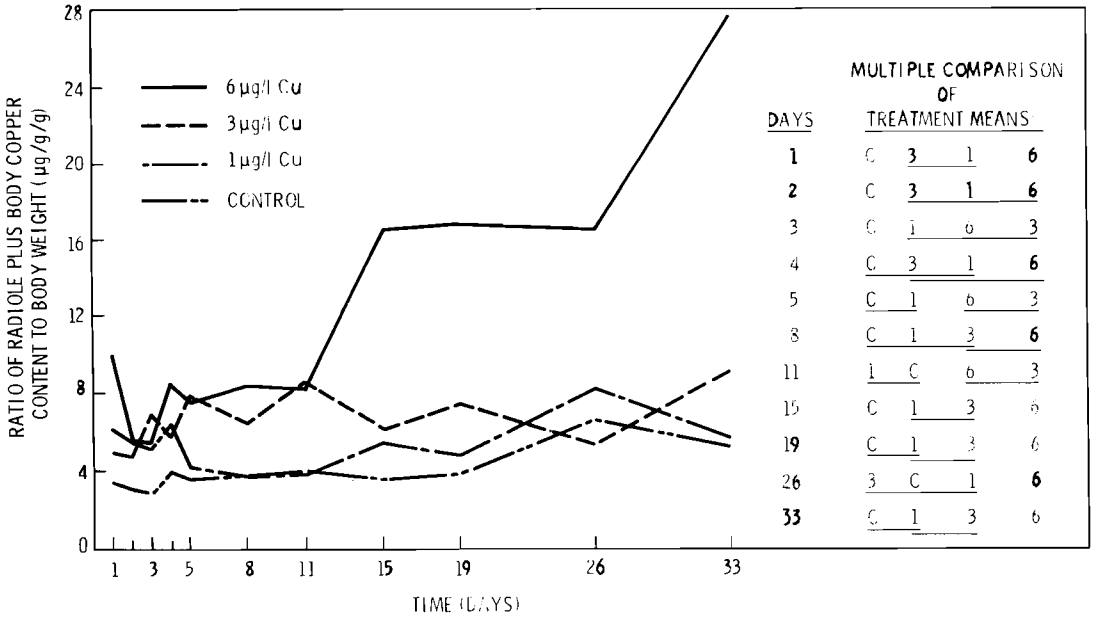


FIGURE 7.2. Ratio of the Sum of Gill and Body Copper Contents to Body Weight After Exposure to Copper - Means and Their Multiple Comparisons Over Time

A major task in FY-78 is to determine a threshold copper level for gill injury in Eudistylia, copper uptake rates and accumulation as injury progresses, its histopathology, and probably its ultrastructural characteristics. The last will help define the toxic mechanism. Evidence indicates cell membrane breakdown and lysosome rupture. In time, this will be compared to ultrastructure of gill injury in Pandalus.

Other tasks include the examination of the behavioral response to copper in Eudistylia and 1 yr exposure of Eudistylia at low level copper to examine long-term accumulation and chemical injury. Under winter conditions Eudistylia begins to accumulate increasing copper with time, at a concentration between 3 and 6 µg/ℓ Cu^{++}. It is essential to determine whether a steady state will occur with time or whether body levels will continue to increase. The tissue level responses such as gill necrosis will also be examined, along with the organisms ability to avoid exposure via behavioral responses.

For the copper regulator Pandalus, the following have been established: 1) the 96-hr LC_{50} for Cu^{++} is 72 µg/ℓ; 2) all tissues examined, except muscle, accumulate copper. Gills and the hepatopancreas gain the highest levels during a given exposure period; 3) the level of copper accumulated in tissues increases with its exposure concentration; 4) this response (Figure 7.3) has been described histologically along with an inflammatory response. Gill necrosis developed in one week at 50 µg/ℓ Cu^{++}; 5) necrotized gill tissue upon elimination of the moult is not regenerated with continued exposure to copper.

Since Pandalus is mobile and it may move in and out of copper contaminated areas studies are underway to determine how well it can recover by examining copper accumulation and depuration. Research completed this past year indicates that depuration is rapid in all tissues except the hepatopancreas, and that a major release mechanism is through the moult. Studies in FY-78 will look at other tissue level responses to determine their role in the movement and accumulation of copper in Pandalus. Pandalus larvae developed through all stages at 10 µg/ℓ Cu^{++}, but not at 20 µg/ℓ. Experiments are planned to define the threshold for these larvae since larvae of many aquatic animals have been shown to be more sensitive to toxicants than adults.

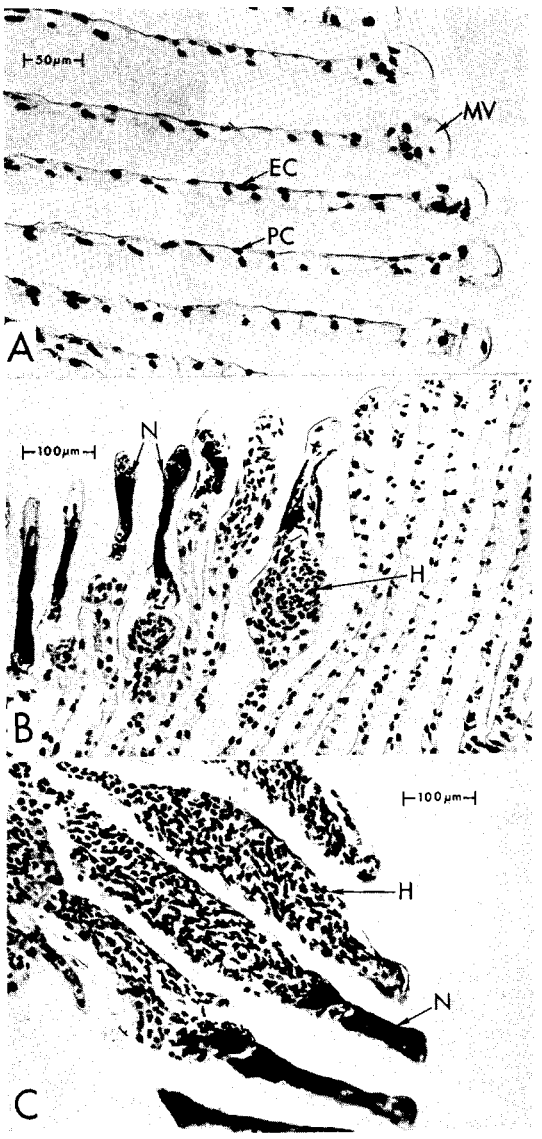


FIGURE 7.3. (A) Section of Gill from Control Shrimp; MV = marginal vessel, EC = epithelial cell, PC = pillar cell. (B) Section of Gill from Shrimp Exposed to 20 ppb Cu, Showing Affected Lamellae Adjacent to Normal Appearing Lamellae; H = hemocytes, N = necrosis. (C) Section of Affected Gill Showing Extensive Hemocyte Infiltration.

• Bioavailability of Energy Effluent Materials in Coastal Ecosystems

Principal Investigators: E. A. Crecelius, C. I. Gibson, L. D. Kannberg, J. E. Rogers, R. L. Schmidt, J. S. Young, K. H. Abel, and D. E. Robertson

The bioavailability program is engaged in research to provide an understanding of the processes involved in the cycling of materials resulting from energy technologies in the marine coastal ecosystem. The primary goal of the program is to define the fate and ultimate ecological consequences of energy technology-produced materials in the coastal ecosystem.

One of the major problems limiting the ability to predict the effects of a contaminant in the marine environment is the inability to equate an analytically-defined fraction of the total amount of material in the environment with a biological response. This problem is particularly limiting for long-term effects where chemical modification is likely to occur because of chemical, physical and biological influences. The bioavailability program has begun a number of tasks to solve this problem and provide an understanding of the processes and mechanisms that control the bioavailability and cycling of materials in the coastal ecosystem.

This year, our work was concerned with: 1) the chemical forms and quantities of radionuclides being released by a nuclear-fueled steam electric station, 2) the chemical form and reactions of copper, 3) the biological uptake of a specific form of copper over long periods of time, 4) the reaction of Cl_2 in seawater and its resulting compounds, 5) the sediment-water interactions of metals in natural and altered systems, and, 6) the development of a model capable of including physical, chemical; and biological interactions in predicting the dispersion of contaminants from a point or multiple sources.

Radionuclide Studies

Physicochemical characterization studies of aqueous process streams at San Onofre Generating Station Unit #1 were conducted to provide source term information concerning the forms of radionuclides being released into the marine environment. This information is needed for future field sampling efforts and laboratory studies of the bioavailability of these materials.

Sampling was conducted on the primary coolant, secondary steam condensate, processed low level wastes and tertiary coolant at several different times in the nuclear fuel cycle. Radionuclides were partitioned into particulate, cationic, anionic, and nonionic species in the reactor process streams, and into particulate and soluble species in the tertiary seawater coolant. Characterization of the particulate species included a detailed size distribution. No measurable radioactivity was found upon examination by a large volume concentration technique of secondary steam condensate. Similarly, no observable activity was found in the tertiary coolant during normal operations when the

reactor was not discharging low level waste. Both of these samples indicate negligible cross-leakage between coolant loops.

Examination of low-level waste immediately prior to release has indicated extremely variable radionuclide compositon and variable and sometimes unexpected physicochemical forms in the waste. Predominant γ emitting nuclides in the low level waste are ^{57,58}, ^{60}Co, ^{110m}Ag, ^{134,137}Cs and ^{124,125}Sb. Detailed examination of particulate material in the low level waste has shown the presence of a wide spectrum of particulates in the low level waste and also significant variability in the particle size spectrum during a release.

An investigation at San Onofre Nuclear Generating Station included evaluation of the efficiency of our in-plant water sampler for soluble species. Sampler efficiency for ionic species was evaluated through the use of multiple resin beds. Cation removal was demonstrated to be quantitative in the first resin bed for all cationic species. Anion removal was less efficient and ranged from 85% for cobalt species (^{58}Co, ^{60}Co), to 75% for ^{124}Sb and ^{125}Sb.

Large volume water sampling conducted at the tertiary coolant outfall has shown detectable radioactivity during low-level releases. Preliminary measurement and mass balance indicate either: 1) incomplete mixing at our sampling point with rapid adsorption to particulate matter present in seawater, or 2) less rapid phase redistribution upon release, but a gradual increase in particulate-associated radioactivity in the area of the outfall.

Further studies are underway to examine the release of radionuclides from the particulate phase and the behavior of soluble rad waste upon mixing with seawater. The results of these studies will give insight into environmental behavior after release of the low-level waste to the marine system and further elucidate the processes occurring in the outfall.

Copper Studies

Copper is a product of energy-related effluents that enter the marine ecosystem. The recent refinement by us and others of the use of Anodic Stripping Voltammetry (ASV), an electrochemical analysis to measure and characterize copper in its dissolved chemical forms, has enabled us to further define the form of copper existing in natural systems and monitor our controlled systems. For example, we have found by ASV that "clean" Pacific Northwest marine waters contain from 0.04 to 0.40 µg/ℓ total copper. Waters near the Seattle industrial complexes contained 1.4 µg/ℓ. Little of the copper is in the ionic (Cu^{++}) form; most is complexed with other molecules. The seawater used in our laboratory has an excess complexing capacity of between 10 and 20 µg Cu/ℓ, depending on season and other water conditions.

The relationship of electrochemically measured copper forms and their availability to marine organisms was investigated to determine how the complexation capacity of the seawater influences copper uptake. This information is needed to assess and predict the effects of metals from energy-related effluents. Copper is a micronutrient that is accumulated by organisms, but regulated at a constant body level. However, above a certain threshold concentration, copper accumulation increases with time, provided the metal is in an available form. In general, the literature has not identified the form of copper that is responsible for measured effects. When a seawater/copper solution is not aged, accumulation is greater than if it is aged, which indicates a change in the copper form, probably from an ionic form to a complexed one. The bonding of weakly-complexed copper may be broken by an organism to make it available or may act as a copper carrier. The objective is to define these relationships, understand some of the mechanisms of metal bioavailability and, thereby, be better able to produce long-term consequences of such ions in energy effluents.

Chlorine Studies

A number of experiments were conducted to determine the relationship of our measurements of total residual oxidants (TRO) in seawater to those being made by researchers at other sites. The results of these investigations are that the developed procedure for preparing the sample for analysis by polarographic technique did not cause the large errors reported by others.

Since chlorine is an oxidant and is known to react with organics, we hypothesized that its addition to seawater could cause changes in the copper-organic complexes we observed in our metal studies. It could reduce the total amount of organics available for complexation with copper or release some of the bound copper.

It was necessary to determine whether the chlorination of seawater would convert complexed copper forms to the toxic or bioavailable ionic form. The electrochemical form of copper was measured with ASV present before

and after chlorination. Seawater samples were spiked with ionic copper, then aged for specific periods of time to allow copper to complex with dissolved organic matter. After aging, the seawater samples contained 5-35 ppb of complexed copper. With the addition of 1.5 ppm chlorine, 1-4 ppb of complexed copper was converted to ionic copper, and the complexation capacity of the water was reduced. In another experiment, however, seawater was spiked with both chlorine and copper, then aged a day. After aging, the copper speciation was similar to unchlorinated seawater, indicating that the chlorination had only a temporary effect on copper speciation. These experiments suggest that chlorination of seawater will increase the amount of bioavailable copper and decrease the complexation capacity of seawater for at least a brief period of time. This fact needs to be integrated in field sampling programs and interpretation of field data relating effluent quantities of metals with bioaccumulation by local species. Further testing is being conducted to define the extent of change caused by different levels and durations of chlorination in seawater.

Ozone Studies

Potentially, ozone could be used in place of, or in combination with, chlorine for treatment of municipal and industrial waste water and to prevent biofouling of power plant cooling systems. To identify the differences of chemical reactions resulting from ozonization of seawater, a series of tests were run, using sodium chloride solutions and natural seawater. The compounds analyzed were: bromine, chlorine, bromate and ozone. All but ozone were identified in chlorinated seawater. The results indicate that ozonization of seawater has chemical results similar to chlorination. Bromide was oxidized to bromine and, eventually, bromate. Ozone also slowly oxidized chloride ions to chlorine, although this reaction is much slower than the bromide to bromine reactions.

When power plant cooling seawater is chlorinated to prevent biofouling, usually enough chlorine is added to produce approximately 1-2 ppm of TRO. Most of this TRO is bromine (assuming low ammonia levels). If ozone was used to produce a TRO level of 1-2 ppm, the major oxidant species would also be bromine. Therefore, the environmental consequences of using ozone in place of chlorine to treat seawater should be similar.

Sediment Studies

Coastal marine sediment is known to be an important reservoir of contaminants such as metals, halogenated organics, and hydrocarbons. Thus, as part of the investigations of the cycling and bioavailability of energy effluent materials, we have conducted field and laboratory studies of the biogeochemical processes occurring in sediment and suspended particulates.

We have emphasized studies on the biogeochemistry of copper, an ecologically important metal that has potential for release, directly or indirectly, by operation of energy technologies. In a laboratory study, ionic copper was added to a sediment/water microcosm previously characterized for physical and chemical variability. Copper was rapidly removed from the water column to the sediment layer. The initial major site of copper sorption was the sediment organic fraction. However, after 4 weeks, a fraction of the copper in the organic material and all of the copper initially sorbed to sediment Fe and Mn oxides was mobilized. Although these studies are ongoing, their preliminary implication indicates that organic material and the oxides of Mn and Fe exert a major control over trace metal partitioning.

Under ambient field conditions in Sequim Bay, sediment organic material and Mn and Fe oxides also appear to be the primary factors controlling metal cycling. In relatively organic-rich sediments, equal amounts of Cu were found to be associated with sediment organic and reductant soluble (Mn-Fe oxides) phases. In sediments containing less organic matter, Cu associated with Mn and Fe oxides predominates. We are preparing a manuscript describing the sediments of Sequim Bay and the relationship of trace metal distribution to sediment parameters.

Organic molecules have a direct effect on metals cycling by providing reactive functional groups that readily coordinate with metals to form stable linkages. They also indirectly affect metal biogeochemistry through changes in pH and Eh caused by microbial metabolic processes. For example, sorption or desorption on hydrous oxides of Mn and Fe occur in response to pH-Eh variations. The chemical and microbiological research are continuing to determine the potential for release of metals from sediments. Separation and analyses of mobile organic compounds produced by sediment microbial processes that influence the bioavailability of metals are a significant part of our study. These studies will be meshed with other investigations by our staff to estimate the accumulative and toxicological effects of organometallic complexes.

The transport of trace metals by suspended particulates was investigated using Sequim Bay as a model tidal system. We found that

the concentration of particulate copper in the surface waters increased significantly during periods of phytoplankton growth. While a normal surface-to-bottom gradient of particulate metals was present, higher surface particulate Cu levels were not due to resuspension of bottom materials since particulate Ti exhibited no seasonal changes. Surface particulate Cu also had areal variations with higher levels occurring at the lower end of the bay. This increase could reflect an accumulation of phytoplankton in that region or the mobilization of sediment-bound Cu in response to reduced oxygen levels. The analysis of additional samples are being completed preparatory to issuing a report describing our observations. Further studies are ongoing to identify the processes controlling the seasonal and areal changes.

Transport of trace metals through the entrance channel of Sequim Bay appears to be related to resuspension of bottom particulates by tidal currents. This observation, based on the comparative distribution of particulate metals, was also noted in surface waters at Admiralty Inlet, which connects Puget Sound to the Strait of Juan de Fuca. Transport of suspended matter through the channel at Sequim Bay in the summer appears to be greater on the flood tide, causing a possible net accumulation of sediment trace metals in the bay. However, limited observations indicate that winter storms may resuspend much of this material for transport on ebb tides.

In addition studies of the transport, cycling and bioavailability of copper were conducted on a series of cruises to Jervis Inlet, B.C., in cooperation with Dr. A. G. Lewis, Institute of Oceanography, University of British Columbia. Preliminary analysis shows that the quantity of suspended matter and concentration of particulate Ti in Jervis Inlet waters is about an order of magnitude less than in Sequim Bay. However, the amount of trace metals in the particulates is such that the concentration of total particulate Cu and Zn is about equal for both waters. The concentration of Mn in Jervis Inlet particulates is about two orders of magnitude greater than in Sequim Bay material, and the increased quantities of particulate trace metals may be due to sorption on Mn oxides.

Model Studies

During FY-77 development began on a hydrodynamic computer model which uses field data to iteratively improve on successive computer model estimates. The purpose of the model is to provide a mass conservative flow field for use on sediment transport, pollutant transport and biotransport models. These transport models require mass conserving flow fields in order to accurately simulate the advection, suspension and deposition of sediments, biota and chemicals in the waterbody. Since advective transport is the dominant mode of transport (as opposed to turbulent mixing) the accuracy of the estimated flow field is paramount to the accuracy of the predicted transport.

Normally, tidal hydrodynamic models solve the full two-dimensional equations of motion to obtain flow field estimates. Obtaining such estimates can often require considerable time, effort, expense and expertise. A rather novel technique has been developed for bypassing some of these problems, provided limited field data are available. By using the stream function equation (with vorticity) one can obtain mass conservative flow fields, provided the vorticity can be estimated. The vorticity is estimated by obtaining the difference between local model computed velocities and field data. The vorticity estimate is now entered into the stream function equation and solved to obtain new velocities. By performing this sequence iteratively, the flow field will be forced closer and closer to the field data and still will be mass conservative. The development of the model is nearly complete and testing will begin early in FY-78.

• Marine Chemistry of Energy-Generated Pollutants

Principal Investigators: E. A. Crecelius, D. E. Robertson, K. H. Abel, D. A. Cochran and W. C. Weimer

This program is designed to increase our understanding of the biogeochemical and physical processes that control the fate of energy-generated pollutants that enter the marine environment. The increased energy needs of our country and increased utilization of the coastlines for siting energy generating facilities and related industries has resulted in the introduction of energy-related pollutants to the oceans from two main sources: 1) the emission of large quantities of material to the atmosphere and subsequent deposition in the oceans, and 2) direct discharges to the oceans from coastal effluents. This program is combined from Chemistry of Ocean Solutions and Geochemical Ocean Sections Study (GEOSECS).

The information generated by this program is vital to the U.S. DOE's interests in understanding: 1) the natural origins, distributions and concentrations in baseline data of trace metals and other contaminants in the oceans; 2) the input rates and mixing rates of pollutants introduced to the oceans; 3) the behavior and fate of the anthropogenic pollutants entering the oceans from the atmosphere and the continents; and 4) an assessment of the potential environmental impact of energy-generated pollutants on the marine environment. Specific tasks that were accomplished during the last year include determining the deposition rate of elements on the Washington Coast, shipboard analysis of mercury in seawater by a new, more sensitive technique, analysis of trace elements in coastal and oceanic waters, and the evaluation of the BLWS for radionuclide sampling.

Atmospheric Deposition of ^{7}Be and Other Elements on the Washington Coast

Atmospheric fallout of anthropogenic dust is a major source of trace metals to coastal marine waters. During the last 2 yr, the concentrations of ^{7}Be and 15 other elements have been measured in air filters and total deposition samples collected at Quilayute on the Washington Coast. These data were used to calculate three parameters: 1) the deposition velocity (Vd), 2) the total elemental deposition/area/yr, and 3) the ratio of element/^{7}Be in air to element/^{7}Be in the deposition collector. With these data, the atmospheric input of elements to coastal waters can be estimated and the atmospheric input to the open ocean can be predicted.

The Vd for elements at Quilayute are shown in Table 7.1. These numbers were calculated by dividing the rate of element fallout (g $m^{-2}yr^{-1}$) by the concentration of element in air (g m^{-3}). The ^{7}Be Vd measured at Quilayute (0.96 cm sec^{-1}) is very similar to that estimated by Young and Siker (1977) off the Washington Coast (1-1.2 cm sec^{-1}). This close agreement between the ^{7}Be Vd strengthens our assumption that Quilayute is a representative site for coastal air chemistry studies. The

TABLE 7.1. Average Annual Deposition Velocity (Vd) for Airborne Elements at Quilayute, Washington, and Ratio of Element/^{7}Be in Air Filters and Deposition Samples

Element	Vd, cm sec^{-1}	Air Element/^{7}Be Deposition/^{7}Be
Cl	6.4	0.11
K	2.6	0.17
Ca	2.6	0.15
Br	2.3	0.30
Fe	0.37	1.30
Mn	0.59	0.60
Ti	0.67	0.50
Cr	0.60	0.50
Pb	0.38	1.50
Se	0.24	1.80
As	0.70	0.30
V	0.24	1.40
Cu	3.00	0.08
Ni	4.00	0.12
Zn	1.80	0.24
^{7}Be	0.96	1.00

Vd for other elements (Table 7.1) can be divided into two groups, those that have Vd of >2 and those <0.7. The high Vd group is believed to be associated with sea-salt aerosols that are typically several μm in diameter. The low Vd group are believed to be of continental origin and in the <1μ size range. The Vd measured at Quilayute are similar to those measured by Cambray in 1975 at six coastal sites on the North Sea. This gives us confidence that our Vd can be applied to all rural coastal and oceanic areas in midnorthern latitudes.

The high Vd for Cu, Ni and Zn have not been explained. These metals appear to be associated with a marine source. A similar effect was reported by Cambray in 1975 for the North Sea.

The ratios of element/^{7}Be in air to element/^{7}Be in total deposition were calculated to determine if ^{7}Be is a good predictor of element deposition in the marine environment. The data in Table 7.1 show that ^{7}Be Vd is a good predictor of elements associated with continental origins (Fe, Mn, Ti, Cr, Pb, Se, As and V), but is a poor predictor of elements associated with marine-derived aerosols (Cl, Br, Ca, K, Cu, Ni and possibly Zn). The conclusion is that ^{7}Be is associated with aerosol of <1μ size and, therefore, has a Vd similar to other elements associated with small aerosol. The elements associated with marine-generated aerosol (sea-salt) has Vd 2-6 times greater than ^{7}Be and, therefore, ^{7}Be deposition rate will greatly underestimate their deposition rates.

The ^{7}Be Vd data for the world oceans and marine air chemistry data will be used to estimate atmospheric input to the ocean of pollutants associated with small aerosols. We plan to compare these atmospheric input rates with other major inputs such as rivers.

Observations of Ultra-low Mercury Concentrations in Oregon-Washington Continental Shelf Waters

Mercury concentrations in ocean water collected off the Oregon-Washington continental shelf have been found to be approximately ten times lower than the best previous measurements. Utilizing a newly developed procedure, which is 50 times more sensitive than previous techniques, measurements of the mercury concentrations in seawater were made on board the R/V Cayuse immediately after the samples were collected. This procedure has greatly reduced the contamination of seawater with mercury during sample storage. Surface seawater was collected in a carefully precleaned plastic pail thrown from the bow of the ship while slowly cruising forward. Deep samples were collected in teflon-coated Niskin Go-Flo® bottles. Immediately after sampling, 800 mℓ of the seawater was transferred to carefully precleaned 1-ℓ Pyrex reagent bottles and acidified with 2.5 mℓ of low-mercury nitric acid and heated in a hot water bath to 60°C. Several minutes after sampling, the mercury was reduced to elemental mercury vapor by adding 20 mℓ of 10% $SnCl_2$ solution and the seawater was purged with nitrogen gas for 12 min at a flow rate of 1 ℓ/min. The purged elemental mercury vapor was collected on a tube of gold-coated glass beads. The gold bead tube was then heated in an induction coil to quickly drive the mercury, in a stream of nitrogen gas, through an LDC® mercury analyzer for detection and quantification. Standard curves were prepared by spiking low-mercury seawater with known quantities of standard mercury solutions and running the spiked samples through the above procedure. The detection sensitivity of this procedure is about 0.1 ng of mercury. Seventeen stations were occupied in Oregon-Washington coastal waters. Samples of Newport Bay and Puget Sound seawater and seawater collected off the Oregon-Washington continental shelf typically contained 0.4 to 1.0 ng mercury/ℓ of unfiltered seawater. These

values are about ten times lower than the best previous measurements which estimated that mercury was present in these waters at a concentration of several ng/ℓ. The ultra-low levels of mercury observed in seawater (0.5 ng/ℓ) are of great significance. They indicate that mercury must have a very short residence time in the oceans. They also mean that phytoplankton, which contains tenths of a ppm of mercury, must concentrate the mercury from seawater by about one millionfold. This efficient bioaccumulation, together with the ultra-low levels of mercury present in the oceans, emphasizes the vulnerability of the marine biosphere to man-made contamination of the oceans by this highly toxic heavy metal.

Trace Element Distributions at Pacific GEOSECS Stations

Instrumental neutron activation analysis of Pacific Ocean GEOSECS seawater samples are establishing the geographical and vertical distributions of Zn, Co, Sb, U, Cs and Rb. In general the concentrations and distributions of these elements are very similar to those observed in the Atlantic Ocean. Extremely low cobalt concentrations averaging around 0.01 µg/ℓ and ranging from 0.006-0.096 µg/ℓ were observed. Considerable variability in the depth distribution of cobalt was observed. Zinc concentrations ranged from 0.56-11.3 µg/ℓ, and averaged about 1.8 µg/ℓ. Zinc concentration maxima existed at discrete depths at each station, but did not systematically correlate with the cobalt maxima. The Sb, U, Cs and Rb distributions were very homogeneous, and averaged about 0.2, 3.5, 0.30 and 120 µg/ℓ, respectively. Preconcentration procedures for measuring arsenic and silver in these samples are presently being conducted.

We have redirected our trace element program towards the continental shelf regions of the Pacific Coast of the U.S. Seawater samples from two cruises in this area are being readied for trace element analysis. We have commenced the analysis of vanadium, an element of considerable interest in the marine environment because of its potential for contamination during oil spills. Vanadium is present in crude oils in relatively high concentrations. The vanadium concentration in Pacific coastal waters off Washington and Oregon is about 1.8 µg/ℓ, and little variability in its distribution in these waters was observed.

Determination of Soluble Aluminum in Seawater

Soluble aluminum levels in seawater are generally thought to be on the order of 1 µg/ℓ; however, few measurements have been made in the past, partially due to limitations in sensitivity and contamination. It is important to assess the levels and variations in soluble aluminum, since one expects particulate aluminum and, thus, possibly soluble aluminum, to vary greatly in coastal environments due to natural processes. It is also possible that anthropogenic activities influence these levels over small areas presently and may be of greater influence in the future.

Toward this end, we have developed a sensitive and quantitative procedure for the determination of soluble aluminum. We utilized an iron hydroxide coprecipitator ion reported previously for determination of arsenic, vanadium, and selenium. Evaluation was conducted using ^{26}Al radiochemical tracer and previously filtered and acidified seawater.

Iron carrier (5 mg) and phenol red indicator were added to six 100 mℓ aliquots of seawater. Sodium hydroxide (1N) was then added dropwise during stirring until the indicator endpoint was reached. The precipitate formed was allowed to age approximately 15 min. It was then centrifuged, the supernatant discarded, and the precipitate then washed with 0.5 M ammonium acetate. The samples were then centrifuged, the supernatant discarded and the final precipitate dissolved in acid and brought to 70 mℓ. The samples and appropriate standards were then γ counted for 1000 min using NaI(Tl) multidimensional gamma ray spectrometers to determine efficiency of recovery during the precipitation procedure. Recovery for the six samples was 99.7 ± 2.0%, indicating the procedure was quantitative and exhibited excellent precision.

Efficiency Evaluation of the Battelle Large Volume Water Sampler

The Battelle Large Volume Water Sampler (BLVWS) has been used for a number of years for concentrating radionuclides from very large volumes of fresh and ocean water (e.g., up to 4000 ℓ). Previous calibrations of this sampler have been conducted during field experiments and in scaled-down laboratory tests. However, no efficiency tests have ever been performed in which large volumes of seawater have been equilibrated with radioactive tracers and then processed through the

BLVWS. This study was conducted to evaluate the adsorption efficiencies for the radionuclides on activated aluminum oxide from large volumes of seawater.

Six 200-ℓ samples of seawater were pumped from Sequim Bay into plastic-lined polyethylene drums and spiked with μCi amounts of the following radionuclides: ^{51}Cr, ^{54}Mn, ^{59}Fe, ^{60}Co, ^{65}Zn, ^{95}Zr-Nb, ^{106}Ru, ^{110m}Ag and ^{144}Ce. The tracers were allowed to equilibrate in the seawater for 2 days and the seawater samples were then pumped through six BLVWS loaded with a series of eight fiberglass filters (30 cm dia) and five beds of 0.7 cm thick by 30 cm dia activated aluminum oxide. The 200 gal spiked seawater samples were diluted with 800 ℓ of raw Sequim Bay seawater before entering the BLVWS by simultaneously pumping from a large reservoir. The combined seawater stream was pumped through the BLVWS at a flow rate of 28 ℓ/min and a total volume of 1000 ℓ of seawater was processed through each sampler. After each sampling, the BLVWS was taken apart and the filters combined for analysis. The five aluminum oxide beds were separately packaged. The filters, oxide beds and plastic drum liners were packaged in standard counting geometries and counted on a large Ge(Li) gamma-ray spectrometer.

During the course of this experiment the amount of tracers lost by adsorption onto the plastic drum liners ranged from 0.05% for ^{124}Sb to 16% for ^{59}Fe. The amounts of particulate species and the percent of the total soluble radioactivity removed on each aluminum oxide bed are shown in Table 7.2.

TABLE 7.2. Efficiency Evaluation of the Battelle Large Volume Water Sampler

	Average % Particulate	Average % of Total Soluble Activity Removed on Each Al_2O_3 Bed				
		1st	2nd	3rd	4th	5th
^{51}Cr	66.2 ± 7.4	55.0 ± 9	14.5 ± 2.1	7.1 ± 1.2	4.3 ± 0.8	2.9 ± 0.4
^{144}Ce	58.0 ± 5.9	96.0 ± 12	11.5 ± 3.6	4.2 ± 1.2	2.5 ± 0.6	2.0 ± 0.5
^{95}Zr	7.4 ± 1.2	73.0 ± 5	16.8 ± 2.9	3.8 ± 1.5	1.1 ± 0.4	0.45 ± 0.13
^{106}Ru	32.7 ± 4.1	49.0 ± 6	18.6 ± 3.2	7.4 ± 1.7	3.8 ± 1.2	2.5 ± 0.5
^{65}Zn	11.1 ± 1.2	85.0 ± 5	8.3 ± 2.2	0.83 ± 0.43	0.30 ± 0.05	0.18 ± 0.03
^{59}Fe	75.9 ± 7.7	59.0 ± 19	23.1 ± 8.3	15.7 ± 4.2	11.6 ± 2.9	8.0 ± 2.3
^{60}Co	5.8 ± 2.3	19.0 ± 3	13.9 ± 2.2	10.0 ± 1.1	7.8 ± 1.0	6.1 ± 0.7
^{54}Mn	20.3 ± 4.3	5.0 ± 1.2	3.5 ± 0.8	2.7 ± 0.5	2.4 ± 0.5	2.0 ± 0.5
^{110m}Ag	26.9 ± 7.7	2.7 ± 1.5	2.3 ± 1.4	1.9 ± 0.9	2.1 ± 1.4	1.9 ± 1.2
^{124}Sb	<1	2.1 ± 0.71	2.4 ± 0.8	2.7 ± 1.0	2.9 ± 1.0	3.2 ± 1.2

Particulate forms (greater than 0.5μ) ranged from <1% for ^{124}Sb to 75.9% for ^{59}Fe. The first Al_2O_3 bed was very efficient in removing ^{144}Ce, ^{65}Zn, ^{95}Zr, ^{59}Fe, ^{51}Cr and ^{106}Ru. The ^{60}Co, ^{54}Mn, ^{110m}Ag and ^{124}Sb retention efficiencies decreased in that order. The first Al_2O_3 bed appeared to be slightly more efficient than succeeding beds, indicating the presence of possibly microparticulates or colloids which might be trapped on the first bed. The retention efficiencies of the second to fifth beds decreased linearly. For the most part the results of this tracer experiment were in good agreement with previous scaled-down efficiency tests.

^{55}Fe and Stable Iron-Comparative Biogeochemical Behavior

Studies of atmospheric aerosols have demonstrated that much of the ^{55}Fe associated with the aerosol input to the oceans is present as either an amorphous or hydrous iron oxide or as very small particulate species attached to the surfaces of the large aerosol particles. By comparison, nearly all of the stable iron is bound in the mineral phase of aerosol particles. This difference in the chemical and physical forms of the radioactive and stable iron isotopes results in the ^{55}Fe being more biologically available than is the stable iron. This difference in availability is responsible for the transfer of a much higher specific activity ^{55}Fe to certain ocean organisms and man than that present in aerosol particles or in seawater. This differential biological uptake of the radioactive iron and its stable element counterpart indicates the problem of always relying on the stable elements in the marine environment to effectively dilute radioelements or other stable elements of anthropogenic sources. The effectiveness of dilution by natural sources depends on the chemical and physical forms of the materials in both the source terms and the receiving environments. The large difference in specific activities of ^{55}Fe in aerosols and seawater relative to ocean organisms reflects the independent behavior of ^{55}Fe and stable iron. Full details of this work are being submitted for publication.

- **In Situ Pollutant Measurements**

Principal Investigator: N. A. Wogman

Other Investigators: K. K. Nielson and H. G. Rieck

This program provides feasibility evaluation, development, and application of instrumental technology for the in situ analysis of the wide spectrum of inorganic, organic, and radionuclide species in ocean and freshwater sediments.

In Situ X-ray Fluorescence System Development

The in situ x-ray fluorescence analyzer developed by this laboratory has successfully measured elemental concentrations in sediment surfaces to water depths of 100 m. With modifications now being incorporated, the equipment will also give accurate measurements down to 6 cm deep in the onsite sediment and allow system use in water depths of 300 m.

Underwater measurements in Puget Sound, Washington were made with in situ x-ray fluorescence equipment mounted in a framework which allowed the probe to just contact the sediment surface. The resulting measurements provided elemental concentrations for the top few millimeters of the sediment column. Although this provided information on current elemental concentrations, greater use could be derived if it were possible to measure elemental concentrations in successive layers of the sediment column. The existing in situ x-ray fluorescence analyzer is currently being modified to provide a system with this capability. Excavating hardware being added can remove a few millimeters of sediment. Following sediment removal the analyzer sensor is lowered to the new surface and the measurement made. This process is repeated successively to a sediment depth of about 6 cm. Excavation of sediment from under the sensor is accomplished by a series of blades attached between roller chains located on each side of the existing x-ray fluorescence analyzer as shown in Figure 7.4.

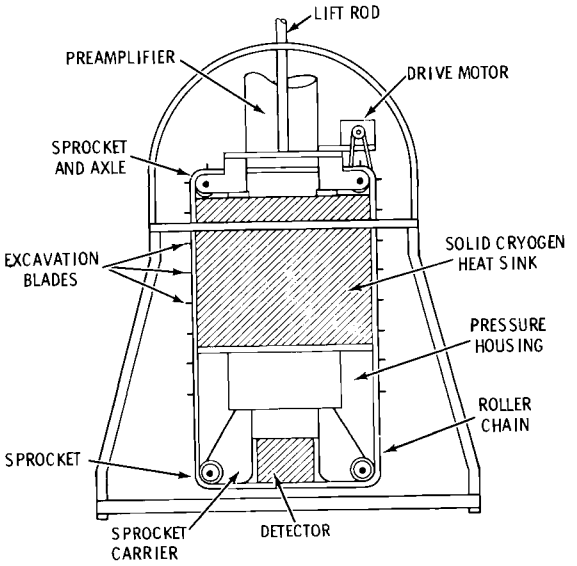


FIGURE 7.4. In Situ X-Ray Fluorescence Probe Modified to Allow Sediment Analysis with Depth.

The pressure housing for the x-ray fluorescence sensor has also been reduced to 7 cm diameter to minimize the excavation area required. The present pressure housing is equipped with a 0.05 mm thick 5-cm diameter Be window allowing operation of the system to water depths of 100 m. By changing to a 0.5 mm thick Ti window, sediments can be analyzed in water depths of 300 m. In the latter

case, ^{241}Am is used to excite the sediment elements instead of ^{109}Cd. The ^{241}Am has a higher energy exciting proton which penetrates the window more easily. The Ti window does decrease the transmission of low energy x-rays (<15 keV), thus, the sensitivity for Fe, Ba, and the intermediate Z elements are reduced by factors of from 3 to 5.

In a typical operation, the modified in situ x-ray fluorescence system will be lowered from a surface vessel or submarine to the sediment region. Following measurement of surface pollutants, power is applied to the excavation assemblies. Chain operation is monitored topside by detection of blade material as it passes under the detector. Shutdown of power releases a brake which allows the x-ray fluorescence system to lower to the new surface and sediment analysis to begin. Use of the modified in situ x-ray fluorescence analyzer is providing knowledge not only of pollutants on the sediment surface but also the changing concentrations of pollutants as a function of sediment depth.

Data Reduction Methods for In Situ X-ray Fluorescence Seabed Analyzer

Continuing method development on data reduction programs allows quantitative analysis of in situ x-ray fluorescence data from the sea bed. Analyses are complicated by the extreme variations in sample matrix (0-90% suspended sediment in water), causing large potential errors from self-absorption, particle size effects, enhancement effects, and the variable mass and volume of sample being "viewed" by the analyzer. A computer code has been developed to relate the backscattered exciting radiation to the mass and average atomic number Z of the material being viewed by the analyzer.

The newly developed computer program estimates appropriate self-absorption, particle size, and enhancement corrections. The measurement of sample mass and average Z is accomplished by separate integration of the coherent and incoherent backscatter peaks and solution of simultaneous equations relating the backscatter intensities to masses of two representative like elements. These representative elements are chosen by the incoherent-to-coherent intensity ratio. Since elements Z greater than 25 are directly measured by fluorescence x-ray peaks, like elements are chosen to represent the Z less than 25 fraction of the sample which comprises most of its mass. Since all mass estimates and corrections are computed from fundamental parameters and an initial spectrometer calibration, the analyses are independent of sample matrix. The light variations in going from fresh water to salt water, or from packed sediments to colloidally-suspended organic matter, are thus accounted for. Preliminary tests with this method suggest a strong correlation ($R^2 = 0.97$) between the computed sample mass and its water content (as determined from grab samples). If further work confirms the relationship, the known water content will permit estimation of the results as part-per-million in sediment, part-per-million in water, or part-per-million in combined suspension.

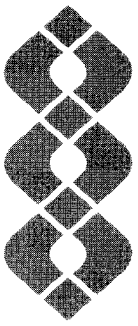
X-ray Fluorescence Capabilities for Uranium Ore Analysis

A comparison of various instrumental methods has been completed for the determination of uranium in a variety of ores. Included in this comparison were energy dispersive x-ray fluorescence (XRF) methods, wavelength dispersive x-ray fluorescence methods, and several commonly used nonfluorescence photon analysis methods. The XRF methods were examined for both thin and thick samples. Uranium sensitivities and detection limits are compared in Table 7.3. Nearly all of the XRF methods listed provide suitable sensitivity and detection limits for rapid assay of uranium in commercial-grade ores (greater than 0.01% uranium).

Part-per-million uranium detection limits can be routinely achieved using an isotope, secondary source, or direct tube excitation with Si(Li) detectors, or direct tube excitation with wavelength dispersive detectors. The lowest detection limits are obtained with L x-rays for energy dispersive XRF and with either L or M x-rays for wavelength dispersive XRF. Even when uranium standards are not readily available, rapid quantitative analyses may be completed using mathematical methods to compute corrections for an unknown matrix. The XRF capabilities for uranium ore analysis exceed those of direct gamma ray photon spectrometry, or of methods using ^{252}Cf neutron-induced fission activation of the uranium.

TABLE 7.3. Comparison of Bulk Sample XRF Analyses for Uranium.

Peak	Excitation	Detection	Count Rate (c/sec)	Sensitivity (C/S · ppm)	60 sec 3σ Detection Limit (ppm)	Sample
U M_α	Cr Tube, 2.5 kW	Pet		1.1	1.6	71 mg/cm²
U $M_{\alpha\beta}$	^{55}Fe, 35 mCi	Si(Li)	500	0.0093	46.0	63 mg/cm²
U $M_{\alpha,\beta}$	Ti Secondary (1.5 kW W-Tube)	Si(Li)	1000	0.15	20.0	63 mg/cm²
U L_α	Mo Tube, 2.5 kW	LiF (220)		~4.4	2.3	510 mg/cm²
↓	Rh Tube, 12 W	Si(Li)	14,000	0.19	32.0	63 mg/cm²
↓	Rh Tube, 35 W (Pulsed)	↓	20,000	0.38	25.0	63 mg/cm²
↓	Ag Secondary (2.2 kW W-Tube)	↓	2500	0.53	3.6	63 mg/cm²
↓	Mo Secondary (2.2 kW, W-Tube)	↓	2400	1.14	2.8	63 mg/cm²
↓	^{109}Cd, 57 mCi	↓	1400	0.50	3.9	63 mg/cm²
↓	^{109}Cd, 50 mCi	↓	3000	0.56	20.0	In-Situ, Under Water
↓	^{241}Am, 100 mCi	↓	340	0.0057	61.0	63 mg/cm²
↓	^{109}Cd, 3mCi	NaI(Tl), Zr + Y Matched Filters		0.075	~300.0	Field or In-Situ
↓	^{109}Cd, 3mCi	Proportional Counter		0.0033	~400.0	Field or In-Situ
U K_α	^{57}Co 0.5 mCi	Intrinsic Ge	170	0.00071	670.0	1.5 g/cm²



8.0 Hanford Project Support

HANFORD PROJECT SUPPORT

- **Rockwell Support Studies**
- **Decommissioning and Decontamination Studies**
- **Service Assessment Studies**

This section reports on ecological work conducted in support of a variety of industry-related operations on the Hanford Site. Past work has been directed toward establishing an ecological data base characterizing the environment. This provided documentation concerning present environmental quality necessary as baseline knowledge when considering possible, future energy technologies.

Much of this year's Rockwell Hanford Operations supported studies centered on preparation of a "status document" summarizing past ecological work on the 200 Area Plateau, assessing the existing data base and projecting future research needs. These studies draw heavily on ecological expertise developed during past years as a result of major program support by the Division of Biomedical and Environmental Research (DBER). In particular, the development of unique ecosystems measurement approaches, as used at the Rockwell sites, and the development of the literature files on radioecology were contributed from DBER's ROWMA and Terrestrial Ecology programs (see, respectively, Sections 2 and 1).

A new effort, this year, involved studies supported under the Decommissioning and Decontamination Program. Environmental studies were designed to ascertain ecological hazards of 300 Area burial grounds in terms of transport of radioactive wastes away from managed areas and risk to biota.

- Intercontractor Supported Studies

o Rockwell Support Studies

Principal Investigator: L. E. Rogers

Other Investigators: D. A. Cataldo, K. A. Gano, R. G. Schreckhise, R. E. Fitzner, D. Paine, R. E. Wildung J. F. Cline, D. W. Uresk and R. H. Sauer

Technical Assistance: M. A. Combs, L. F. Nelson, C. A. Lee, M. J. Harris, V. D. Charles and H. A. Sweany

The Rockwell Hanford Operations (RHO) supported ecological studies are designed to clarify ecosystem structure and functioning as pertaining to the management of radioactive waste control areas. To date, emphasis has been placed on characterizing the abiotic and biotic components of these areas, resulting in publication of over 20 PNL documents and several scientific articles pertinent to the 200 Area Waste Management Program. Future research emphasis will include energy and mineral cycling mechanisms, radionuclide transport by biota and modeling of ecosystem functioning, thereby integrating past characterization and future ecological transport studies for application to waste management operations.

Status Document

Much of this past year's research sponsored by Rockwell has centered on preparation of a document summarizing past ecological research on the 200 Area Plateau waste management facilities (PNL-2253, Rogers and Rickard, 1977). This review permitted an assessment of the present data base and projection of future research needs. Past and projected research needs were identified in terms of six research categories: 1) characterization, 2) biotic production, 3) transfer processes, 4) radionuclide transport, 5) environmental assessment, and 6) management modeling.

Future research tasks scheduled for particular categories were presented in a Five Year Milestone Chart with Task Interrelationships illustrated in a flow diagram. This documents the need to accomplish a variety of seemingly unrelated research tasks in a timely manner. Studies concerning the impact of radiation dose on deer mouse populations or the diets of grasshoppers may seem esoteric when considered independently, but are essential when viewed as a part of a complete program.

Ground-Dwelling Beetles

Darkling beetles are an important component of most semi-arid ecosystems and are common inhabitants of waste burial sites at Hanford. The food habits of darkling beetles were determined as part of an overall program for identifying major transport pathways within the confines of the Hanford Site.

The relative frequency of food item consumption is shown in Table 8.1. There were 52 different food items recorded: 8 grass species, 33 forb species, 6 shrub species and 5 miscellaneous food types. Considering all darkling beetles together, the most frequently ingested food item was western tansymustard (Descurainia pinnata). This small plant, with its yellow flowers, is ubiquitous

TABLE 8.1. Plant Taxa and Frequency of Ingestion by Darkling Beetles on the Hanford Site[a]

Taxon	Consumption Frequency, %
GRASSES	
Agropyron spicatum	<1
Bromus tectorum	9
Festuca octoflora	<1
Poa cusickii	<1
Poa sandbergii	1
Stipa comata	<1
Stipa thurberiana	1
Unknown grass	1
FORBS	
Ambrosia acanthicarpa	<1
Amsinckia spp.	1
Antennaria dimorpha	<1
Astragalus spp.	<1
Balsamorhiza careyana	<1
Calochortus macrocarpus	<1
Castilleja sp.	<1
Chenopodium leptophyllum	3
Chenopodium sp.	<1
Crepis atrabarba	1
Cryptantha spp.	1
Cymopterus terebinthinus	<1
Descurainia pinnata	15
Draba verna	<1
Erigeron filifolius	<1
Erysimum asperum	1
Helianthus sp.	1
Lomatium macrocarpum	<1
Lupinus spp.	<1
Microsteris gracilis	<1
Phacelia linearis	<1
Plantago patagonica	<1
Polemonium micranthum	<1
Psoralea lanceolata	<1
Salsola kali	2
Sisymbrium altissimum	5
Sphaeralcea munroana	3
Taraxacum officinale	<1
Townsendia florifer	<1
Tragopogon dubius	<1
Compositae	<1
Liliaceae	<1
Unknown forbs	2
SHRUBS	
Artemisia tridentata	13
Atriplex spinosa	1
Chrysothamnus nauseosus	2
Chrysothamnus viscidiflorus	<1
Eurotia lanata	8
Sarcobatus vermiculatus	4
MISCELLANEOUS	
Arthropod parts	5
Cryptogams	13
Hair	<1
Seeds	1
Pollen	2

[a]N = 959 slides; 19,180 observed fields.

on the Hanford Site but more common at lower elevations. It occurred in the darkling beetle diet with a frequency of 15%. Other common food plants included big sagebrush (13%), mosses and lichens (13%), cheatgrass (9%), winterfat (8%), Jim Hill mustard (Sisymbrium altissimum) 5%, and arthropod parts (5%). The remaining 45 food plants were consumed with a frequency less than 5%.

The list of food plants utilized indicates that darkling beetles select a wide range of plant species. But this list represents the food source of an entire beetle family. Individual species may be more specific feeders.

Mule Deer Diets

Dietary preferences of mule deer (Odocoileus hemionus) were determined. Fecal pellets were collected from three diverse areas and analyzed by microscopic technique to determine their plant composition. Three study areas were characterized for plant frequency of occurrence and canopy cover. Two sites were adjacent to man-made ponds with differing amounts of riparian vegetation, while the third site was in a sagebrush/cheatgrass-sandberg bluegrass community. At all sites cheatgrass was the most abundant understory species. Sagebrush was the major overstory species at B Pond and at the sagebrush/cheatgrass-sandberg bluegrass site. However, at Gable Pond hopsage provided most of the overstory canopy cover. Total canopy cover was 42% at B Pond, while Gable Pond and the sagebrush/cheatgrass-sandberg bluegrass site provided 54% and 59% canopy cover, respectively. The important plant species in the deer fecal pellets varied among the three locations. At B Pond 24 species occurred in the diet with the most important being Russian thistle which provided 64% of the diet. Thirty-six species occurred in pellets collected from the Gable Pond site with the most abundant being goldenrod, which constituted 32% of the total diet. Pellets collected from the sagebrush/cheatgrass-sandberg bluegrass site had 13 plant species with the major species being bitterbrush. Bitterbrush made up 69% of the plants found in the pellets. The similarity in diets among sites was low, ranging from 10% to 16%. Preference indices among the sites were not similar (6-18%) indicating that deer are selecting food plants that are site specific.

Coot Studies

The American coot (Fulica americana) is a common inhabitant of all ponds located on the Hanford Site. The biology and radioecology of the coot is presently under investigation since they represent a "worst-case" example for the potential transport of waste materials away from managed areas. This report concerns the biology of the American coot in southcentral Washington. A later report will summarize results of the radioecological studies. Five ponds of similar vegetational succession and water depth were selected for the investigation. These were Gable Mountain

Pond, U Pond and B Pond on the U.S. DOE Hanford Reservation, and Royal Slough and Morgan Lake within the Columbia National Wildlife Refuge near Othello, Washington. Gable Mountain, U and B Ponds differed from the other two study sites in that they received small amounts of man-made radionuclides introduced along with coolant water from production chemical facilities.

The investigation compared several facets of coot nesting biology on the five ponds but one of the major points of the study dealt with food habits. Table 8.2 provides a listing of the food eaten by these birds on the study areas in southeastern Washington. These data were similar to other published reports and reveal that the coot is strongly vegetarian with similar food habits for study areas located both on and remote to the Hanford Site (Table 8.2).

Soil Affixants

Shallow burial has been a technique long employed for low-level radioactive dry waste disposal at Hanford. This technique involves digging a shallow trench, placing waste material on the bottom of the trench, and backfilling with earth. Since many of the natural soil profiles at burial sites have a layer of cobbles overlain by a sandy topsoil, they are subject to wind erosion.

Nevertheless, buried wastes have remained successfully removed from biological accessibility in almost all instances. However, there have been a few instances where deep-rooted plants or burrowing animals have brought the buried radioactive materials to the surface where they can enter ecological food chains. These incidents have prompted usage of soil affixants for soil erosion control and as biobarriers to prevent plant roots and burrowing animals access to buried wastes.

This report describes results of a study concerning possible adverse environmental effects associated with soil affixant usage in terms of seed germination, plant water usage and yield (Table 8.3). Layers of latex emulsion and asphalt emulsion mixtures applied to soil surfaces did not reduce seed germination significantly nor was there any evidence of induced plant toxicity. Asphalt emulsion and asphalt-neoprene affixants reduced the loss of soil water, but water utilization by plants growing in latex-treated pots was nearly the same as for controls. This study suggests that these treatments can be used as soil stabilizers to control the moisture flow from the soil as required by the burial procedures and to help control wind erosion.

TABLE 8.2. Foods Eaten by the American Coot in Southeastern Washington.

Plants	Study Areas: Gable Mountain Pond % Frequency ± S.E. N = 89	U Pond % Frequency ± S.E. N = 14	B Pond % Frequency ± S.E. N = 25	Morgan Lake and Royal Slough % Frequency ± S.E. N = 15
Thallophyta				
Algae				
Filamentous algae	22.2 ± 0.02	22.4 ± 0.06	25.0 ± 0.04	7.2 ± 0.02
Chara sp. (muskgrass)	5.2 ± 0.02	—	1.8 ± 0.01	1.5 ± 0.01
Spermatophyta				
Najadaceae				
Potamogeton sp. (pondweed)	20.1 ± 0.02	23.3 ± 0.05	22.4 ± 0.03	21.1 ± 0.04
Potamogeton pectinatus (Sago pondweed)	13.2 ± 0.02	26.1 ± 0.05	14.3 ± 0.03	27.8 ± 0.18
Cyperaceae				
Scirpus sp. (Bulrush)	2.1 ± 0.01	0.1 ± 0.01	0.01 ± 0.01	0.01 ± 0.01
Lemnaceae				
Lemna sp. (duckweed)	0.3 ± 0.01	2.2 ± 0.01	—	3.01 ± 0.01
Typhaceae				
Typha latifolia (cattail)	0.5 ± 0.01	—	—	—
Ranunculaceae				
Ranunculus aquatilis (white water crowfoot)	3.1 ± 0.01	2.0 ± 0.01	—	—
Haloragidaceae				
Myriophyllum sp. (watermilfoil)	18.6 ± 0.02	7.2 ± 0.03	17.7 ± 0.03	19.8 ± 0.04
Animals				
Invertebrates	12.5 ± 0.02	7.5 ± 0.02	17.2 ± 0.03	17.5 ± 0.05
Undetermined				
Vegetation	2.4 ± 0.03	1.3 ± 0.02	1.6 ± 0.02	2.0 ± 0.02

TABLE 8.3. Effects of Soil Affixants on Plant Growth

Treatment	Seed Germination (%) 1st Crop	Seed Germination (%) 2nd Crop	Water Used (ml/da/pot) 1st Crop	Water Used (ml/da/pot) 2nd Crop	Yield (g/pot) 2nd Crop
			- - - Cheatgrass - - -		
1-1/2% Latex	46 ± 5	89 ± 5	99 ± 2	95 ± 1	0.31 ± 0.02
10% Latex	30 ± 3	96 ± 1	89 ± 4	95 ± 1	0.33 ± 0.01
20% Latex	36 ± 3	92 ± 3	98 ± 2	87 ± 3	0.36 ± 0.01
Asphalt Emulsion	49 ± 9	90 ± 3	39 ± 3	26 ± 1	0.54 ± 0.03
Asphalt-Neoprene	24 ± 6	72 ± 2	12 ± 1	11 ± 5	0.27 ± 0.02
Control	106 ± 5	90 ± 2	107 ± 1	88 ± 1	0.35 ± 0.02
			- - - Russian Thistle - - -		
1-1/2% Latex	30 ± 8	83 ± 2	91 ± 2	64 ± 2	0.65 ± 0.04
10% Latex	18 ± 7	56 ± 4	92 ± 1	52 ± 1	0.35 ± 0.04
20% Latex	19 ± 3	59 ± 9	103 ± 1	61 ± 1	0.27 ± 0.04
Asphalt Emulsion	46 ± 5.0	53 ± 9	40 ± 2	13 ± 1	0.29 ± 0.03
Asphalt-Neoprene	10 ± 3	1.2 ± 3	11 ± 0.4	12 ± 2	0.02 ± 0.02
Control	36 ± 13	86 ± 6	110 ± 1	68 ± 1	0.48 ± 0.04

$\bar{X}$ ± SE

U Pond Mammals

The effects of radiation dose to biota inhabiting waste management areas are of increasing concern as detailed by the recent Hanford Environmental Impact Statement. This study was designed to ascertain body burdens and dose for a population of small mammals inhabiting U pond environs, a low level radioactive waste pond. Four species of mice were trapped: the Great Basin pocket mouse (Perognathus parvus), deer mouse (Peromyscus maniculatus), house mouse (Mus musculus), and the Western harvest mouse (Reithrodontomys megalotis). Tissue samples were collected throughout the study for gamma scan analysis. An analysis for $^{238-240}Pu$ and ^{241}Am was also performed. The most abundant gamma emitter was ^{137}Cs with the highest levels associated with Z-19 Trench and two areas adjacent to the pond. House mice from the Z-19 Trench showed the highest levels with 1600 pCi ^{137}Cs/gm dry weight in a GI tract sample. Radiochemical analysis suggests that the potential for plutonium uptake by mice is very low. The highest value detected for either plutonium or americium was 2.03 pCi ^{239}Pu/gm dry. This sample came from house mice inhabiting the meadow transect near the pond. Results of radiochemical analyses indicate that of the four species of mice examined all but the pocket mouse show a potential for radionuclide uptake. This is attributed to food and habitat preference. Pocket mice, in general, prefer the dry desert Artemisia tridentata - Bromus tectorum habitat over the moist, riparian habitat associated with the pond and trenches.

Dosimeters were implanted beneath the neck skin of living mice. Mice were released and subsequently recaptured, dosimeters removed and read. The data show that mice adjacent to the pond received the highest doses. The highest levels were associated with house mice which had an average dose of 54.9 R/yr (Table 8.4). The distribution of radioactivity was also measured according to soil profile depth. The surface and top decimeter of soil for the meadow area showed the highest contamination levels with a mean of 75 R/yr. Neutron dose in the soil profile was also measured and was also highest near the surface averaging 74 m rem/yr.

TABLE 8.4. Mean Gamma Doses to U Pond Mice (Roentgens per year)

	Pocket Mouse R/y ± S.E.	Deer Mouse R/y ± S.E.	House Mouse R/y ± S.E.
R.S. Control[a]		0.07 ± 0.003	
C.P. Control	0.09 ± 0.005	0.07 ± 0.003	
Z-Trench	0.14 ± 0.01	0.93 ± 0.51	
U-Trench	0.35 ± 0.11		
Forest	3.99 ± 2.5	2.93 ± 0.41	13.4 ± 8.8
Meadow	42.7 ± 7.32	21.1 ± 1.9	54.9 ± 1.7

[a]Rattlesnake Springs Control

o Decommissioning and Decontamination Studies

Principal Investigators: D. W. Uresk and J. F. Cline

Other Investigators: K. A. Gano, L. E. Rogers and R. E. Fitzner

Technical Assistance: H. A. Sweeny and V. D. Charles

The decommissioning of retired Hanford facilities requires careful consideration of environmentally-related factors. Applicable ecology programs have been designed to: 1) develop the technology associated with burial ground stabilization, thereby minimizing biotic access and transport of radioactive wastes and, 2) characterize present 300 Area burial grounds to ascertain the potential biotic transport of waste materials away from managed facilities.

Burial Ground Stabilization

This task is designed to test the ability of a "biobarrier" (a rock-gravel-sand-topsoil cover) to prevent plant and animal penetration of contaminated burial grounds. An effective biobarrier must have the following features: 1) uniform thickness and continuous over the entire area; 2) enough elasticity to withstand soil shifting and heaving during construction, freezing and thawing, etc; 3) must prevent roots from entering the buried material; 4) must remain effective for the life of buried radioisotopes; and, 5) should be adaptable to mechanical installation under field conditions.

Different biological barriers are presently being tested in a replicated plot design using soil mixed with lithium chloride to simulate radioactive wastes. Ant colonies and pocket mice have been introduced to the test area. Ant carcasses and soil particles collected from the ant hill will be analyzed for lithium content. The mice will be trapped and feces sampled for lithium content to determine if barrier penetration has occurred. Russian thistle plants have been harvested and analyzed for lithium content. Two plants had significant quantities of lithium in the aboveground parts indicating that the roots had reached the lithium-spiked soil below the cobble layer.

A plant growth test facility has been established within 200 West Area boundaries to ascertain the availability of waste nuclides to plants cultivated under field conditions. Soil properties have been analyzed and the following radionuclides detected in quantities ranging from 50 to 100 pCi/g dry soil: ^{235}U, ^{241}Am, ^{137}Cs, ^{155}Eu, and ^{239}Pu. Several other radioelements were observed in very low concentrations. The plot has been tilled, fertilized, and planted to beans, barley, and carrots. Plant tissues from these crops will be analyzed throughout the growing season.

300 Area Burial Ground Characterization

This study was initiated to gather data pertinent to the question concerning potential environmental hazards associated with the present burial ground technology and includes tasks evaluating the role of plants, small mammals and invertebrates as potential transport vectors of waste materials.

Vegetation samples have been collected from the 300 Area burial grounds and from nearby control areas for radionuclide analyses. Radioisotopes of interest include ^{144}Ce, ^{106}Ru, ^{137}Cs, ^{95}Zr, ^{54}Mn, ^{65}Zn, ^{40}K and ^{60}Co. Significantly greater activity levels were observed for vegetation growing in the Wye and North burial grounds. Cerium-144 was a factor of $\sim$2 higher, ^{106}Ru a factor of $\sim$6 greater, and ^{137}Cs a factor of $\sim$3 greater for the North burial ground. However, it should be noted that the elevated levels observed for plants from the North burial ground resulted from a single, highly-contaminated plant.

The Wye burial ground revealed mean concentration orders-of-magnitude greater than those observed for control areas, for all radioisotopes except ^{40}K and ^{60}Co. However, this observation, again, was highly dependent on the large degree of spatial heterogeneity of nuclide concentrations. All elevated levels were a result of a single contaminated plant. In general, except for the two contaminated vegetation samples out of a total of 191, the concentrations (dpm/gm) of all radioisotopes investigated were not significantly different from observed concentrations in vegetation samples collected from control areas.

Ant Roles in Management of Radioactive Waste Storage Areas

Ants have long been known to be important contributors to soil formation. The activity of these animals over extended periods of time results in the pulverization, granulation, and transfer of large amounts of soil. The soil transfer function may prove significant in managing radioactive waste disposal sites.

There are only a few estimates in the literature concerning the amount of soil ants move to the surface. A study concerning the importance of ants in soil genesis in New England estimated that 50 g/yd^2 were brought to the surface each year. The ant Lasius neoniger Emery was estimated to have brought 86 g/m^2 of soil to the surface in an old field study in Michigan. A harvester ant common in arid areas of the West (Pogonomyrmex occidentalis) was estimated to have excavated 2.8 kg of soil during the life of a colony. This species is closely related to the large red harvester ant common to the Hanford environs. In waste management areas, harvester ants are of particular importance since they tend to invade disturbed areas and usually tunnel to great depths. Their colonies have been reported to extend to depths of 3 m to 4.5 m in arid regions, but depths of 2 m to 2.5 m are probably more common.

This initial investigation was designed to document colony density for the common harvester ant (Pogonomyrmex owyheei). This was accomplished by counting colonies actually located on 300 Area burial grounds. Counts were also made of colony density from adjacent control areas to determine if the burial grounds provide preferred nesting areas (Table 8.5). Results from these studies show that nearly all 300 Area burial ground support substantially greater colony densities than nearby control areas.

TABLE 8.5. Ant Colony Density Counts, 300 Area and Adjacent Burial Grounds

	(No. Colonies/ha)	
	Treatment	
Burial Ground	(Inside Exclosure)	Control
Wye	96	24
300 North	32	14
300 6 and 7	55	2
300 West	0	0
300 4	12	9
Total	195	49

Little is currently known concerning the depths that these ants tunnel into the soil. An initial excavation on the 200 Area Plateau revealed their nest to extend more than 2.5 m into the soil. Additional information is needed concerning colony depths and tunnel construction patterns before estimating the amount of contaminated wastes these ants could bring to the surface.

Mammal Studies

These studies are designed to ascertain radioactive exposure levels to native small mammals inhabiting 300 Area waste management areas. The technique involves live trapping burial ground inhabitants and surgically implanting small dosimeters. The animals are then released and retrapped at a later time. Extraction and analysis of dosimeters then permits a determination of radioactive dose levels impacting resident small mammal populations. An initial comparison of exposure data is shown in Table 8.6.

TABLE 8.6. Exposure to Pocket Mice in mR/Week During April 21, 22, 1977.

Burial Ground	Control Plot
1.68	1.54
1.75	1.40
1.75	2.03
	1.54
	2.24

This shows close similarity between exposure levels for burial ground and control populations of pocket mice. Additional data are currently under analysis.

o Service Assessment Studies

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Other Investigator: R. E. Fitzner

Technical Assistance: H. A. Sweany

Service Assessment studies, supported through the Richland Operations Office, are related to long-term ecological monitoring of important animals on the Hanford Site. This report concerns the Canada goose, the great blue heron and chinook salmon spawning.

Biological Monitoring: Canada Goose Nesting

Biological monitoring has received increasing amounts of attention and funding since the advent of the National Environmental Policy Act of 1969. Some of the most advertised biological monitoring studies are those associated with the siting and operation of commercial nuclear power stations. Such studies are designed to obtain biological data for 1 or 2 yr prior to power station operations to provide baseline information concerning the kinds, and an estimate of abundance, of organisms most likely to be affected by either construction activities or by extended station operations.

Although biological monitoring is generally accepted as a worthwhile practice, there is some concern that the experimental designs of sample-taking, which relate operational factors to changes in wild animal populations, are mostly inadequate. Another concern is that monitoring will need to be conducted over long periods of time, i.e., 30-40 yr, necessitating a long-term funding commitment.

One of the distinct advantages of surveying nesting goose populations on the Hanford Site is that the entire nesting population can be censused rather than sampled, thereby eliminating some of the statistical uncertainties associated with sample-taking. Goose nests are relatively easy to locate since they are almost always restricted to islands that are relatively small and only sparsely covered with plants.

There was a general lack of information concerning the biological fate and effects of radioactivity released into the Columbia River from operating plutonium production reactors. Thus, a program to monitor goose nests was initiated in 1953 to determine if the hatchability of goose eggs was deleteriously affected by the radioactive effluent water being released to the river. These studies showed that the release of radionuclides to the Columbia River by Hanford's production reactors had no deleterious impact on the hatchability of goose eggs.

The production reactors began to be phased out in 1964 and by 1971 eight of nine reactors were no longer operating. Nevertheless, the total number of nests found on the islands diminished from more than 250 nests in the 1950's to about 120 in the 1970's (Figure 8.1). Although this is a dramatic decline, it is not nearly so severe as compared to the virtual loss of the entire island nesting population associated with the nearby Snake River. The decline of the Snake River population is attributed to the inundation of nesting islands by the construction of four hydroelectric dams that have created slack water from the confluence of the Columbia

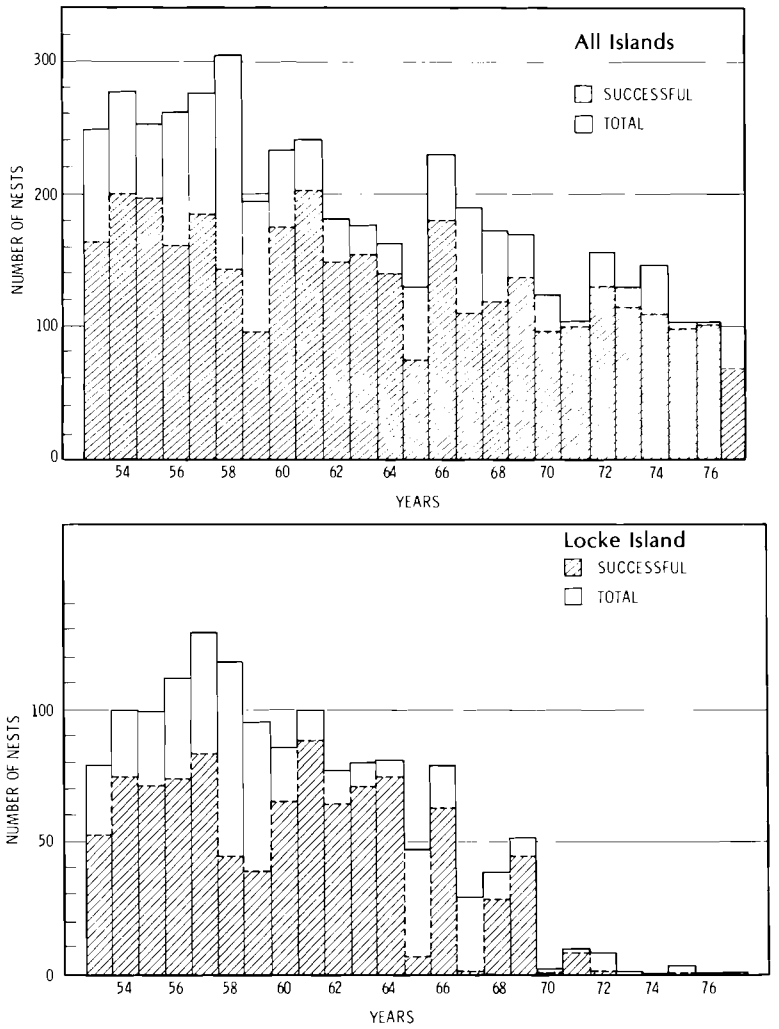


FIGURE 8.1. Total number of goose nests on 20 islands in the Hanford Reach of the Columbia River from 1953-1977: the number of nests that successfully hatched (upper diagram) and the same data for Locke Island (lower diagram), an island that has been occupied by coyotes since 1970.

River to the Idaho border. In contrast, the Columbia River Reach of the Hanford Site has not been dammed.

The steady decline in goose nests can be attributed to human interferences, since the lower reaches of the Columbia River have been subjected to recreational uses over the past decade. Coyote predation is also an important factor. The most dramatic example of coyote predation occurred on Island 6 (Locke Island), which is not subject to human recreation. At one time this island supported more than 100 nests. In autumn 1966, two coyotes swam to Island 6 and remained there until they were shot in February 1968. The number of goose nests increased in 1969 but in 1970 nesting was a complete failure as a coyote occupied the island prior to the nesting season. Since 1970, coyote control has been abandoned altogether and its destructive influence on goose nesting is clearly shown in the following years (Figure 8.1).

The data are now very useful because of the time span involved and their continuity. They now serve as a baseline from which to judge the impact of future environmental perturbations associated with the development of the Columbia River as an energy source and as a water transportation route. A research opportunity is also present to determine if the nesting goose population can be restored to Locke Island by selectively controlling an important goose predator, the coyote.

Radionuclide Content of Rejecta from Heron Nests: A Colony Comparison

One of the environmental concerns associated with the proliferation of nuclear reactors and selection of new sites for the chemical processing of irradiated fuel is the potential for escapement of long-lived radionuclides into the adjacent environment. Once radionuclides enter the biological environment they may participate in food chains. Food chains leading to man are usually well defined and are amenable to radiological surveillance procedures. However, the food chains of higher trophic level animals, other than man, have seldom been investigated in terms of radiological surveillance.

Conventional radiological surveillance techniques involve the collection of domestic meat, fruits and vegetables, milk, eggs, the shooting or trapping of wild animals from time to time and then subjecting organs or tissues to radiochemical analysis. This procedure is useful for sampling wild, gallinaceous birds and waterfowl (birds with high reproductive potential) but is not entirely acceptable for raptorial birds and certain kinds of wading birds with low reproductive potential. For example, shooting for radiological surveillance purposes could seriously deplete small populations of ospreys and great blue herons, fish-eating birds with low reproductive potential.

The great blue heron, Ardea herodias, is a large, fish-eating bird that characteristically nests in the same grove of trees year after year. Nesting begins in early spring with nest refurbishing and egg-laying. Two to five eggs are laid in each nest. When the young birds hatch, parent birds bring food, mostly fish, to the nest to feed the growing young. Although different parents may select different foraging areas located at various distances from the colony, food items are transported to the colony to feed the growing young. Food scraps and fecal material collectively called "rejecta" are cast over the sides of the nests and can be collected and chemically analyzed for year-to-year and site-to-site comparisons.

Cheesecloth blankets spread on the ground beneath heron nests and beneath trees without heron nests at Chatcolet Lake, Idaho, and at the Hanford Site, Washington, showed that worldwide fallout contributed small amounts of radionuclides to cheesecloth through direct aerial deposition (Table 8.7). Control blankets on the Hanford Site accumulated more ash weight than blankets at Lake Chatcolet, largely because of wind-blown dust from the sparsely vegetated ground at the

TABLE 8.7. Average Ash Weight and Radionuclide Content (pCi/g ash) of Cheesecloth Blankets With and Without Accumulations of Heron Rejecta at Hanford, WA and Lake Chatcolet, ID.

Location	Ash wt.	^{137}Cs	^{60}Co	^{40}K	^{144}Ce
		Blankets with Rejecta			
Hanford	290	4.7	0.55	29	7.1
Chatcolet	106	1.2	0.28	16	15
		Control Blankets			
Hanford	66	3.2	1.2	37	20
Chatcolet	6	26	8.7	123	224

Hanford Site. Lake Chatcolet is in a climatically moist and heavily forested region with little blowing dust. When radionuclide content is expressed as pCi/individual control blanket then Hanford blankets accumulated more radioactivity than Chatcolet control blankets. However, when expressed as pCi/g ash weight, the less dusty Chatcolet control blankets had higher concentrations of ^{137}Cs, ^{60}Co, ^{40}K and ^{144}Ce.

Cheesecloth blankets spread beneath heron nests at Hanford accumulated more nest rejecta than blankets of Chatcolet i.e., 289 g at Hanford as compared to only 106 g at Chatcolet. Cheesecloth blankets with accumulated heron rejecta at Hanford also had greater concentrations of ^{137}Cs, ^{60}Co, ^{40}K and ^{144}Ce than Chatcolet blankets. When expressed as pCi/g ash, Hanford blankets with rejecta had greater concentrations of ^{137}Cs, ^{60}Co and ^{40}K, but lesser amounts of ^{144}Ce.

The naturally-occurring radionuclide ^{40}K derived from parent soil and rock was the most abundant radionuclide measured in heron rejecta. Cerium-144, a short-lived fission product, probably had its origin in worldwide fallout. It is possible that the slightly elevated levels of ^{137}Cs and ^{60}Co in rejecta originated from past operations at the Hanford Site but the levels are too low to be of biological significance.

Radionuclide (gamma emitters) content of heron rejecta was very low but measurable at Hanford, Washington, and at Lake Chatcolet, Idaho, indicating that 30 yr of processing nuclear fuels at the Hanford Site have contributed little, if any, radioactivity to present day heron foods. Surveillance of heron rejecta appears to be a sensitive, nondestructive way to survey radionuclide

levels in the foods of a top trophic level animal during a critical phase of its life cycle. The cooperation of Dr. Don Johnson, University of Idaho in locating the Lake Chatcolet heron colony and permission of Heyburn State Park to conduct the study are gratefully acknowledged.

Fall Chinook Salmon Spawning Near Hanford-1976

Annual estimates of fall chinook salmon spawning in the Hanford Reach of the Columbia River, begun in 1947, were continued this year. Continuation of these studies provides baseline ecological data that are potentially useful in the estimation of impacts resulting from continued development of the nuclear industry in this area. Results of past year's surveys are given in previous Annual Reports.

Counts of the numbers of salmon redds (nests), through aerial surveys of the river, serve as a relative index of spawning population size, which is useful for comparing changes among and between years. Because the effectiveness of these surveys is largely dependent on highly variable weather and river conditions, they have limited use in the precise estimate of population size for any 1 yr.

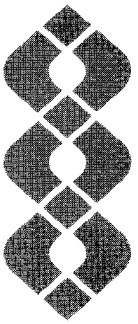
In 1976, surveys were conduted on 12 and 26 October and on 5 and 15 November. Estimates of the number of redds were made from a small aircraft of the Columbia River from river mile 342, near Richland, upstream to Priest Rapids Dam at river mile 396. Survey conditions ranged from poor to good. The spawning area at Midway (river mile 393) continued to be the most difficult to survey due to high river elevations resulting from daytime release of water at Priest Rapids Dam. The start of spawning in most areas was on or after the first survey of 12 October. The exception to this was in the vicinity of Ringold (river mile 354) where spawning was well underway at the time of the first survey. There is a possibility that spawning in this area occurred as early as late September and that some of the early redds had lost their definition and were no longer visible at the time of the first survey of 12 October. The early spawning in the Ringold area may result from past year's releases of juvenile fall chinook from the Washington State Department of Fisheries rearing facility at this location. These releases are from Columbia River salmon stocks obtained from the river drainage downstream of Hanford.

On the surveys of 12 and 26 October and 5 and 15 November, 26, 1305, 1868, and 1618 redds were counted. Total for the season was 1951. This represents about 16% of the adult fall chinook ascending McNary Dam, the nearest dam downstream of Hanford; and about 4% of the fall run over Bonneville Dam, the dam nearest the river mouth (Table 8.8). The 1976 fall chinook passage over both Bonneville and McNary Dams was the highest for the past 10 yr. This was not reflected by high counts at either Priest Rapid or Ice Harbor Dams, upstream of McNary, or by the estimated number of spawners in the Hanford Reach of the Columbia River. It is possible that an underestimation of Hanford spawning contributed to part, but certainly not all of this discrepancy.

TABLE 8.8. Fall Chinook Salmon Passage Over River Dams and Spawning Near Hanford 1967-1976.

Year	Bonneville Dam	McNary Dam	Ice Harbor Dam	Priest Rapids Dam	Hanford[a]	Hanford as % Bonneville	Hanford as % McNary	Unaccounted[b] %
1967	185,643	73,087	19,022	17,330	22,869	12	31	19
1968	159,048	72,757	24,377	22,165	24,920	16	34	2
1969	231,828	79,375	17,507	16,042	31,556	14	40	18
1970	208,902	61,554	10,385	19,884	26,691	13	43	8
1971	202,274	69,718	11,004	12,345	25,200	12	36	30
1972	137,486	49,307	9,436	9,104	6,132	4	12	51
1973	211,127	73,253	8,353	10,083	20,755	10	28	47
1974	186,328	62,009	2,814	7,619	5,096	3	8	75
1975	277,111	68,719	2,558	13,366	18,781	7	27	50
1976	324,786	87,991	1,474	10,780	13,657	4	16	71

[a] No. redds x 7
[b] [McNary-(Hanford + Ice Harbor + Priest Rapids)] ÷ McNary



9.0 Energy Research for Other Agencies

ENERGY RESEARCH FOR OTHER AGENCIES

Nuclear Regulatory Commission

Bonneville Power Administration

Electric Power Research Institute

National Institute of Environmental and Health Sciences

National Oceanic and Atmospheric Administration/ Bureau of Land Management

The purpose of this section is to correlate energy research conducted for other sponsors to U.S. DOE energy programs where interagency and other mutual agreements exist to support work on a nonduplicative basis. The research topics identified in this section are complementary to work discussed in other sections of this report. The data bases being developed provide a unique long-term reference for environmental and energy assessment in the arid West.

Biocide Byproducts in Aquatic Environments (Project Manager: D. R. Anderson; Principal Investigators: R. M. Bean and G. Roesijadi; Other Investigators: R. G. Riley, T. O. Thatcher, S. W. Li; Specialist: E. W. Lusty and C. S. Abernethy)

More electrical generating plants are being built each year to meet the additional demands for electricity. Assuming the continued use of chlorine as a biocide in power plants, there will be a significant increase in the volume of chlorinated effluents entering aquatic environments. Associated with these effluents will be chlorinated (halogenated) organic byproducts.

This program is designed to define an information base that can be used to generate or modify regulations concerning chlorination byproducts. The first step in determining the magnitude of this potential problem is to identify the chlorination byproducts and their effects and fates on biota and in aquatic ecosystems. The study has been divided into analytical chemistry and biological studies, which includes freshwater and marine subdivisions. The objective of the analytical phase is to identify those chemical compounds, other than the "free and combined available halogen," which result from the addition of chlorine to fresh or saltwater. The objectives of the biological studies are to investigate the immediate and relatively long-term toxicity of several chlorination byproducts; to follow their pathways of action; to analyze for bioaccumulation or biomagnification; and to evaluate these effects on rainbow trout, (Salmo gairdneri), and the littleneck clam (Protothaca staminea). The program is expanding to encompass more freshwater and marine species during the second year of work.

Analytical Chemistry of Chlorination Byproducts

Initial experimentation has been directed toward isolating and identifying halogenated components formed in natural waters under conditions similar to those used in cooling water treatment, particularly those halogenated compounds that might be expected to be absorbed and biomagnified in the lipids of aquatic biota. Thus, we investigated the nonpolar, presumably lipophylic organohalogens formed. The procedures used to isolate these compounds would specifically exclude more polar component types such as halogenated phenols, amines, and nitrogen heterocycles that are being investigated using other methodologies.

Samples of chlorinated natural waters were obtained from a continuous flow apparatus designed to simulate conditions of current power plant cooling water treatment practice. Organic components were concentrated by forcing chlorinated and unchlorinated water through columns of XAD-2 resin using a positive displacement pump. Ether extracts of the XAD-2 columns were analyzed for haloforms by gas chromatography, using a 30 m wall-coated, open tubular capillary column. An electron-capture gas chromatogram obtained from a sample of chlorinated seawater is given in Figure 9.1. Bromoform was found to be the major constituent in all chlorinated seawater samples. In contrast, chloroform was the only haloform found from chlorinated freshwater.

The procedures used for the analysis of XAD-2 extracts for components other than haloforms involved a number of preliminary separation steps prior to gas chromatographic analysis. Gel permeation chromatography was first used to remove material having molecular weight greater than 800. Silica gel chromatography using hexane, then hexane-ether, was used to separate the sample into two fractions. Fraction A contained nonpolar components (e.g., hydrocarbons and halocarbons) and Fraction B contained components of intermediate polarity (e.g., ketones and esters).

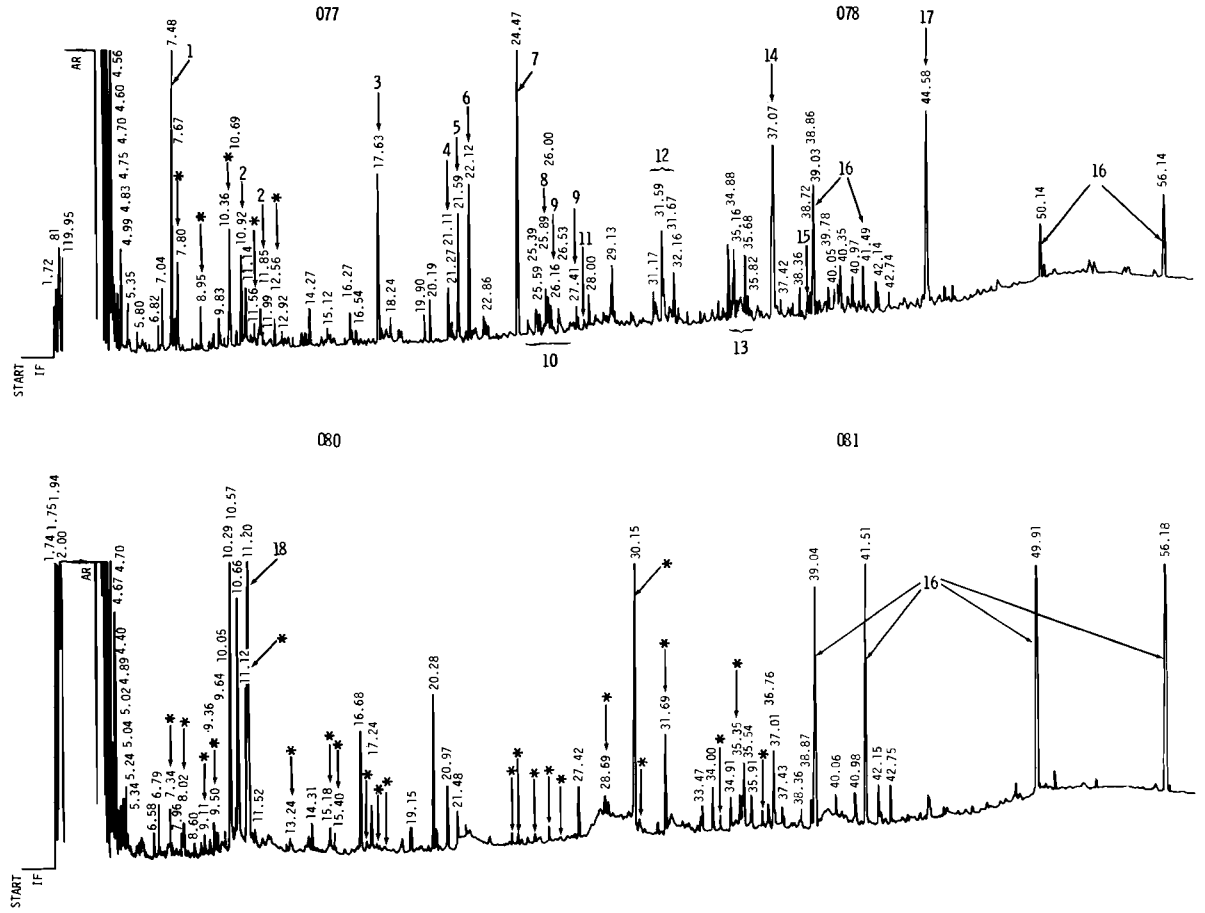


FIGURE 9.1. Above, Gas Chromatogram of Fraction A from XAD-2 Extract of Seawater Using Flame Ionization Detector; Below, Chromatogram of Fraction B. Chromatographic peaks denoted by an asterisk represent unidentified bromine-containing components (confirmed by GC/MS and electron capture). Peaks denoted by numbers have been identified and are listed by number on Table 9.1.

Capillary chromatograms obtained from Fraction A and B using a flame ionization detector are shown in Figure 9.2. These two fractions were derived from the XAD-2 extract of over 600 ℓ of chlorinated seawater. Final volume of the fractions was adjusted to less than 50 μl prior to GC analysis. The chromatographic peaks denoted by asterisks or numbers represent components that met two criteria: 1) exhibited response from the electron capture detector, and 2) gave chemical ionization mass spectra having peaks characteristic of bromine isotope ratios. A component was required to meet both of these criteria to positively designate it as bromine-containing. The identities of numbered peaks in Figure 9.2 are listed on Table 9.1.

Many of the components identified in Fraction A are aromatic hydrocarbons which were found in all samples studied. Procedural blanks did not contain these hydrocarbons. Our present belief is that these components are initially present in the XAD-2 polymer matrix in an unextractable form and are released during the sampling operation, possibly the result of fracturing of the resin particles as the bed is compacted. The brominated aromatic components in Fraction A are probably formed by reaction of bromine with the hydrocarbon impurities during sampling.

On the basis of our results thus far, the concentrations of nonpolar and presumably lipophylic halogenated components generated by the low level chlorination of relatively pristine natural waters appear to be very low, in the ng/ℓ range, with the exception of the haloforms. Although destruction of active chlorine prior to XAD adsorption should eliminate brominated artifacts, continued problems with contaminates can be expected since many substances are ubiquitous in the environment at these levels.

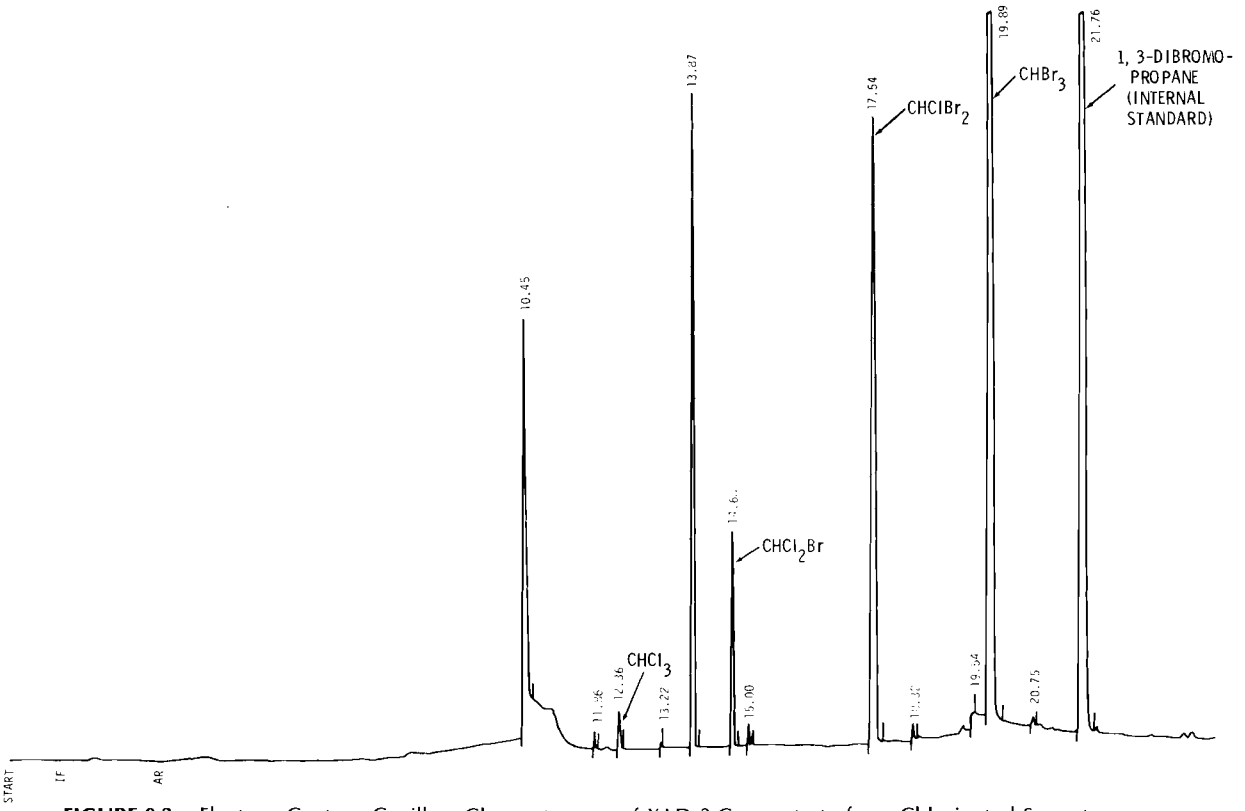


FIGURE 9.2. Electron Capture Capillary Chromatogram of XAD-2 Concentrate from Chlorinated Seawater

TABLE 9.1. Components Identified by GC/MS in Fractions A and B Obtained from XAD-2 Extract of Seawater[a]

1. Bromoform
2. Trimethylbenzene
3. Naphthalene
4. Bromotrimethylbenzene
5. 2-Methylnaphthalene
6. 1-Methylnaphthalene
7. Biphenyl
8. Bromotetralin
9. Brometetramethylbenzene
10. Dimethylnaphthalenes
11. Bromonaphthalene
12. Bromomethylnaphthalenes
13. Bromobiphenyls and Bromodimethylnaphthalenes
14. Phenanthrene
15. Bromotrimethylnaphthalene
16. Phthalate Esters
17. Pyrene
18. Bromoacetal

[a]Hydrocarbons were identified by comparison of spectra with authentic samples; identities of brominated components were assigned solely on the basis of spectral interpretation.

Biological Studies of Chlorination Byproducts

The objectives are to investigate the immediate and relatively long-term toxicity of several chlorination byproducts; to analyze for bioaccumulation or biomagnification; and to evaluate these effects on the growth of rainbow trout, (Salmo gairdneri), and the marine littleneck clam (Protothaca staminae). The biological studies are subdivided into freshwater and marine or estuarine programs.

In the marine studies, clams were exposed in a chronic bioassay to chlorinated seawater for 6 months to examine the uptake of halogenated organic compounds and their effect on growth and mortality of Protothaca staminea. Chlorination and dilution in a chlorine contact chamber produced total residual oxidant (TRO) levels ranging from 0.01 to 0.14 mg/l. The resultant exposure chamber concentrations ranged from undetectable to 0.02 mg/l. This decrease is presumably due to chlorine demand of the seawater. Samples of exposed and control clams were taken at 1, 2, 3 and 6 month intervals and preserved. Specimens were frozen for later analysis of tissues for halogenated compounds, and others were preserved in an appropriate fixative for histological examination. Growth of exposed clams, measured as increase in length of shell, was also examined during the 6-month exposure.

New exposure systems were recently completed which supply both relatively high concentrations necessary for chemical characterization of chlorinated seawater or freshwater and the low concentrations required for long-term fish or clam exposures. In order to simulate power plant conditions, the systems incorporate a temperature increase during passage through the chlorine contact chambers. The level of chlorination in the contact chamber is approximately 1.5 mg/l TRO.

In the upcoming year, growth experiments will be continued, emphasizing an energetics approach to measure clam productivity. Samples for bioaccumulation will continue to be collected and preserved for analysis. Chronic bioassays are being conducted with juvenile rainbow trout to determine the long-term effects of chlorination byproducts. The test water is chlorinated in a chlorine contact chamber prior to entering the bioassay system. Fish are exposed in a naturally fluctuating temperature regime to chlorination levels of less than 0.04 mg/ℓ total residual chlorine (TRCl). Although the mortality rate throughout the experiment is low, less than two fish/week, the total number of mortalities after 6 months indicates an interesting trend with mortality and TRCl, or the corresponding concentration of chlorination byproducts. Preliminary indications are that trends in mortality have a positive correlation with increasing levels of chlorinated organics. Using Fulton's coefficient of condition $K=\frac{W}{l^3}$ as an indication of the general health of the fish, there is a significant difference between controls and 50% of the contact chamber levels of chlorination byproducts.

Design and Analysis of Aquatic Monitoring Programs (Principal Investigators: D. H. McKenzie, K. L. Gore and L. D. Kannberg)

Our efforts during the past fiscal year have been devoted to: 1) reviewing the statistical methods used in impact analyses, 2) developing what we have termed the paired station analysis of variance approach and applying the technique to several existing data bases, 3) examining the appropriate error rates, detectable differences and sample size requirements for this technique, 4) reviewing the state-of-the-art of hydrological monitoring, modeling techniques and model verification and 5) examining the sensitivity of a hydrological model to the quality and quantity of the monitoring data.

The project addressed some of the problems of designing, conducting and analyzing aquatic environmental monitoring programs for impact assessment. Our objective was to develop usable guidelines that would hopefully avoid some of the problems and pitfalls of earlier monitoring programs. The reports from an earlier "three laboratory study" (ANL, PNL and ORNL) provided the impetus and data bases for this project.

The concept of paired control and treatment station pairs is the fundamental basis for the experimental method proposed in this report. This concept is based on the hypothesis that the relationship between the two stations forming the pair can be estimated from the preoperational period. Any changes observed in this relationship during the operational period are assumed to be the result of the power plant impacts. Thus, it is important that station pairs are selected so it can be assumed that they respond to natural environmental changes in a manner that maintains this relationship.

The major problem in establishing the station pairs will be the location of the control station. The universal heterogenity in the environment will prevent the establishment of identical station pairs. The requirement that the control station remain unaffected by the operation of the power plant dictates a spacial separation with its associated differences in habitat. Thus, selection of the control station will be based upon balancing the following two

criteria: 1) far enough away from the plant site to be beyond the plant influence, and 2) close enough to the treatment station that the biological communities will respond consistently to natural environmental changes.

It appears that the paired station requirements can be achieved for some components of the aquatic ecosystem, i.e., phytoplankton, zooplankton, and benthos. The Zion Nuclear Power Station monitoring program is a good example of the application of this concept to a large lake environment, Lake Michigan. The application of the method to riverine, estuarine and marine environments is complicated by varying hydrodynamic factors and environmental gradients. Additional study is required to evaluate the station concept in these situations.

The application of the station pairing concept to the fish population monitoring program does not appear promising. The concept of a control station unaffected by the power plant is unlikely to be valid. The mobility and migratory behavior of most fish species tends to distribute any plant-induced impact over the entire population. Thus, the density at a "control" station would not be independent of the plant-induced impacts. Therefore, the assessment of impact on fish populations must proceed at a population level and cannot be solely based on station differences. Additional study is required to evaluate the role of the fish monitoring program in the assessment of fisheries impacts.

Another important concept in the design of aquatic monitoring programs is the collection of balanced data sets. One should avoid monitoring schemes that produce data sets which contain missing observations. In addition, the design should be such that each of the analysis of variance factors has equal numbers of observations at each level of the factor. For example, month effects are usually included in the analysis of variance model and a balanced data set would contain an equal number of samples collected each month. Other common factors for which balancing is desirable include: water depth, substrate type, current velocities, upstream-downstream directions, and preoperational-operational periods. Balanced data sets are much easier to analyze and interpret, both biologically and statistically.

The requirements placed on the data collection phase of the monitoring program can be summarized in one word: consistency! This would seem to be intuitively obvious, but nearly all of the nine monitoring programs reviewed by the three laboratories contained design changes that affected the usefulness and interpretations of the data base. Changes in collecting gear, station locations, and personnel (usually environmental monitoring contractors) can seriously confound the interpretation of monitoring data. When changes are unavoidable or fully justified, an orderly transition period is essential to preserve the comparability of the data bases.

There is a general lack of uniformity between the level of detail in the data collection phase and the analysis and interpretation phases. The data collection phase generally measures and records everything in the greatest detail practical. Conversely, the analysis and interpretation appears to proceed at the least level of detail possible. For example, phytoplankton data bases usually contain densities for the organisms identified to the genus or species level, but the analysis and interpretation of change may only consider total densities. Guidelines for the level of data identification and analysis are presented in an attempt to resolve the lack of uniformity and increase the cost effectiveness of monitoring programs.

The proposed statistical techniques have been applied to portions of the nine power plant data bases. While the methodology represents an improvement over the majority of the methods used by the three laboratories, additional work is required to verify the generic applicability of the method and the parameter estimates. With the above caveat, the proposed method is an improvement in the ability to statistically detect changes in the organism densities. Based on the Zion zooplankton data the proposed method is four to seven times more efficient than the commonly used, unpaired analysis.

Guidelines were also proposed for some of the problems encountered during the statistical analysis of the data. An experimentwise error rate was suggested where the lower trophic levels and economically important species are treated as the experimental unit. Levels of the statistical error rates for testing hypotheses were proposed. The testing of the hypothesis about plant-induced changes indicates that the sample sizes presently being collected permit the detection of approximately 50% or smaller changes at the 0.10 significance level and with a power of 0.8. Additional study is needed to establish whether 50% changes are appropriate to the interpretation and evaluation of impacts.

Environmental Development Plans - Nuclear (Principal Investigator: R. F. Foster)

In December 1976, ERDA issued Immediate Action Directive No. 0500-4 establishing the requirement for Environmental Development Plans (EDP's) for energy systems. One of the purposes of EDP's is "to provide the Assistant Administrator for Environment and Safety with the basis: 1) for planning support of technical programs; 2) for determining common needs for environmental, health, and safety research and development control technology; and 3) for discharging responsibility for corrective environmental overview and assessment."

PNL staff assisted ERDA in the development of EDP's for several energy systems, but the contributions of the Ecosystems Department were especially great on those managed by Nuclear Technology Overview Programs. These contributions included input to the conceptual design for the nuclear EDP's and the generation of major portions of the plans for: 1) Defense Waste Management, 2) Special Nuclear Materials, and 3) Decommissioning and Decontamination. Because of this participation and the future implementation of the EDP's, PNL will be able to focus even more sharply on the priority problems in planning future environmental research work.

Bonneville Power Administration

Biological Studies at the Site of an 1200-KV Prototype Transmission Line (Principal Investigator: L. E. Rogers)

This study, supported by the Bonneville Power Administration, is designed to determine whether enviromental effects, detectable by current field sampling techniques, exist which are associated with operation of a 1200-KV power line.

Preliminary analysis in terms of organic productivity and morphology of plant species, behavioral disturbances in cattle and honey bee populations and in the population dynamics of small mammals indicates a lack of significant impact. Some discoloration of Douglas fir tree needles was noted for branches near the line. This is the type of effect which has been associated with corona discharge although the actual mechanism accounting for needle necrosis is not clear at this time. Needle damage does not necessarily imply that adverse effects to tree growth has occurred. Data on annual tree growth are currently being analyzed.

Electric Power Research Institute

Synthesis and Analysis of Cooling Impoundment Information (Principal Investigators: C. D. Becker, C. E. Cushing and K. L. Gore)

This program was initiated in January 1977 for the Environmental Assessment Department of the Electric Power Research Institute. Electric utilities are presently experiencing a number of power plant siting difficulties as a result of: 1) local, state and Federal environmental protection requirements, 2) the action of environmentalists, and 3) community concern regarding societal impacts. One specific problem area concerns the disposal of waste heat added to circulating cooling water during plant operation. For this purpose, the use of semienclosed cooling impoundments, as managed aquatic ecosystems, appears to be an economically and environmentally acceptable alternative.

We are evaluating the ecological impact of power plant operation on cooling impoundments with load ratios (pond surface acres/plant capacity in MWe) less than ten, which imparts relatively high thermal loading. We believe that adverse impacts on aquatic environments could be detected and quantified most readily under such conditions. Consequently, "monitoring" data available in the published and unpublished literature is undergoing synthesis and analysis for technically sound conclusions concerning the effects of power plant operations.

This research is integrated with and complements several studies concerning environmental impact assessment supported by the U.S. DOE in the Freshwater Sciences Section.

National Oceanic and Atmospheric Administration/ Bureau of Land Management

Accumulation of Organic Constituents and Heavy Metals from Petroleum Impacted Sediments by Marine Detritivores (Principal Investigator: J. W. Anderson)

This research complements that conducted under U.S. DOE funding, since it relates to bioavailability of hydrocarbons from sediment. While the techniques and organisms differ, the findings will aid us in evaluating the fate and effects of petroleum hydrocarbons in marine organisms and the marine environment.

Research on the bioavailability of hydrocarbons and trace metals from oil-impacted sediments during FY77 has resulted in publication or submission of the five manuscripts listed below:

Reports

Anderson, J. W., G. Roesijadi, D. L. Woodruff and E. A. Crecelius. 1977. Accumulation of Organic Constituents and Heavy Metals from Petroleum Impacted Sediments by Marine Detritivores. Annual Report to NOAA, April, 1977.

Anderson, J. W., G. Roesijadi and E. A. Crecelius. 1977. Bioavailability of Hydrocarbons and Heavy Metals to Marine Detritivores from Oil-Impacted Sediments. OCSEAP Program Review Report, November, 1977. To be published in NOAA Technical Report Series.

Publications

Roesijadi, G., D. L. Woodruff and J. W. Anderson. 1978. Bioavailability of naphthalenes from marine sediments artificially contaminated with Prudhoe Bay crude oil. Environmental Pollution (in press).

Roesijadi, G., J. W. Anderson, and J. W. Blaylock. 1978. Uptake of hydrocarbons from marine sediments contaminated with Prudhoe Bay crude oil: Influence of feeding type of test species and availability of polycyclic aromatic hydrocarbons. J. Fish. Res. Bd. Canada (in press).

Roesijadi, G., and J. W. Anderson. 1978. Condition index and free amino acid content of Macoma inquinata exposed to oil-contaminated marine sediments. In: 1977 Symposium on Pollution and Physiology of Marine Organisms. Georgetown, S.C., ed. by Winoma and F. J. Vernberg, Academic Press, New York (in press).

National Institute of Environmental and Health Sciences

Fate of Heavy Metals and Heavy Metal Complexes in Soils and Plants (Principal Investigator: R. E. Wildung)

The program underway for the National Institute of Environmental Health Sciences is directed toward understanding the influence of soil, microbial and plant physiological processes on the form and behavior in soils and plants of trace metals arising from energy production.

Trace elements may enter the environment as a result of extraction, combustion, upgrading and refining of fossil fuels; production of synfuels; and the utilization of geothermal energy. Little is known of the effects on the environment and on human health resulting from alteration of the sensitive concentration balance of trace metals in the environment, as investigations of these phenomena are relatively recent. However, a background of information to predict the behavior of these elements is available from studies of radionuclides. Furthermore, use of radiotracers is essential to studies of trace metals where problems of contamination and detection require rapid, sensitive, and specific detection methods.

The studies are providing an integrated view of the potential for trace metal-organometal complex formation in soil, uptake by plants, localization in edible tissues and gastrointestinal transfer in animals. This information has provided a basis for assessment of problems

associated with entrance of trace metals into the environment as a result of energy production and a unique insight into the effect of similar transformations on the long-term behavior of radionuclides. Nuclear science has, in turn, provided the tools and a part of the background necessary to conduct experiments appropriate to development of this understanding.

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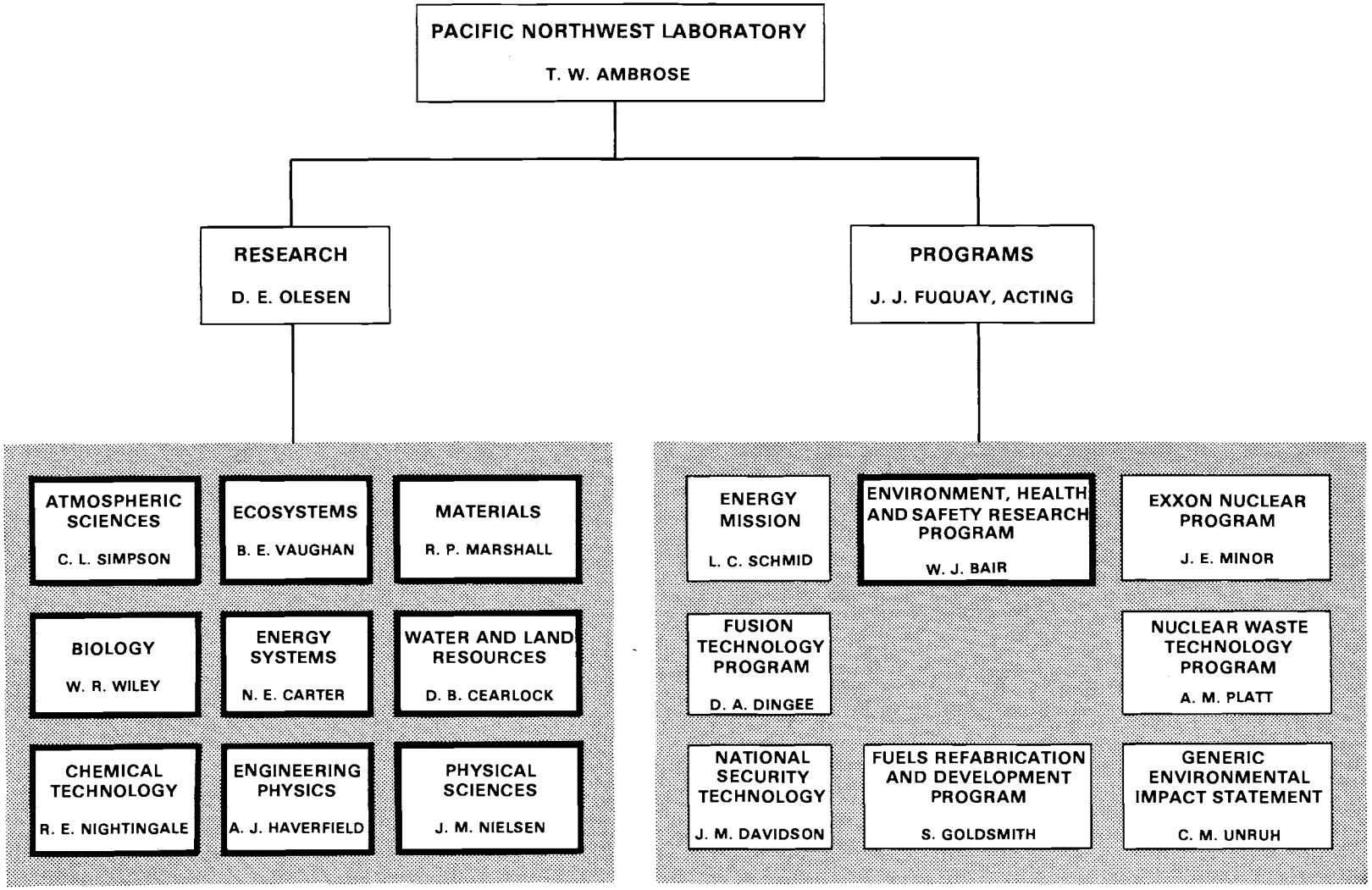

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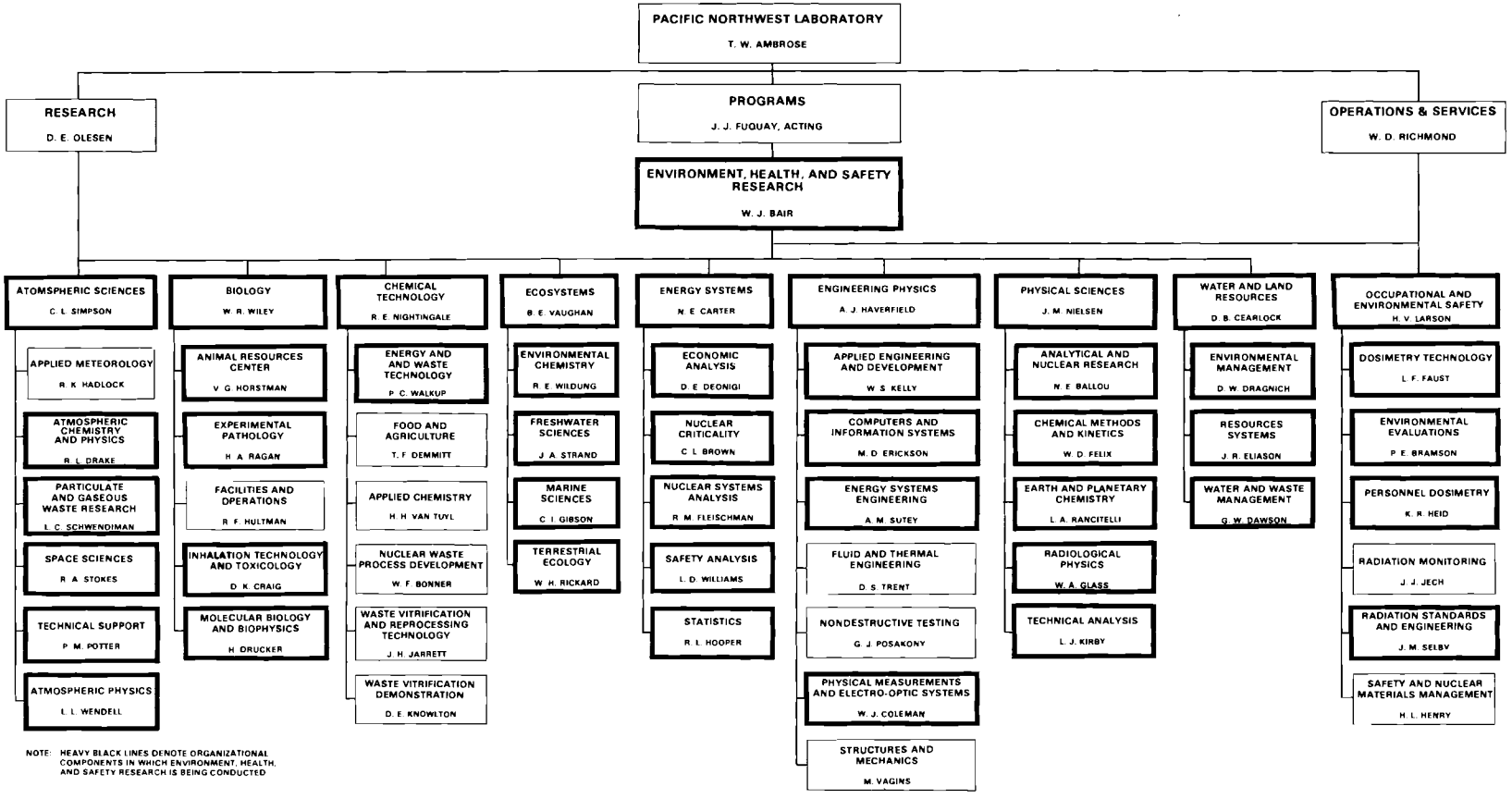
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