the ground-water migration of this fission product (*9*). In addition, Mono Lake is well known for its elaborate $CaCO_3$ structures (*10*) formed as stream and spring water tributaries deliver dissolved calcium to the lake (*11*) which has extremely high concentrations of $CO_3{}^{2-}$ (~ 0.3*M*). The ratio of fallout $^{137}Cs$ to $^{90}Sr$ reported here for Mono Lake is the highest for any natural water of which we are aware.

The total quantities of fallout isotopes in Mono Lake are similar to those that would be expected from regional fallout data. The most unusual aspects of the fallout nuclide distributions in Mono Lake are that fallout plutonium has remained in the water column and $^{90}Sr$ has largely left the water column. The behavior of radioisotopes (including both anthropogenic and natural nuclides) in natural waters can be strongly dependent upon the major-element chemistry of the water, and information of this type must be included explicitly in any projections of the behavior of radioactive material in natural water systems.

H. J. SIMPSON, R. M. TRIER
J. R. TOGGWEILER, G. MATHIEU
B. L. DECK, C. R. OLSEN*
*Lamont-Doherty Geological Observatory* and *Department of Geological Sciences, Columbia University, Palisades, New York 10964*
D. E. HAMMOND, C. FULLER
T. L. KU
*Department of Geological Sciences, University of Southern California, Los Angeles 90007*

### References and Notes

1. H. J. Simpson, R. M. Trier, C. R. Olsen, D. E. Hammond, A. Ege, L. Miller, J. M. Melack, *Science* **207**, 1071 (1980).
2. J. M. Cleveland, in *Chemical Modeling in Aqueous Systems: Speciation, Sorption, Solubility, and Kinetics*, E. A. Jenne, Ed. (Symposium Series 93, American Chemical Society, Washington, D.C., 1979), pp. 321–338.
3. Measurements of $^{238}U$, $^{234}U$, $^{232}Th$, $^{230}Th$, and $^{228}Th$ were made on small samples (< 1 liter) filtered through glass fiber filters, returned to the laboratory, and analyzed by chemical preparation and α-spectrometry procedures frequently used for environmental samples. For $^{234}Th$, we used β-counting after chemical preparation with a 2-liter sample of filtered lake water. We concentrated $^{239,240}Pu$, $^{238}Pu$, $^{210}Pb$, and $^{210}Po$ from large samples in the field (80 to 240 liters) by precipitation with $Fe(OH)_3$ and analyzed them by α-spectrometry. We measured $^{228}Ra$ by γ-counting of its daughter, $^{228}Ac$, using $CaCO_3$ precipitated from a large field sample (80 liters). We measured $^{226}Ra$ by scintillation counting of its gaseous daughter product, $^{222}Rn$. We measured $^{137}Cs$ by γ-counting of an exchange resin frequently used for the extraction of fallout $^{137}Cs$ from environmental samples (ammonium molybdophosphate). We measured $^{90}Sr$ by β-counting, using chemical preparation procedures developed for the analysis of coral samples. We measured $^{3}H$ by liquid scintillation counting after enrichment by electrolysis. The $^{231}Pa$ was coprecipitated with $Fe(OH)_3$ from a 20-liter sample, purified, and measured by α-counting [T. L. Ku, *J. Geophys. Res.* **73**, 2271 (1968)].
4. D. Thurber, *Bull. Volcanol.* **28**, 1 (1965); R. F. Anderson, thesis, Massachusetts Institute of Technology–Woods Hole Oceanographic Institution (1980).
5. G. H. Coleman, *The Radiochemistry of Plutonium* (Publication NAS-NS-3058, National Academy of Sciences–National Research Council, Washington, D.C., 1965); R. E. Connick, in *The Actinide Elements*, G. T. Seaborg and J. J. Katz, Eds. (McGraw-Hill, New York, 1954), pp. 221–300; R. P. Larsen and R. D. Oldham, *Science* **201**, 1008 (1978); D. M. Nelson and M. B. Lovett, *Nature (London)* **276**, 599 (1978); E. A. Bondietti and J. R. Trabalka, *Radiochem. Radioanal. Lett.* **42**, 169 (1980).
6. The total delivery of fallout $^{137}Cs$ to collection stations at San Francisco and Medford, Ore., through 1970 was 30 to 40 $mCi/km^2$ (average precipitation rate at both stations, ~ 50 cm/year), assuming a $^{137}Cs/^{90}Sr$ ratio of 1.5 [*Report HASL-308* (Atomic Energy Commission, Washington, D.C., 1976), appendix].
7. M. A. Wahlgren and D. M. Nelson, *Verh. Int. Verein. Theoret. Angew. Limnol.* **19**, 317 (1975).
8. W. S. Broecker, E. R. Bonebakker, G. G. Rocco, *J. Geophys. Res.* **71**, 1999 (1966).
9. K. E. Cowser and T. Tamura, *Health Phys.* **9**, 687 (1963); J. A. Varga and D. G. Jacobs, *Report ORNL TM-3130* (Oak Ridge National Laboratory, Oak Ridge, Tenn., 1970).
10. P. Cloud and K. R. Lajoie, *Science* **210**, 1009 (1980).
11. D. T. Mason, *Limnology of Mono Lake, California* (Publications in Zoology vol. 83, Univ. of California Press, Berkeley, 1967); H. J. Simpson, thesis, Columbia University (1970).
12. T. L. Ku, K. G. Knauss, G. G. Mathieu, *Deep-Sea Res.* **24**, 1005 (1977).
13. W. S. Moore and W. M. Sackett, *J. Geophys. Res.* **69**, 5401 (1964).
14. S. G. Bhat *et al.*, *Earth Planet. Sci. Lett.* **5**, 483 (1969).
15. A. Kaufman, *Geochim. Cosmochim. Acta* **33**, 717 (1969); W. S. Moore, *Earth Planet. Sci. Lett.* **53**, 419 (1981).
16. K. G. Knauss, T. L. Ku, W. S. Moore, *Earth Planet. Sci. Lett.* **39**, 235 (1978).
17. A. Kaufman, R. M. Trier, W. S. Broecker, H. W. Feely, *J. Geophys. Res.* **78**, 8827 (1973).
18. K. W. Bruland, M. Koide, E. D. Goldberg, *ibid.* **79**, 3083 (1974).
19. M. P. Bacon, D. W. Spencer, P. G. Brewer, *Earth Planet. Sci. Lett.* **32**, 277 (1976); Y. Nozaki, J. Thomson, K. K. Turekian, *ibid.*, p. 304; Y. Nozaki and S. Tsunogai, *ibid.*, p. 313; J. Thomson and K. K. Turekian, *ibid.*, p. 297.
20. H. Livingston, D. R. Mann, V. T. Bowen, in *Analytical Methods in Oceanography*, T. Gibb, Jr., Ed. (American Chemical Society, Washington, D.C., 1975), p. 124.
21. The ratio of $^{137}Cs$ to $^{90}Sr$ in surface waters of the deep North Atlantic is assumed to be identical to the fallout ratio of 1.5.
22. V. T. Bowen, V. E. Noshkin, H. L. Volchok, T. T. Sugihara, *Science* **164**, 825 (1969).
23. W. Roether, thesis, University of Heidelberg (1967).
24. We thank S. Emerson of the University of Washington and D. Imboden of the Swiss Federal Institutes of Technology for their help during our 1980 field sampling and especially for providing dissolved oxygen and sulfide profiles of the highest quality. Financial support was provided by Department of Energy contract EY2529. This is Contribution No. 3313 from Lamont-Doherty Geological Observatory of Columbia University.

* Present address: Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, Tenn. 37830.

16 November 1981; revised 29 December 1981

## Elevated Concentrations of Actinides in Mono Lake

Abstract. *Tetravalent thorium, pentavalent protactinium, hexavalent uranium, and plutonium (oxidation state uncertain) are present in much higher concentrations in Mono Lake, a saline, alkaline lake in eastern central California, than in seawater. Low ratios of actinium to protactinium and of americium to plutonium indicate that the concentrations of trivalent actinides are not similarly enhanced. The elevated concentrations of the ordinarily very insoluble actinides are maintained in solution by natural ligands, which inhibit their chemical removal from the water column, rather than by an unusually large rate of supply.*

Many elements are chemically removed from natural waters by adsorption on particle surfaces rather than by precipitation of inorganic salts (*1*). The effective solubility of reactive elements can be regarded as the result of a competition between complexation by dissolved ligands and uptake at adsorption sites on settling particles (*2*). The concentration and mobility of reactive elements in a given body of water will therefore depend on chemical composition. We demonstrate in this report that certain actinide elements that are extremely insoluble under most environmental conditions have greatly elevated concentrations in Mono Lake, a highly alkaline (*p*H ~ 10) lake in eastern central California with a salt content about twice that of seawater (*3*).

Simpson *et al.* (*4*) found unexpectedly high concentrations of plutonium in Mono Lake. The chemical removal of dissolved plutonium by adsorption to particles or bottom sediments is ordinarily very rapid, and in freshwater lakes nearly all the plutonium delivered by atmospheric fallout is found in the sediments. In Mono Lake, on the other hand, about half the plutonium inventory has remained in the water, and plutonium concentrations are about 100 times greater there than in other lakes and estuaries and in coastal seawater. Simpson *et al.* questioned whether the elevated concentrations result from the complexation of plutonium by the high concentration of $CO_3{}^{2-}$ or whether an unusually large proportion of the total plutonium is somehow maintained in the relatively more soluble VI (or V) oxidation state rather than in the chemically reactive and less soluble III and IV oxidation states (*5, 6*). The oxidation state of plutonium in Mono Lake has yet to be measured.

Actinide elements in the same oxidation state tend to have similar chemical properties (*7*). We have measured the concentrations of four natural actinides, actinium, thorium, protactinium, and uranium, and two other actinides derived from nuclear fallout, plutonium and americium, in Mono Lake water. The oxi-

0036-8075/82/0430-0514$01.00/0

dation states would be as follows: actinium, III; thorium, IV; protactinium, V; and uranium, VI. Our objective was to determine whether actinides in all oxidation states are present at elevated concentrations in Mono Lake.

A surface-water sample (~ 11 liters) was obtained from the western shore of the lake and shipped in a plastic Cubitainer to Woods Hole Oceanographic Institution without any pretreatment. A chlorinity analysis showed that our sample contained lake water diluted with fresh spring water (*8*). Visible amounts of particulate material were present. No attempt was made to remove the particles for fear of losing some of the dissolved actinides. The concentrations of the actinides were measured by isotope dilution and α-spectrometry (*9*).

Table 1. Actinide activities in Mono Lake water and seawater; N.D., not determined.

| Isotope | Valence | Mono Lake* (dpm/$10^3$ liters) | Seawater (dpm/$10^3$ liters) |
|---|---|---|---|
| $^{232}$Th | IV | 886 ± 29 | 0.002 to 0.03 (*9*) |
| $^{232}$Th | IV | | < 0.016† (*22*) |
| $^{230}$Th | IV | 1674 ± 43 | 0.4 to 1.0 (*9*) |
| $^{230}$Th | IV | | 0.3 to 1.3 (*2*) |
| $^{230}$Th | IV | | 0.5 to 1.6 (*12*) |
| $^{228}$Th | IV | 966 ± 36 | 0.2 to 2.0 (*9*) |
| $^{228}$Th | IV | | 0.05 to 0.8† (*14*) |
| $^{228}$Th | IV | | 1 to 7‡ (*15*) |
| $^{238}$U | VI | $(2.40 \pm 0.05) \times 10^5$ | $2.5 \times 10^3$ (*23*) |
| $^{231}$Pa | V | 79.4 ± 2.9 | 0.1 to 0.3 (*9*) |
| $^{231}$Pa | V | | 0.13 to 0.24 (*11*) |
| $^{239,240}$Pu | | 24.8 ± 2.4 | 1 to 5 (*24*) |
| $^{239,240}$Pu | | ~ 44 (*4*, *8*) | |
| $^{227}$Ac | III | < 6 | N.D. |
| $^{241}$Am | III | 2.7 ± 0.6 | 0.3 to 0.7 (*18*) |

*This work. Errors are ± 1σ counting statistics. These values are for Mono Lake water diluted with fresh spring water (*8*). †Open ocean surface water. ‡Nearshore surface ocean water.

Table 2. Radioisotope activity ratios in Mono Lake water and seawater; N.D., not determined.

| Ratio | Mono Lake | Seawater |
|---|---|---|
| $^{234}$U/$^{238}$U | 1.14 ± 0.01 | 1.14 ± 0.03 (*23*) |
| $^{228}$Th/$^{232}$Th | 1.19 ± 0.05 | 50 to 100+ (*9*) |
| $^{230}$Th/$^{231}$Pa | 21.1 ± 1.0 | 3 to 5 (*9*) |
| $^{230}$Th/$^{231}$Pa | | 1.4; 3.4 (*11*) |
| $^{227}$Ac/$^{231}$Pa | < 0.08 | N.D. |
| $^{241}$Am/$^{239,240}$Pu | 0.11 ± 0.03 | 0.2 to 0.3 (*18*) |

The actinide activities in the lake water are compared with the seawater actinide activities in Table 1. The activities of the thorium isotopes and $^{231}$Pa show the greatest enhancement, with values $10^3$ to $10^5$ times greater in Mono Lake than in the deep ocean. Steady-state concentrations of chemically reactive actinides such as $^{230}$Th and $^{231}$Pa can be represented as a balance between their rates of supply and removal:

$$dA/dt = S - (\lambda + \psi)A = 0 \qquad (1)$$

where $A$ is the concentration of the actinide, $S$ is its rate of supply, λ is its radioactive decay constant, and ψ is the rate constant for chemical removal. The residence time with respect to chemical removal, τ, is defined as $1/\psi$. Since $A = S/(\lambda + \psi)$, high values of $A$ in Mono Lake compared to other bodies of water could be accounted for either by a higher rate of supply or by a lower rate of chemical removal. Therefore, residence time is a better measure than concentration by which to compare the mobilities of actinides in different bodies of water.

The isotopes $^{230}$Th and $^{231}$Pa are produced in Mono Lake by the radioactive decay of $^{234}$U and $^{235}$U, respectively. In view of the salt budget for the lake (*10*), the supply of $^{230}$Th and $^{231}$Pa by stream runoff should be insignificant, and the quantity of uranium in the lake may be considered constant with time. Therefore, in Eq. 1 we can represent $S$ as $\lambda A_{\mathrm{U}}$, where $A_{\mathrm{U}}$ is the concentration of the appropriate uranium isotope parent. The calculated residence times of $^{230}$Th (668 ± 23 years) and $^{231}$Pa (343 ± 15 years) are much shorter than their radioactive half-lives, and so the loss of both isotopes from Mono Lake water must occur almost entirely by chemical removal rather than by radioactive decay. These residence times are several times greater than in the deep ocean [$\tau_{\mathrm{Th}} = 15$ to 40 years (*8*, *11–13*); $\tau_{\mathrm{Pa}} = 30$ to 130 years (*8*, *11*, *13*)]. However, it is more appropriate to compare $\tau_{\mathrm{Th}}$ and $\tau_{\mathrm{Pa}}$ in Mono Lake with values from surface seawater and estuarine environments where the concentrations of particulate matter are more like those in Mono Lake. The value of $\tau_{\mathrm{Pa}}$ has not been determined accurately in surface seawater, but $\tau_{\mathrm{Th}}$ is less than 1 year in open-ocean surface seawater (*14*, *15*) and is only a few days in coastal and estuarine waters (*16*). The elevated concentrations of $^{230}$Th and $^{231}$Pa in Mono Lake must then reflect a pronounced inhibition of chemical removal as compared to the situation in seawater and not merely elevated rates of supply. The high concentration of $^{232}$Th in Mono Lake (Table 1), which must be derived entirely by leaching from detrital minerals, further attests to the solubilizing effect of Mono Lake water.

Mono Lake is currently drying up as a result of the diversion of inflow to Los Angeles, and so the measured concentrations may not represent steady-state values. However, $\tau_{\mathrm{Th}}$ and $\tau_{\mathrm{Pa}}$ are calculated from the $^{230}$Th/$^{234}$U and $^{231}$Pa/$^{235}$U ratios, respectively. As these residence times are much greater than the period over which the concentrations may have been changing, we believe that the ratios should have changed little and that these residence times give good first-order estimates of the chemical removal rates in Mono Lake.

All actinides are not solubilized to the same extent in Mono Lake. For example, $\tau_{\mathrm{Th}}$ is greater by about a factor of 2 than $\tau_{\mathrm{Pa}}$, whereas $\tau_{\mathrm{Pa}}$ is greater by a factor of 2 or more than $\tau_{\mathrm{Th}}$ in the deep ocean (*9*). Therefore, although chemical removal of both thorium and protactinium is greatly reduced in Mono Lake as compared to seawater, the solubilizing effect is significantly greater for thorium than for protactinium. If the residence times of trivalent actinides in Mono Lake water were as great as those of thorium and protactinium, $^{227}$Ac, with a half-life of 21.8 years, would be in radioactive equilibrium with its parent, $^{231}$Pa (that is, $^{227}$Ac/$^{231}$Pa = 1.0). Only an upper limit for the concentration of $^{227}$Ac could be set for Mono Lake water, but the actinium/protactinium activity ratio is clearly much less than 1.0 (Table 2). Similarly, the $^{241}$Am/$^{239,240}$Pu activity ratio (Table 2) is less than half the ratio observed in seawater and in atmospheric fallout (*17*, *18*). Therefore, Ac(III) and Am(III) are more efficiently removed from Mono Lake than thorium, protactinium, and plutonium.

Simpson *et al.* (*4*) suggested that complexation by $CO_3^{2-}$ may be repsonsible for the high concentrations of plutonium in Mono Lake, and others (*5*, *19*) have given evidence for the increased solubility of actinides in oxidation states IV, V, and VI as a result of $CO_3^{2-}$ complexation. We have attempted to quantify the effect of $CO_3^{2-}$ complexing through the use of computer models but have been frustrated by the lack of suitable thermodynamic data on actinide complexes. Simulations of the solution equilibrium chemistry by means of the computer program MINEQL (*20*), incorporating removal processes similar to those in the ocean (*2*), were made with and without the effect of $CO_3^{2-}$ complexing. Let us consider the case of thorium, for which the solubilizing effect is largest (Table 1). In the absence of $CO_3^{2-}$ complexing, the dominant thorium species is $Th(OH)_4$ (*21*), which would lead to rapid removal by adsorption. The simplest carbonate species we could invoke which would compete effectively with $Th(OH)_4$ is $Th(CO_3)(OH)_2$. If we assign $\log K = 6$ for the formation constant of this species, then 98 percent of the dissolved thorium would be in this form. Our educated guess is that this is a lower limit and that $\log K > 6$ is likely.

We believe, therefore, that the elevated concentrations of actinides in Mono Lake could be maintained largely by complexation with $CO_3^{2-}$. Other ligands may also be important. The effective solubilities of actinides in oxidation states IV, V, and possibly VI in this lake are greatly enhanced relative to seawater by complexation with natural ligands at the concentrations found in Mono Lake. The solubilities of trivalent actinides are not as strongly enhanced. The marked solubilizing effect of natural alkaline brines on the actinide elements has important implications for the management of radioactive waste and its transport in ground water. Our ability to model this effect would be greatly enhanced by the availability of thermodynamic data on actinide carbonate species.

R. F. Anderson*
M. P. Bacon
P. G. Brewer
*Department of Chemistry,*
*Woods Hole Oceanographic Institution,*
*Woods Hole, Massachusetts 02543*

### References and Notes

1. V. M. Goldschmidt, *Geochemistry* (Clarendon, Oxford, 1954); K. B. Krauskopf, *Geochim. Cosmochim. Acta* **9**, 1 (1956); K. K. Turekian, *ibid.* **41**, 1139 (1977).
2. P. W. Schindler, *Thalassia Jugosl.* **11**, 101 (1975); L. Balistrieri, P. G. Brewer, J. W. Murray, *Deep-Sea Res.* **28A**, 101 (1981).
3. H. C. Whitehead and J. H. Feth, *Geol. Soc. Am. Bull.* **72**, 1421 (1961); H. J. Simpson and T. Takahashi, *Init. Rep. Deep Sea Drill. Proj.* **20**, 877 (1973).
4. H. J. Simpson, R. M. Trier, C. R. Olson, D. E. Hammond, A. Ege, L. Miller, J. M. Melack, *Science* **207**, 1071 (1980).
5. J. B. Andelmann and T. C. Rozzell, in *Radionuclides in the Environment*, R. F. Gould, Ed. (American Chemical Society, Washington, D.C., 1970), pp. 118–137.
6. C. N. Murray and R. Fukai, in *Impacts of Nuclear Releases into the Environment* (International Atomic Energy Agency, Vienna, 1975), pp. 179–192; D. M. Nelson and M. B. Lovett, *Nature (London)* **276**, 599 (1978); J. M. Cleveland, in *Chemical Modeling in Aqueous Systems: Speciation, Sorption, Solubility, and Kinetics*, E. A. Jenne, Ed. (Symposium Series 93, American Chemical Society, Washington, D.C., 1979), pp. 321–338; E. A. Bondietti and J. R. Trabalka, *Radiochem. Radioanal. Lett.* **42**, 169 (1980).
7. F. P. Cotton and G. Wilkinson, *Advanced Inorganic Chemistry* (Interscience, New York, ed. 3, 1972).
8. The concentrations of actinides in Table 1 are lower than those reported by H. J. Simpson *et al.*, *Science* **216**, 512 (1982). Our sample, taken near the edge of Mono Lake, was apparently diluted with fresh water from a nearby spring. The sample had a chlorinity of 13.2 per mil as compared with an average chlorinity of ~ 20 per mil for the samples taken by Simpson *et al.* from the center of the lake. The concentrations of actinides in Table 1 normalized to a chlorinity of 20 per mil are in good agreement with the concentrations determined by Simpson *et al.* The major conclusions of this report are not changed by the fact that our sample was diluted with fresh water. Furthermore, the values of $\tau_{Th}$ and $\tau_{Pa}$ are unaffected by this dilution since they are calculated from the ${}^{230}Th/{}^{234}U$ and ${}^{231}Pa/{}^{235}U$ ratios, respectively. We learned of the full extent of the work of Simpson *et al.* on the isotopes of the uranium and thorium series in Mono Lake after the initial draft of this report had been prepared. This report and the accompanying one by Simpson *et al.* were written without reference to each other in order to identify the original contributions of each group.
9. R. F. Anderson, thesis, Woods Hole Oceanographic Institution–Massachusetts Institute of Technology (1981).
10. D. W. Scholl, R. Von Huene, P. St. Amand, J. B. Ridlon, *Geol. Soc. Am. Bull.* **78**, 583 (1967).
11. W. S. Moore and W. M. Sackett, *J. Geophys. Res.* **69**, 5401 (1964).
12. B. L. K. Somayajulu and E. D. Goldberg, *Earth Planet. Sci. Lett.* **1**, 102 (1966).
13. P. G. Brewer, Y. Nozaki, D. W. Spencer, A. P. Fleer, *J. Mar. Res.* **38**, 703 (1980).
14. W. S. Broecker, A. Kaufman, R. M. Trier, *Earth Planet. Sci. Lett.* **20**, 35 (1973); E. Matsumoto, *Geochim. Cosmochim. Acta* **39**, 205 (1975); Y. H. Li, H. W. Feely, J. R. Toggweiler, *Deep-Sea Res.* **27A**, 545 (1980).
15. K. G. Knauss, T. L. Ku, W. S. Moore, *Earth Planet. Sci. Lett.* **39**, 235 (1978).
16. R. Aller and J. K. Cochran, *ibid.* **29**, 37 (1976); Y. H. Li, H. W. Feely, P. H. Santschi, *ibid.* **42**, 13 (1979); P. H. Santschi, Y. H. Li, J. Bell, *ibid.* **45**, 201 (1979); M. Minagawa and S. Tsunogai, *ibid.* **47**, 51 (1980).
17. H. D. Livingston, D. L. Schneider, V. T. Bowen, *ibid.* **25**, 361 (1975).
18. H. D. Livingston and V. T. Bowen, in *Environmental Toxicity of Aquatic Radionuclides: Models and Mechanisms* (Ann Arbor Science, Ann Arbor, Mich., 1976), p. 107.
19. M. H. Kurbatov, H. B. Webster, J. O. Kurbatov, *J. Phys. Colloid Chem.* **54**, 1239 (1950); M. R. Scott, unpublished results; I. E. Starik and L. B. Kolyadnin, *Geochemistry* **2**, 245 (1957); D. Langmuir, *Geochim. Cosmochim. Acta* **42**, 547 (1978).
20. J. C. Westall, J. L. Zachary, F. M. M. Morel, *Technical Note 18* (Water Quality Laboratory, Department of Civil Engineering, Massachusetts Institute of Technology, Cambridge, 1976); in our calculations we assumed a $CO_3^{2-}$ concentration of 0.187*M* and a *p*H of 10 (*3*).
21. C. F. Baes, Jr., and R. E. Mesmer, *The Hydrolysis of Cations* (Wiley, New York, 1976).
22. A. Kaufman, *Geochim. Cosmochim. Acta* **33**, 717 (1969).
23. T. L. Ku, K. G. Knauss, G. G. Mathieu, *Deep-Sea Res.* **24**, 1005 (1977).
24. V. T. Bowen, V. E. Noshkin, H. D. Livingston, H. L. Volchok, *Earth Planet. Sci. Lett.* **49**, 411 (1980).

25. Discussions with H. D. Livingston during the early stages of this work led to the inclusion of plutonium and americium measurements in this study. We thank L. Surprenant for performing the ${}^{241}Am$ analysis. H. J. Simpson arranged for the comparison of the chlorinity of our sample with that of the samples analyzed by Simpson *et al.* (*8*). S. Smith assisted with the use of the MINEQL program calculations. Financial support was provided by NSF grant OCE 78-26318, Department of Energy contract EY-76-S-02-3566, and a fellowship to R.F.A., from the Woods Hole Oceanographic Institution Education Office. This is Woods Hole Oceanographic Institution Contribution 4962.

[*] Present address: Lamont-Doherty Geological Observatory, Palisades, N.Y. 10964.

14 September 1981; revised 29 December 1981

---

# Deep Advective Transport of Lithogenic Particles in Panama Basin

Abstract. *Sediment traps were deployed at several depths between 660 and 3800 meters in the Panama Basin. The flux of lithogenic particles increased with increasing depth. This increase was due primarily to particles of beidellite (a smectite clay), which was identical to the clay occurring in bottom sediment on the continental slope to the west of the sediment trap mooring. The beidellite vertical flux at the Panama Basin station increased when an easterly current prevailed and decreased when the current reversed, indicating that a major portion of smectite was transported horizontally at mid-water depth to the mooring site from the nearby continental slope.*

Six Parflux Mark II sediment traps (*1*) with 1.5-$m^2$ openings were deployed at depths of 667 to 3791 m at 5°21′N, 81°53′W in the Panama Basin. This station, which is 3856 m deep, is located at the center of a small, deep basin between the Coiba and Malpelo ridges (Fig. 1) (*2*). Mass flux, measured by this set of sediment traps, increased significantly from 1268 to 3791 m. The increase was due to the increasing flux of lithogenic material, particularly smectite particles (Table 1).

A Parflux trap deployed at 2265 m was equipped with a receptacle changer, which changes the sediment receiving chamber every 30 days, yielding a time series of sediment flux. A cylindrical trap with an opening of 0.05 $m^2$ was deployed at 1267 m. This trap also had a receptacle changer and was set to collect sediment every 15 days (JZF trap) (*3*). To start a new collecting period in these two traps the timing was synchronized by electronic timers which operated within an error of a few hours (*4*).