PROGRESS REPORT

DOE/ER/60595--3

I. Research accomplishments

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Our research at Yale on the fate of nuclides in natural water systems has three components to it: the study of the atmospheric precipitation of radionuclides and other chemical species; the study of the behavior of natural radionuclides in groundwater and hydrothermal systems; and understanding the controls on the distribution of radionuclides and stable nuclides in the marine realm. In this section a review of our progress in each of these areas is presented. This review is based on the papers and preliminary reports in the Attachment to this Progress Report and Proposal.

A. Atmospheric precipitation of radionuclides and other chemical species:

About 15 years ago we showed the possibility of determining the precipitation flux of ^{210}Pb (22 year), produced by the decay of atmospheric ^{222}Rn (3.8 day) by measuring ^{226}Ra and ^{210}Pb in soil profiles. This led to the beginning of an extensive program of soil profile sampling, atmospheric aerosol sampling and modeling to determine the worldwide fluxes of ^{210}Pb to the earth's surface and also the conjugate flux of ^{222}Rn from soils to the atmosphere. Our fundamental review of the state of the art was published in 1977 (Turekian, Nozaki and Benninger, Ann. Rev. Earth Planet. Sci., v. 7). This work has led to contributions to the GEOSECS program, work on acid rain fluxes, regional studies of soil radon fluxes and the SEAREX program. Although each of these programs took on a life of its own, supported by various agencies over the last 15 years, it is the work we were able to start and sustain as the result of this ongoing DOE grant.

In the Attachment are included two papers resulting in part from contributions of this grant to an atmospheric project. The work on soil profiles in the Hawaiian islands established that the fluxes of ^{210}Pb (and by analogy, other aerosols) and ^{137}Cs are controlled by the precipitation flux and show no difference in the ratio of the fluxes of ^{210}Pb to ^{137}Cs that could be ascribed to orographic effects or

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to the supply flux of ^{210}Pb from volcanic sources (see Attachment: Turekian, Graustein and Cochran).

We have measured the ^{210}Pb flux from a recent Kilauea eruption and made laboratory measurements to determine the controls on ^{210}Pb emissions from volcanoes in order to assess the expected local and worldwide burden of ^{210}Pb in the atmosphere from this source. These results are described in a report by Reinitz in the Attachment.

The orographic effects to which I alluded are seen on continental mountain systems and a recent paper by Graustein and Turekian (Attachment) shows that the continental boundary layer area has a higher $^{210}\text{Pb}/^{137}\text{Cs}$ ratio than mid tropospheric air. High mountains whose crests rise into the low $^{210}\text{Pb}/^{137}\text{Cs}$ domain receive most of their aerosol flux via cloud droplet impact and feeder-seeder mechanisms from orographic lifting of the continental boundary layer air. Thus the impact of aerosols derived adjacent to a mountain system is greater than that of long distance traveled mid troposphere aerosols.

In work performed on our DOE grant on salt marshes and published 10 years ago we showed that atmospherically derived ^{210}Pb accumulating in a salt marsh preserved the total flux without migration and was useful in determining the rate of rise of sea level over the past 100 years. In addition, the fluxes of other atmospherically delivered metals could be determined as a function of time to assess the changing contribution of the anthropogenic burden to the air. We have been continuing that work and established that the flux of metals was diminished along the Connecticut coast, as expected, the more remote the location from the New York metropolitan region. Our recent work also showed the preservation of the atmospheric pattern of ^{137}Cs (but not its absolute flux) in salt marshes thus providing another useful chronometric point (Attachment: Green, Graustein and Turekian).

Our most recent extension into the use of salt marshes was to determine the flux of anthropogenic osmium, with its characteristic isotopic composition, to coastal deposits. We have established that the major source of osmium (as well as other metals) to New Haven harbor was sewer outfalls but we weren't sure how far the impact of this loading was felt in Long Island Sound (Attachment: Esser and Turekian). By demonstrating that the atmospheric flux of osmium in a salt marsh was indistinguishable from the flux of osmium to an adjacent marine sedimentary deposit, the confining of the pollution to New Haven harbor was indicated (Attachment: Esser).

B. The behavior of natural radionuclides in groundwater and hydrothermal systems: As part of our DOE funded research we earlier showed how the use of members of the uranium and thorium decay chains dissolved in groundwater could be used to determine the adsorption, desorption coefficients of ions in the aquifer and thus the retardation factor for these ions. The method depended on the assumption that the recoil flux of ^{222}Rn (3.8 day) could be used as a measure of the recoil fluxes of the other nuclides. This would not be strictly true if recoil took place in "nanopores". The experiment by Krishnaswami and Seidemann (Attachment) using artificially recoiled ^{39}Ar and ^{37}Ar in conjunction with naturally recoiled ^{222}Rn demonstrated that the source of recoil radon, and by extension the other recoiled nuclides, was not in nanopores but surface deposits of radium (and its precursors). This observation validated, for us, the procedure we used for determining retardation factors for nuclides in groundwaters. This procedure has importance for the nuclear waste disposal problem and is being continued with a contract from Lawrence Livermore National Laboratory. The radon loss mechanism is of interest to the DOE radon program and part of it is now supported by that program.

We have also used the radium isotopes (^{226}Ra , ^{228}Ra , ^{224}Ra) in the hydrothermal waters of Yellowstone Park to determine, with our insights into adsorption and desorption coefficient determinations in cool groundwaters, the life

time of reaction of water with rock at depth prior to expansion and supply to the surface as geysers and hot springs. This work is reported in Clark and Turekian (Attachment) and shows that the reaction time of deep water in the Yellowstone plumbing system is about 700 years. This is significant, obviously, to a number of energy and environmental issues including geothermal energy, radioactive waste disposal and the behavior of metals in deep hydrothermal brines associated with oil fields.

C. The controls on the distribution of radionuclides and stable nuclides in the marine realm: Our interest in the oceans has always had a strong component of concerns about the margins (both continental and ocean bottom) and the processes acting there. We have been involved in the SEEP project and been opportunistic in studying the boundaries of the oceans wherever we can.

The problem of the fate of ^{210}Pb in the oceans has occupied us for many years, as it has others as well. Starting with the GEOSECS surveys and our work on Long Island Sound the work has been extended to virtually all the margins of the oceans. This includes the ridge areas of the oceans, the upwelling areas of the Pacific and Atlantic and marginal shelf environments of the northeastern U.S. Recently we published, with our WHOI collaborators, the results of the ^{210}Pb survey of ocean profiles in the SEEP area. The results (Bacon et al, Attachment) indicate that the western margin of the Atlantic Ocean is not an especially strong site for the removal of ^{210}Pb from the deep oceans.

We studied the marginal waters (deep) around the Atlantic sector of Antarctica, using samples collected for us on the AJAX expedition, for ^{210}Pb and found that, despite high productivity in the region, strong ^{210}Pb removal from the water column is not observed (Attachment: Farley and Turekian). Clearly the ^{210}Pb removal, and by extension other particle reactive element removal, must be found at the highly productive eastern edges of the ocean basins and the major oceanic ridge systems.

Our work on the ridge systems indicated the potential importance of these regions because of the deposition of iron sulfides and, more importantly, manganese and iron oxides. During November of 1988 the Hydros expedition, under the direction of Harmon Craig of Scripps Institution of Oceanography, provided an opportunity for close sampling of water above the East Pacific Rise at several locations in conjunction with the sampling being done for ^3He . Two former Yale undergraduates who did their Senior Theses on DOE supported projects (Ken Farley and Jordan Clark), took the samples in the proper manner on that expedition for the determination of ^{210}Pb and sent them to Yale. The results of this work showed a strong flux of ^{210}Pb into the deposits of the ridge axis. This confirms the importance of the ridge as a sink for ^{210}Pb and may explain the large-scale Pacific mid-ocean deficiency in ^{210}Pb (Attachment: Turekian and Rye).

The transport and accumulation of particulate organic carbon on the continental margin of the Middle Atlantic Bight was a major goal of the SEEP multi-institution enterprise. Our contribution to this effort was to measure ^{210}Pb , ^{137}Cs in bulk sediment and $\Delta^{14}\text{C}$ in other calcareous and organic fractions and in one case the separated lignin fraction. It became clear that ^{210}Pb and ^{137}Cs were indices of sediment mixing rather than accumulation and we found no evidence of significant accumulation of new organic carbon anywhere (although of course there must be some). There was no spike of bomb-produced ^{14}C in the most recent sediments so no recent massive transport of organic carbon to the continental slope from the shelf had occurred.

In most of the shelf and slope samples $\Delta^{14}\text{C}$ for coexisting organic carbon and carbonate carbon were indistinguishable. The deepest slope core (~2500 m) however, showed a marked disparity with the organic carbon generally having lower $\Delta^{14}\text{C}$ than coexisting carbonate carbon. The lignin fraction had the same $\Delta^{14}\text{C}$ as the bulk organic carbon indicating that all of the carbon in this deepest slope core could have been terrestrial in origin.

The $\delta^{13}\text{C}$ was about -19 to -20 per mil in all the organic carbon indicating either a marine origin or terrestrial carbon with a significant component of C-4 plant derived carbon. The latter would be possible if salt marsh carbon were exported as well as soils from upland C-3 plant-rich sites.

The sources of water along the Middle Atlantic Bight are known to be the Labrador Sea, the Sargasso Sea and deep upwelling water. We have been trying to determine the sources as a function of time and space by measuring the $\Delta^{14}\text{C}$ and $\delta^{18}\text{O}$ of water collected on the shelf from piers and several traverses from the shelf to the edge of the continental margin by ship. This is part of our effort in the SEEP program.

The $\delta^{18}\text{O}$ studies essentially confirm the results of previous workers with the identification of the freshwater component, the Labrador Sea component and, at depth, the Antarctic Intermediate Water component. The mixing of the different sources occurs but the capacity to diagnose all endmembers is limited by the range of $\delta^{18}\text{O}$ values and the compounding of other tracers such as salinity and temperature as the result of freshwater mixing and surface warming (Attachment: Kashgarian and Rye).

The $\Delta^{14}\text{C}$ tracer is more diagnostic than $\delta^{18}\text{O}$ in that at the present time the Labrador Sea water via the Gulf of Maine has a $\Delta^{14}\text{C}$ value of +60; the Gulf Stream (and Sargasso Sea) a value of +165 and Antarctic Intermediate Water -80. The mean $\Delta^{14}\text{C}$ value of Middle Atlantic Bight shelf water is about +100 which can be explained as a mix of Gulf of Maine water and Gulf Stream water. However, the range of $\Delta^{14}\text{C}$ values over a two year sampling period along the entire Middle Atlantic Bight is from 0 to +165 indicating the varying degrees of contribution of all three water types (Attachment: Kashgarian and Tanaka, Monaghan, Turekian).

The extension of these results backward in time is possible through the use of dated museum specimens of clams taken from the Middle Atlantic Bight and the Gulf of Maine and the use of the $\Delta^{14}\text{C}$ record in a 200 year old coral from Bermuda.

Our results show that there have been times in the past 150 years where upwelling of Antarctic Intermediate Water has been strong enough to impact the waters of Long Island Sound and the shells growing there (Attachment: Tanaka, Monaghan and Turekian). We conclude that $\Delta^{14}\text{C}$ provides an independent insight into the history of water-type mixing and upwelling from the present back to the earlier history of our nation for the Middle Atlantic Bight.

A similar study on water from the South Atlantic Bight, collected along piers, shows the role of upwelling specifically off South Carolina, presumably due to the influence of the Charleston Bump (Attachment: Tanaka, Kashgarian and Turekian).

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