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MASTER

THE FATE OF NUCLIDES IN NATURAL WATER SYSTEMS

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DISCLAIMER

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I. PROJECT SUMMARY

A. <u>Summary of research projects</u>: The northeastern part of the United States is characterized by the use of virtually every form of energy source. Nuclear and fossil fuel sources predominate at the present time. Materials involved in energy production pass in one way or another through the environment and in this region particularly impact the estuarine and coastal zones. Some of the materials related to the environment as the result of energy-related activity can be uniquely identified while some others are virtually indistinguishable from naturally introduced substances or materials mobilized by man's general activities.

It has been our goal to use all types of information resulting from the study of the distribution of specific energy-related substances, materials mobilized by man or natural substances in order to determine the fate of materials in nearshore environments. We believe that a general approach to the problem will be the most fruitful for understanding the ultimate fate of materials in the coastal zone.

In our estuarine and coastal work we have concentrated, in the past few years, on the Long Island Sound and the New York Bight - both areas heavily impacted by man but each with its own characteristic physical setting.

Long Island Sound, although generally obeying the rules of estuarine circulation also behaves like a large basin, protected from the effects of the intensive storm energy of the Atlantic Ocean to which the New York Bight is subject. Long Island Sound, therefore, can accumulate materials on a continuous basis and maintain a record that is not modified significantly by storm activity. Biological activity in the sediment column, however, does have a strong influence in determining the nature of this record.

The New York Bight, on the other hand, is subject to intense bottom reworking as the result of Atlantic storms. It is also subject to strong long-shore currents resulting from this energy. The fine-grained sediments are therefore patchilly and ephemerally distributed throughout the Bight.

Both regions are characterized by high turbidity which is the result of resuspended sediment and biological debris and this markedly affects the properties of the water column. Both regions are the recipients of materials from the natural erosion of the adjacent drainage basins and from the activities of man.

The following is a review of our results during the past year and the plans we are making for next year growing out of our acquired knowledge of the coastal system. The relationship of each research program to energy-related activities is identified.

1. The residence time of dissolved "reactive" metals in the coastal zone is strongly controlled by the particle concentration in the water column. We have determined this in two ways: (a) In the New York Bight we have used ^{234}Th (24 day half life) and ^{228}Th (1.9 year half life) produced in the water column from ^{238}U and ^{228}Ra respectively to determine the residence time of thorium in the water column as a function of distance from the Long Island-New Jersey coast. The residence time increases from less than 10 days at the Bight Apex to greater than 100 days at the continental shelf margin. (b) In Long Island Sound an assay of the ^{234}Th standing crop in the sediments indicates that virtually all of the ^{234}Th is taken to the sediment column on a very short (less than 10 days, probably 1 day) time scale.

We had earlier established that the ^{210}Pb, Pb, Cu and Zn delivered to Long Island Sound are also quantitatively removed to the sediments there.

The energy-related consequence of these studies is that it indicates that "reactive" nuclides impacting the coastal zone, whether from nuclear reactors, atmospheric emission from fossil fuel reactors, or industrial sources, will all end up primarily in estuarine deposits and be retained there.

2. Redistribution of metals in the sediment column follows three paths. In the storm protected Long Island Sound, tidal currents, reinforced by storm coupling at times, result in the frequent if not continuous redistribution of the top one centimeter of the sediment pile. This results in rapid homogenization of chemical properties perpendicular to the axis of the Sound (tidal displacement is along this axis) with redistribution along the axis proceeding on a much slower time scale. The role of the benthic organism community in terminally sequestering pollutants in the Sound appears to be important. The deeper parts of the Sound have deeper bioturbating activity and thus a greater penetration and storage of "reactive" metals and other contaminants.

The energy-related aspect of this study is that it identifies ultimate repositories of "reactive" nuclides introduced into the coastal zone. Of particular significance is our observation that the behavior of plutonium (as well as the other metals) can be predicted by the behavior of natural ^{210}Pb. Both nuclides have their highest standing crops and deepest sediment penetrations in the deeper parts of the Sound.

Thus not only are metals sequestered by particles out of the water column but they are homogenized in small areal domains and finally permanently sequestered most efficiently in the deep burrowing habitat regions of the system.

The third mode of transport is primarily found in the New York Bight where intense Atlantic storm activity redistributes the fine-grained material periodically. This tends to homogenize the properties of this material to yield virtually homogeneous fine-grained pockets of an ephemeral nature throughout the Bight. The behavior of plutonium can again be predicted from the behavior of ^{210}Pb.

3. Where streams meet the sea, radical changes in chemical properties take place not only because of the change in salinity but also because of the establishment of estuarine circulation resulting in the renewal of nutrients in the surface waters and the development of characteristic ecosystems such as salt marshes. We have been studying the behavior of the uranium decay series nuclides and ^{228}Ra in this region to determine the changes that take place in the water and on particles as this boundary is encountered. We have shown that in the virtually pristine Connecticut River estuary radium is released into the water column at low tide at least. As silicon and barium are also released there is a suspicion that salt marshes, so prominently present in this system, may be taking part in modulating the chemistry of the boundary region. This has important consequences for other elements as well especially in causing us to reevaluate the way of determining riverine fluxes of metals to the oceans.

The energy-related aspect of this research is immediately obvious. The Connecticut Yankee nuclear reactor is on the Connecticut River stretch we are studying and the role of salt marshes and the estuarine zone in general, in response to radionuclides that might be injected into the system, must be assayed.

4. We have been studying the sources and properties of organic carbon delivered to estuarine systems. This was the direct result of our detailed study of a Long Island Sound core for carbon-14 distribution with depth. We observed the effects of a sharp increase in the use of fossil fuel in the region in the latter part of the 19th century. We also observed that the high flux of biogenic carbon produced in the Sound is eliminated with a mean residence time in the top 5 cm of the sediment of 28 days. We believe this rate of degradation also holds for sewage sludge based on our few analyses of the New York Bight sediments.

The most striking observation was that the sediments of Long Island Sound, long before fossil fuel was introduced, contained about 1.2% organic carbon with a C-14 age of 2300 years BP at the moment of deposition. As planktonic material would have a zero age at the time of introduction this obviously is not dominantly refractory marine organic debris but soil organic matter brought down by the rivers.

This kindled an interest in the very complex field of particulate organic transport and storage. The significance to the energy program is twofold. First, this part of the carbon cycle is obviously so poorly known that it becomes a serious constraint on our full understanding of the behavior of the carbon dioxide system as it relates to land plant and organic matter storage. We do not know the residence time of "refractory" carbon in soils well enough for modelling of the carbon system. Secondly, the accumulation of land derived organic matter has long been known to be a possible source of petroleum after suitable metamorphosis. Although one does not think of Long Island Sound as a future petroleum resource certainly the mechanism and rate of emplacement of detrital land-derived organic matter will assist us in sharpening some parts of our search for additional petroleum resources. For these reasons we intend to continue our investigation of the coupling of the land and marine carbon system using the radioactive and stable isotopes of carbon as tracers and timepieces.

The reports in the following sections represent the status of the work we are engaged in. Some of these are not addressed in detail in this section as they represent new work which is still being developed.

B. Communication of results: It has been our policy to prepare the results of our research for publication in journals as they become completed. Prior to final publication, however, preliminary results are presented at informal internal seminars, scientific meetings, and as seminar talks at various institutions by members of the research staff. The institutions this past year include: Woods Hole Oceanographic Institution, Lamont-Doherty Geological Observatory (Columbia University), University of Chicago, UCLA, Scripps Institution of Oceanography, University of Connecticut, University of Ottawa, and the Geological Survey of Canada.

In addition we have participated in the DOE Contractors' meeting in March at Rockville, the Workshop of SCOR Working Group 46 (RIOS) in March in Rome, and the Long Island Sound Conference in May at the University of Bridgeport.

Visitors from other institutions have come to Yale and participated in seminars and extended discussions about the general subject matter of the grant.

C. Personnel: Various members of our laboratory have participated in research supported by the DOE grant even if most or part of their salary support comes from other sources.

Dr. Y. Nozaki was a Research Associate until November 30, 1979. He participated in the radiocarbon work and also the PARFLUX experiments (see last year's Progress Report) in collaboration with Woods Hole Oceanographic Institution. He has continued this research at Woods Hole as a member of their staff.

Dr. J. Kirk Cochran is supported primarily on a grant from the Petroleum Research Fund of the American Chemical Society while working on projects associated with this grant. W. C. Graustein has been involved in an informal way pending the completion of his thesis after which he will join our staff.

Eric P. Dion is an advanced graduate student supported wholly by the DOE grant. He has been involved in a number of projects but his main effort has gone into establishing our Connecticut River estuary program which will constitute the major part of his thesis.

Technical and secretarial support were provided in part from this grant. These include A. Phelps (secretary) and E. Aaboe (technician).

II. Papers Published or Submitted for Publication

A. Radiocarbon dating of a core from Long Island Sound

B. The ^{210}Pb chronology of a core from Mirror Lake, New Hampshire

C. Trace metals in the sediments and biota of New Haven harbor and Long Island Sound

D. Tracking particle associated processes in nearshore environments by use of ^{234}Th/^{238}U disequilibrium

E. Cosmic-ray produced ^{7}Be: its application to the study of particle mixing rates in near-shore and lake sediments

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