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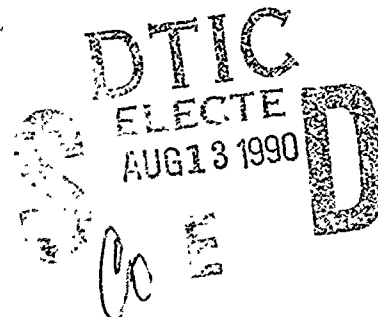
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Radium Isotopes in the Orinoco Estuary and Eastern Caribbean Sea

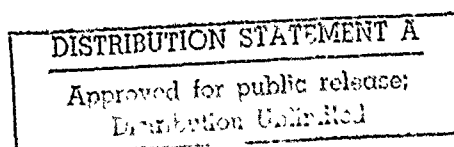
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Abstract

Radium isotopes provide a means of identifying the source of freshened waters in the ocean and determining the time elapsed since these waters were in the estuary. We present evidence that, during April, waters from the Amazon mixing zone pass within 50 km of the mouth of the Orinoco mouth. These Amazon waters are characterized by a lower $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio than are waters from the Orinoco at similar salinities. Within the Caribbean Sea waters of Orinoco origin carry a characteristic radium signature including excess activities of ^{224}Ra . This isotope may be used to estimate the time elapsed since the waters were removed from contact with sediments. Current speeds based on ^{224}Ra dating ranging from 5 to 20 cm/sec during April.

The radium isotopes also provide an assessment of sediment mixing in the estuary. During low discharge (April), considerable mixing of older sediment by physical or biological processes or dredging maintained high activities of ^{228}Ra in the estuary and produced the highest $^{228}\text{Ra}/^{226}\text{Ra}$ A. R.'s yet measured in any estuary. During high discharge (September) a large fraction of the ^{228}Ra was derived from desorption from fresh sediment rather than mixing of older sediments. Activities of ^{224}Ra were high in the estuary during both high and low discharge, indicating that considerable mixing of recently introduced sediment must occur during each period. The high $^{224}\text{Ra}/^{228}\text{Ra}$ A. R.'s measured in September imply that the same sediments were being resuspended during high discharge, and that there was little exposure of older sediments which had been stored in the estuary for months to years. During April ^{224}Ra and ^{228}Ra activities in the water were about equal, indicating that most of the sediment being resuspended had been stored in the estuary long enough to reestablish radioactive equilibrium in the ^{232}Th decay series, 20 years.



Radium Isotopes in the Orinoco Estuary and Eastern Caribbean Sea

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1. Introduction

Mixing zones at the mouths of major rivers supply pulses of freshened water to the ocean. Because of their lower density, these waters may move across the surface of the ocean undergoing little mixing with surrounding waters and retaining for long distances chemical properties acquired in the river or mixing zone.

Satellite imagery has proved to be an invaluable means of tracing these freshened plumes into the ocean (Muller-Karger et al. 1988). These images can provide data on the pattern of the dispersal of these waters and how these patterns change through the year. The primary signal recorded by the satellites is ocean color which, in the case of freshened plumes, is due mainly to chlorophyll from primary productivity, phaeopigments and dissolved organic matter (Muller-Karger et al. 1988). The interpretation of the satellite data assumes that the path of the water and the pattern of the stimulation of productivity are the same. Plumes which do not stimulate productivity and contain only a small component of dissolved organic matter from the river may not be recognized.

Directly measured chemical tracers also provide information on the dispersal of river water in the ocean. Although these techniques do not cover the spatial scale of the satellites, they do provide a means of quantifying the amount of water from the mixing zone and of determining chemical fluxes associated with the plumes. Salinity is an obvious chemical tracer of freshened plumes. It is said that from the presence of low salinity water in the open ocean, Columbus deduced that he was near a continent. Another tracer which has been used to identify the influence of rivers is dissolved silica, a component with high concentrations in river water. Froelich et al (1978) determined that at least 60% of the freshening of the surface Caribbean during Oct. - Nov. was due to river water rather than direct precipitation. Moore et al (1986) used radium isotopes to determine that even at distances of 1500 km from the mouth, up to 30% of the water sampled in the Atlantic Ocean had recently passed through the Amazon estuary.



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To determine the time scale of the transport of freshened plumes, oceanographers have relied on surface drifters, devices which are designed to float within a current system. These devices will provide information on the direction and speed of the current as long as they stay within the main current field. Because of the possibility that the drifter may exit the current field, multiple drifters are often deployed to track a current system.

In this paper we extend the use of radium tracers to the region of the Orinoco River plume in the Atlantic Ocean and Caribbean Sea. In addition to the commonly measured isotopes, ^{228}Ra ($t_{1/2} = 5.7$ yrs.) and ^{226}Ra ($t_{1/2} = 1600$ yrs.), we introduce the use of the short lived isotope, ^{224}Ra ($t_{1/2} = 3.6$ days) to establish the time scale of mixing and transport between freshened river plumes and the adjacent coastal ocean.

2. Radium isotopes as tracers

a. Generation of the signal

In river waters Ra strongly adsorbs to particles; in sea water it is primarily dissolved. These differences in chemical behavior are due to a change in the adsorption coefficient of Ra between fresh and salt water and to a change in the average particle concentration between terrestrial and ocean waters. From measurements in the Hudson River estuary, Li et al (1977) demonstrated that activities of ^{226}Ra in the estuary were higher than either river or ocean activities. They postulated that desorption of Ra bound to the surfaces of particles occurs as the particles enter the high ionic strength estuarine water. Experimental measurements of the release of Ra from river borne particles proved that this mechanism was important in supplying Ra to the estuary.

Elsinger and Moore (1980) found that activities of ^{226}Ra in Winyah Bay, S. C., were considerably higher during average and low flow of the Pee Dee River than they were during high river flow. The authors postulated that upstream advancement of the salt wedge during periods of lower discharge released ^{226}Ra from sediments initially deposited in fresh water.

Key et al (1985) established that the ^{228}Ra activity in the Amazon estuary far exceeded that which could be supplied by desorption from riverborne sediments. They separated the source functions for ^{226}Ra and ^{228}Ra into four components: (1) river dissolved (2) river suspended (3) ocean dissolved and (4) estuarine sediments. From measurements of the distribution of the Ra isotopes among the various components, they concluded that most of the excess ^{226}Ra was supplied by desorption from river borne sediments,

but most of the excess ^{228}Ra was released from estuarine sediments. This observation requires that the estuarine sediments maintain contact with the overlying water for some time after being deposited so that new ^{228}Ra may be generated from the decay of the long lived parent, ^{232}Th . As these recently deposited sediments mix with the overlying waters, they release Ra isotopes which have been generated by Th decay since the sediments were deposited. Because this mixing time scale is on the order of years, this mechanism provides an efficient source of freshly generated ^{228}Ra with little ^{226}Ra .

Generation of ^{224}Ra activity in the sediments occurs on a time scale of days. Frequent mixing of the upper few cm of estuarine sediments thus provides an efficient source of ^{224}Ra activity accompanied by little ^{228}Ra or ^{226}Ra activity. ^{224}Ra produced in the estuary may be used to identify estuarine water in the open ocean; and, if the estuarine endmember activity can be determined, changes in the ^{224}Ra activity of waters isolated from input of ^{224}Ra provides information on the time since the water was in the estuary. A fourth radium isotope, ^{223}Ra ($t_{1/2} = 11.4$ days), complicates the picture somewhat. When the measurements reported in this paper were being made, we could not easily discriminate between ^{224}Ra and ^{223}Ra . Because of the far greater abundance of ^{224}Ra parents, we assumed that none of the activity was due to ^{223}Ra . We have since learned that the measuring system is more sensitive to ^{223}Ra than expected (although the ^{224}Ra sensitivity is at least twice as great as ^{223}Ra) and that the initial ^{223}Ra activity may be on the order of 5% of the ^{224}Ra activity. This does not affect our qualitative observations concerning the distribution of ^{224}Ra in the estuary; but, it does mean that the ages we calculate for waters removed from the source region may be too young.

b. Mixing models

Moore et al (1986) measured extremely high $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios in samples collected in the Atlantic Ocean and Caribbean Sea at distances of up to 1500 km from the mouth of the Amazon River. They concluded that these high A. R.'s were generated in the Amazon mixing zone (or the Orinoco mixing zone) and calculated that as much as 30% of the water had passed through one of these systems. They pointed out that unlike other tracers of surface water which change by evaporation, precipitation and biological activity, the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio will only change by radioactive decay if the water is isolated from inputs of Ra. This ratio thus provides a sensitive tracer of the presence of estuarine water in the ocean. In order to calculate the fractions of estuary and ocean water as well as changes due to evaporation and precipitation for slightly freshened water parcels in the open ocean, Moore et al (1986) constructed a mixing

model based on the conservation of water, salt, ^{228}Ra and ^{226}Ra . The relevant equations are:

$$f_{oc} = \frac{S_{obs} - S_{es} f_{es}}{S_{oc}} \quad (1)$$

$$f_{es} = \frac{\frac{S_{obs}}{S_{oc}} [AR_{obs} {}^{226}\text{Ra}_{oc} - {}^{228}\text{Ra}_{oc}]}{AR_{obs} {}^{226}\text{Ra}_{es} - {}^{228}\text{Ra}_{es} - \frac{S_{es}}{S_{oc}}} \quad (2)$$

$$f_{E-P} = 1 - f_{oc} - f_{es} \quad (3)$$

where

- obs = observed value
- oc = ocean end member
- es = estuary end member
- E = evaporation
- P = precipitation
- f = fraction
- AR = activity ratio

To use the activity of ^{224}Ra in a water sample as an index of the time since the water left the estuary, we use the equation

$$t = - \frac{\ln\left[\frac{{}^{224}\text{Ra}_{obs}}{{}^{224}\text{Ra}_{es} f_{es}}\right]}{\lambda_{224}} \quad (4)$$

where λ_{224} is the decay constant of ^{224}Ra , 0.191 days⁻¹.

The basic assumptions in these models are that the endmembers do not change over the time period of interest and that no additions or subtractions of Ra isotopes except for mixing and radioactive decay occur. (The latter applies only to ^{224}Ra , since the time scale of mixing with which we are concerned is much shorter than the decay constant of ^{228}Ra). In equation 4, the effect of mixing is removed by the f_{es} term to give the time elapsed since the water left the estuary.

Of course the selection of the endmember is an important consideration of the modeling. For the Amazon system Moore et al (1986) used an estuarine endmember at 31.8 ‰ salinity, but noted that if an endmember of 21.5 ‰ had been selected, the computed estuarine components would have been reduced by about 30 %.

3. Measurement techniques

Surface water samples were pumped from the bow pump of the R/V COLUMBUS ISELIN into plastic barrels. The nominal depth of the intake was 2 m; however, ship motion and wave activity often caused considerable variability. When the ship was underway the intake was higher than it was on station. Thin freshened plumes observed on the salinity sensor for the SAIL system during steaming sometimes disappeared when the ship stopped for station. Therefore, salinity and silica samples were taken from the large volume homogenized samples before radium analyses. Changes in the depth of the intake and relative movement on station with respect to the initial position of freshened plumes sometimes made the correlation between the radium samples and samples collected for other measurements difficult.

Between 80 and 360 liters of water was processed for radium isotope measurements. If the water was clear and low in suspended solids, it was filtered directly through a column of MnO_2 -coated fibers (i.e. Mn-fiber) to quantitatively remove radium (Moore 1976). Samples which contained significant suspended solids were first passed through a 1 μm CUNO Micro Wynd (CUNO Inc., Meriden, CT) cartridge filter. In some cases highly turbid samples were allowed to settle 10 - 20 hours before filtering.

The Mn-fiber was drained and placed in a column for ^{224}Ra measurements as described by Rama et al (1987). Air was circulated through the column and decays of ^{220}Rn ($t_{1/2} = 54.5$ sec) and ^{216}Po ($t_{1/2} = 0.16$ sec) were measured in an alpha scintillation detector attached to the circulation system. The efficiency of the system was established by using standards prepared from a ^{232}Th solution. Backgrounds were checked frequently because of the buildup of ^{212}Pb ($t_{1/2} = 10.6$ hrs) in the detector.

Some samples were remeasured in the ^{224}Ra system 1 - 2 months after collection to determine the ^{228}Th activity on the fiber. The waiting time was necessary to allow initial excess ^{224}Ra to decay and a new generation of ^{224}Ra activity to equilibrate with ^{228}Th . Excess ^{224}Ra activity in the water was calculated by subtracting the ^{228}Th supported activity from that initially measured.

We recognize that in addition to ^{224}Ra , the samples may contain ^{223}Ra . Like ^{224}Ra , ^{223}Ra has a short-lived radon daughter (^{219}Rn , $t_{1/2} = 4$ secs) which is detected in the system we use to measure ^{224}Ra with an efficiency on the order of 50% of the ^{224}Ra detection efficiency. Because the abundances of the parent isotopes of ^{223}Ra are much lower than the parents of ^{224}Ra , initial $^{223}\text{Ra}/^{224}\text{Ra}$ activity ratios should be quite low, on the order of 0.05. However, the longer half life of ^{223}Ra causes this ratio to increase with time. Therefore, the ages which we calculate assuming all of the activity is due to ^{224}Ra are too young.

In the laboratory the ^{226}Ra activity on the Mn-fiber was measured by a technique described by Butts et al (1988). The Mn-fiber was placed in a sealed glass equilibration vessel and purged of all ^{222}Rn . After 5 - 20 days of accumulation of ^{222}Rn , a measured fraction of the air in the equilibrator was transferred to a Lucas cell and decays of ^{222}Rn and daughters were measured.

After the ^{226}Ra measurements the radium was removed from the Mn-fiber by leaching in a solution of hydroxylamine hydrochloride and HCl. Ra was coprecipitated with BaSO_4 and stored in a small vial to allow ^{222}Rn to equilibrate with ^{226}Ra . Then the $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. of the sample was measured in a well-shaped intrinsic germanium (WeGe) detector (Moore 1984). This A. R. multiplied by the ^{226}Ra activity in the water gave the ^{228}Ra activity in the water.

4. Results

A map of the study area is presented in the introductory paper in this volume. The results of these measurements are presented in Tables 1 and 2. The errors estimated for the Ra activities are about $\pm 10\%$. In addition to the radionuclide data, measurements of salinity and silica from the large volume samples are included in these tables. To correct the ^{224}Ra values for ^{228}Th -supported activity, ^{228}Th was measured on some samples by allowing the initial excess ^{224}Ra to come to equilibrium with ^{228}Th on the fiber. The ^{228}Th activities were very low, resulting in errors on the order of $\pm 20\%$. Excess ^{224}Ra was calculated by subtracting the ^{228}Th activity from the initial ^{224}Ra activity and assuming that ^{223}Ra activity was negligible. Samples not measured for ^{228}Th were corrected using interpolated values.

5. Distribution of Ra isotopes during April

a. Estuary samples

During low discharge conditions in April, the freshwater front of the Orinoco River was located upstream of the most inland station, therefore we did not obtain a sample of the freshwater endmember during low discharge. The lowest salinity sampled was about 7 ‰.

Low water levels in the River channel during the spring necessitate continual dredging to keep the channel open for ship traffic. The channel is maintained at 9.9 m during low water and 13.7 m during high water. The Pilot Guide cautions that

A feature of the Rio Orinoco is that the bottom consists of a layer of very soft mud, virtually in suspension and partially mobile, which is locally known as "calambrina". The existence of the calambrina and its mobility makes it difficult to calculate the actual depth of water in the channel. Mariners are warned that the calambrina may severely hinder the passage of vessels across the bar, particularly during the summer months. Vessels drawing more than 7.5 m are likely to have to force a passage through a layer of mud a meter more thick with a consequent loss of speed up to 50%.

Except for slight increases in the 25-30 ‰ salinity range, ^{226}Ra activities were rather constant through the estuary (Figure 1). This is probably due to the small quantity of fresh sediment being supplied to the estuary during low flow. Increases of ^{228}Ra did occur in the salinity range 17-33 ‰ (Figure 1) causing the $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. to reach a value of 6.74, the highest value ever measured in an estuary. Continued release of ^{228}Ra generated from ^{232}Th decay on sediments deposited in the estuary must cause a high flux of ^{228}Ra with little addition of ^{226}Ra . Similar mechanisms of ^{228}Ra enrichment in estuaries have been reported (Elsinger and Moore 1983; Elsinger and Moore 1984; Key et al. 1985; Moore 1981; Santschi et al. 1979); but, in no other reported system is the effect as great as in the Orinoco. Increases of ^{224}Ra generally paralleled increases of ^{228}Ra through the estuary (Figure 1).

b. Atlantic Samples

One of our goals in this study was to determine if Ra isotopes could unambiguously identify the source of freshened waters in the ocean. We recognized that even though the study was concentrated near the mouth of the Orinoco, much of the freshened water further offshore could have an Amazon River source.

The presence of high activities of excess ^{224}Ra in samples near the Orinoco mouth was convincing evidence that the Orinoco was the source of these freshened waters. However, the absence of excess ^{224}Ra in a freshened plume did not unequivocally eliminate the Orinoco as a source. The freshened plume could have left the estuary two weeks earlier and become entrained in an eddy or simply moved slowly away from the estuary.

Samples from stations R1/S and R2/S (# 13 & 14) are a case in point. These samples are slightly fresher than samples collected nearer the River mouth and they do not contain significant ^{224}Ra . Does the freshening represent an Amazon or Orinoco source?

Figure 2 is a plot of the $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. of all of the Atlantic samples as a function of salinity. The slightly freshened samples without excess ^{224}Ra form a distinct grouping with A. R.'s below 1.0. Also included on this plot are data collected during the TTO NAS cruise in the Atlantic during May 1981 (Moore et al. 1986). These data, which almost certainly represent waters derived from the Amazon estuary, lie in the same field as the April Atlantic samples which do not have excess ^{224}Ra . We consider this convincing evidence that the origin of these slightly freshened samples only 40 to 120 km NE of the Orinoco sea buoy is the Amazon estuary, almost 1300 km to the southeast.

c. Gulf of Paria Samples

During low flow most of the freshened water from the Orinoco plume flows northward along the coast and enters the Gulf of Paria (Muller-Karger et al. 1988). Within the Gulf of Paria the salinity, ^{226}Ra and ^{228}Ra contents do not change significantly; but, the ^{224}Ra activities decrease from 18 to 11 dpm/100L (Table 1). If this were due entirely to decay of ^{224}Ra , it would imply that the residence time of water in the Gulf was on the order of 2.5 days. However, because of the close proximity to coastal sediments, additions of ^{224}Ra unaccompanied by ^{226}Ra or ^{228}Ra could occur, making the residence time calculation based on ^{224}Ra a minimum. If some of the activity which we attribute to ^{224}Ra was due to ^{223}Ra , this would also cause the residence time estimates to be low.

d. Caribbean Samples

As the slightly freshened waters from the Gulf of Paria enter the Caribbean, rapid mixing with surface Caribbean water makes their identification based on salinity alone tenuous. However, the Gulf of Paria water carries a substantial ^{228}Ra signal into the Caribbean. Northern Caribbean waters have a characteristic $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. in the range 0.5-0.6 (Table 1). The waters near the Gulf of Paria have an activity ratio greater than 2.0 (Table 1). The presence of waters with an abnormally high $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. persist to a salinity of 36.4 ‰, making the Orinoco component easy to identify (Figure 3).

We have applied the mixing model described in equations 1-3 to these Caribbean waters to estimate the fraction of the Gulf of Paria endmember which they contain. For the ocean endmember we used data from samples 2,3,4,62,63 (Salinity=36.39, ^{226}Ra =6.85 dpm/100L, ^{228}Ra =3.78 dpm/100L). For the Gulf of Paria endmember we averaged all April samples from the Gulf (Salinity=34.15, ^{226}Ra =9.02 dpm/100L, ^{228}Ra =29.3 dpm/100L). The results of the endmember calculations which range from 0-100% are given in Table 2.

Some of the Caribbean waters exhibiting high $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. also contain excess ^{224}Ra . After these waters enter the Caribbean, they float as a buoyant surface layer isolated from contact with sediments which could add ^{224}Ra . Therefore, the decrease of ^{224}Ra in these waters may be used to estimate the time elapsed since the water left the Gulf of Paria. Figure 4 is a plot of the age of these waters (based on equation 4 and the endmember calculations) as a function of their distance from the Gulf of Paria. Most of the data indicate that the freshened plumes traveled with current speeds on the order of 5-20 cm/sec. If the samples contained significant activities of ^{223}Ra , the ages would be older and the speeds required would be lower. However, even if all of the activity which we attribute to ^{224}Ra was actually ^{223}Ra (an extremely unlikely possibility), the ages would only increase a factor of 3 and the speeds would decrease a factor of 3.

These freshened plumes also carry significant concentrations of methane (Jones and Amador, this volume). As expected, the waters which contain excess ^{224}Ra also contain methane, in fact these two tracers are highly correlated in the Gulf of Paria and Caribbean (Figure 5). The almost linear trend of this plot with a slope of 1.6 indicates that the methane has a mean residence time in the water on the order of 3 days (compared to the ^{224}Ra mean life of 5 days). Jones and Amador (this volume) have determined that the oxidation rate of methane in the water is too slow to have any impact on this time scale; therefore, the methane must be lost to the atmosphere. A stagnant film thickness of 40 μm and a plume depth of 15 m would be consistent with a residence time of methane on the order of 3 days.

6. Distribution of Radium isotopes During September-October

a. Estuary

Two river transects were completed during the fall, the first between 25-28 September and the second on 8 October. Although these were less than two weeks apart, significant changes in River and mixing zone properties were observed.

The mixing profiles of silica were considerably different in slope and intercept (Figure 6). The September sampling had a River endmember concentration of $156 \mu\text{M/kg}$; on 8 October the endmember Si had decreased to $126 \mu\text{M/kg}$. Both mixing profiles were essentially conservative with only a few points falling below the trend at salinities $>20 \text{‰}$.

During peak river flow in September, significant additions of ^{226}Ra occur in response to its release from fresh sediment entering the estuary. These additions increase the ^{226}Ra activity from 3.8 to 17.9 dpm/100L (Figure 7). However, this large pulse of ^{226}Ra was not present in the profile collected on 8 October; the highest activities recorded in the estuary during this sampling were only 10.8 dpm/100L (Figure 7). This change in ^{226}Ra activity must reflect a sharp decrease in the supply of fresh sediment to the mixing zone. Meade et al. (1983) have shown that the sediment discharge is highly correlated to water discharge; however, the ^{226}Ra data suggest that the sediment discharge drops abruptly after high discharge. A curious, unexplained feature of both the September and October ^{226}Ra data is a depression of the concentration in the samples collected in the 11-12 ‰ salinity range. Sampling density was not adequate to determine if this reflects removal of Ra within the system.

^{228}Ra increases in the mixing zone during September and October parallel the ^{226}Ra increases without exception (Figure 8). The increases of ^{228}Ra are much less dramatic than in April; and, because the ^{226}Ra activities double in the fall, the $^{228}\text{Ra}/^{226}\text{Ra}$ A. R.'s only reach values of 2.6 in September and 3.1 in October. This must be a function of the extremely short residence time of water in the estuary during high discharge and lower exposure of older sediments by mixing or dredging. The low $^{228}\text{Ra}/^{226}\text{Ra}$ A. R.'s require that either the sediments being mixed do not contain significant ^{228}Ra which has accumulated from ^{232}Th decay or the water does not remain in the system long enough to acquire a high activity from the sediment source.

^{224}Ra activities do increase dramatically during Sept.-Oct. (Figure 8). Activities are 2 to 3 times greater than ^{228}Ra throughout the estuary. This means that the water does remain in the estuary

long enough to acquire a ^{224}Ra signal from sediments which have produced ^{224}Ra from ^{228}Th decay. Sediments regenerate ^{224}Ra activity rapidly, but ^{228}Ra activity much more slowly. The high $^{224}\text{Ra}/^{228}\text{Ra}$ A. R.'s imply that sediment mixing during Sept.-Oct. comprises primarily the same sediments being recycled, rather than exposure of older sediments which have been stored in the estuary for months to years. The high release of ^{228}Ra accompanied by an equal activity of ^{224}Ra during April implies that much of the sediment being mixed during low discharge has been stored in the estuary for a considerable time (1-20 yrs). Dredging of the shipping channel during the spring or deep sediment mixing by physical or biological processes must expose these older sediments to the water in the mixing zone.

b. Atlantic Ocean Samples

The $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. in the Atlantic shows the same features as in April; but, the magnitude of the changes is more subdued (Figure 9). Three samples had $^{228}\text{Ra}/^{226}\text{Ra}$ A. R.'s below 1.0 at salinities of 34-35 ‰. These are almost certainly of Amazon origin. The characteristic $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. for Orinoco samples of this salinity is in the range 1.4-2.4.

All of the Atlantic samples except for the 3 of Amazon origin had excess ^{224}Ra which correlated well with ^{228}Ra (Figure 8). Apparently these waters move quickly into the Atlantic during high discharge.

c. Gulf of Paria

Waters entering the Gulf of Paria had ^{224}Ra activities about a factor of two higher than ^{228}Ra ; but, within the Gulf this A. R. decreased to about one. However, it is especially risky to use the Ra activities in these waters during high discharge to deduce residence times because numerous discharge channels of the Orinoco into the Gulf are active during this period. These additions of water and sediment certainly add Ra isotopes to the Gulf. Short term changes in the $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. at the River mouth and additions of Ra isotopes to the Gulf make the definition of a Gulf of Paria endmember uncertain during the fall.

d. Caribbean Sea

The water entering the Caribbean carries a strong signal in its $^{228}\text{Ra}/^{226}\text{Ra}$ A. R. which ranges from almost 2.5 at the entrance to the Gulf of Paria to 0.5 at salinities of 36 ‰ (Figure 10). There is no evidence of significant Amazon influence on the waters sampled in the eastern Caribbean.

Most of the waters sampled within 500 km of the Gulf of Paria contained excess ^{224}Ra . However, we cannot use the steady state model represented by eq. 4 to calculate water ages in the fall because the Gulf of Paria ^{224}Ra endmember is not constrained to a constant value or to a constant relationship to ^{228}Ra during this time period (Table 2).

7. Conclusions

The data from this study have demonstrated the usefulness of Ra isotopes in studies of water and sediment mixing processes at the mouth of a major river. Distributions of ^{226}Ra were controlled by the supply of fresh river sediment to the estuary. ^{224}Ra and ^{228}Ra activities were derived from this source and from the mixing of sediment which had been in the estuary for weeks to years.

During low discharge (April), considerable mixing of older sediment by physical or biological processes or dredging maintained high activities of ^{228}Ra in the estuary and produced the highest $^{228}\text{Ra}/^{226}\text{Ra}$ A. R.'s yet measured in any estuary. During high discharge (September) a large fraction of the ^{228}Ra was derived from desorption from fresh sediment rather than mixing of older sediments. Activities of ^{224}Ra were high in the estuary during both high and low discharge, indicating that considerable mixing of recently introduced sediment must occur during each period. The high $^{224}\text{Ra}/^{228}\text{Ra}$ A. R.'s measured in September imply that the same sediments were being resuspended during high discharge, and that there was little exposure of older sediments which had been stored in the estuary for months to years. During April ^{224}Ra and ^{228}Ra activities in the water were about equal, indicating that most of the sediment being resuspended had been stored in the estuary long enough to reestablish radioactive equilibrium in the ^{232}Th decay series, 20 years.

^{224}Ra released in the estuary provides a means of estimating the length of time since the water was in contact with sediments. The most fruitful region for such studies during this investigation was the eastern Caribbean during April. Samples collected at various distances from the Gulf of Paria revealed that current speeds on the order of 5 to 20 cm/sec were required to transport the freshened waters hundreds of km into the Caribbean.

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Table 1 April Data

Sampl	Station	Latitude	Longitude	Temp	Salinity	Silica	Ra-224	Th-228	ex Ra-224	Ra-226	Ra-228
		°N	°W	°C	ppt	µm/L	dpm/100L	dpm/100L	dpm/100L	dpm/100L	dpm/100L
1											
2											
3	1	21 34	70 40	25.5	36.21		1.42	0.98	0.45	8.51	3.40
4	D/S	17 17	66 53	27.0	36.45		2.03	1.17	0.87	7.34	3.97
5	F/S	15 41	65 33	27.1	36.12		0.94		0.14	6.62	3.71
6	G/S	15 20	65 18	27.5	36.15		0.83	0.68	0.15	6.28	3.20
7	I/S	11 11	64 43	26.9	36.19		1.26		0.56	6.42	4.62
8	J/S	13 45	64 24	27.0	36.20		0.95	0.71	0.24	6.70	3.69
9	L/S	12 53	63 46	27.2	36.22		1.06	0.33	0.73	6.58	4.51
10	M/S-1	11 49	62 50	26.8	36.02		0.92		0.62	6.71	5.70
11	UW	11 25	61 58	27.1	36.20		4.32	0.28	4.05	7.18	9.69
12	UW	11 18	61 33	27.0	36.17		2.07	0.25	1.82	6.96	5.92
13	UW	11 04	61 01	27.0	36.08	0.32	1.37	0.23	1.14	7.87	5.75
14	UW	10 14	59 37	27.4	35.85	0.81	0.77	0.25	0.52	6.96	5.51
15	R1/S	09 50	59 10	27.6	35.45	1.22	0.57	0.31	0.45	7.00	7.35
16	R2/S	09 32	59 31	27.6	35.45		0.76	0.18	0.39	7.04	7.96
17	R3/S	09 10	59 50	27.5	35.75	1.05	0.65	0.38	0.27	7.34	7.27
18	R4/S	08 57	60 11	26.8	35.51	9.88	17.98		17.98	9.14	45.70
19	R5/S	08 38	60 30	28.6	7.40	98.26	55.05		54.95	13.53	35.58
20	17A UW	08 38	60 30	28.6	7.90		50.28	0.01	50.27	13.30	40.30
21	18 UW	08 42	60 27	29.5	10.73	72.90	48.59		48.49	13.34	40.55
22	19 UW	08 42	60 27	29.4	17.49	49.30	39.05		38.95	12.12	44.36
23	20 UW	08 43	60 26		23.83	39.50	57.91		57.81	11.64	66.46
24	20A UW	08 43	60 26		25.76	30.86	82.21		82.11	14.77	74.59
25	21 UW	08 45	60 24		30.63	28.40	105.70		105.40	17.62	113.47
26	21A UW	08 45	60 24		32.71	19.40	88.08		87.78	13.15	88.63
27	22 UW	08 53	60 16		35.28		42.35		42.05	9.68	47.14
28	23 R4/S	08 57	60 11		35.58		28.85		28.55	8.35	34.57
29	24 R4/S	08 57	60 11		35.76		14.68		14.38	8.24	29.00
30	25 UW	09 10	60 23	27.0	35.99		18.28		17.98	7.55	25.29
31	26 R6/S	09 30	60 36	27.0	36.04	3.92	9.98		9.68	8.32	25.21
32	27 UW	09 49	60 56	26.7	34.78	12.12	30.75		30.45	8.88	27.00
33	28 R7/S	09 53	61 07	27.2	34.13	8.87	14.09		13.79	8.29	29.51
34	29 UW	09 56	61 24	27.5	33.31	12.71	18.13		17.83	9.15	28.55
35	30 R9/S	09 58	61 45	27.9	32.16	14.83	29.43		29.13	10.98	27.78
36	31 R9/S	09 57	61 38	27.3	34.46		22.09		21.79	8.15	22.49
37	32 UW	10 00	61 41	27.2	34.99	4.90	9.84		9.54	8.23	22.30
38	33 UW	10 00	61 46	27.1	34.92	4.22	12.85		12.55	7.67	22.78
39	34 UW	10 02	62 00	27.0	34.38	6.08	22.61		22.31	10.04	30.06
40	35 UW	10 06	62 05	27.2	34.66	5.57	16.66		16.36	8.58	22.74
41	36 P1/S	10 15	62 10	27.5	33.27	6.93	17.62		17.32	9.11	29.70
42	37 P3/S	10 30	62 00	28.2	34.22	4.90	20.85		20.55	9.48	31.00

Table 1 April Data

Sampl	Station	Latitude	Longitude	Temp	Salinity	Silica	Ra-224	Th-228	ex Ra-224	Ra-226	Ra-228
43	38	P5/S	10 44	61 48	27.7	34.16	11.08		10.78	8.90	34.35
44	39	P5/S	10 44	61 49	27.4	34.19	11.45		11.15	9.20	31.10
45	40	P5/S	10 44	61 49	27.3	34.42	11.89		11.59	8.83	26.89
46	41	UW	10 49	61 53	24.8	35.45	10.79		10.49	7.35	15.87
47	42	UW	10 55	61 57	24.4	36.29	1.23		0.93	6.06	9.24
48	43	UW	11 00	62 01	25.7	35.93	4.64		4.34	7.28	17.14
49	44	UW	11 06	62 06	26.8	35.92	3.57		3.27	9.04	10.79
50	45	UW	11 14	62 12	27.0	36.21	0.77		0.47	8.54	5.60
51	46	N/S	11 18	62 20	27.2	36.16	0.85		0.55	8.05	6.25
52	47	UW	11 31	62 34	27.1	35.94	3.13		2.83	7.88	13.68
53	48	M/S-2	11 44	62 43	27.3	36.14	1.05		0.75	6.82	7.35
54	49	M/S-2	11 44	62 43	27.3	36.18				6.89	5.79
55	50	M/S-2	11 42	62 41	26.9	36.23	1.70		1.40	7.76	7.75
56	51	M/S-2	11 47	62 45	27.0	36.22	1.14	0.48	0.66	7.87	5.75
57	52	UW	11 45	64 05	24.9	36.53	1.31		1.01	6.33	4.18
58	53	T/S	11 41	64 59	25.8	36.88	0.81	0.50	0.31	7.76	5.20
59	54	S/S	11 22	64 46	25.3	37.02	1.10		0.80	7.31	4.31
60	55	UW	10 30	64 13	25.8	36.98	4.54		4.24	7.64	4.06
61	56	U/S	12 20	65 40	27.0	36.25				7.50	6.50
62	57	H/S	14 39	64 44	27.8	36.12	0.34			7.50	4.80
63	58	Y/S	15 22	62 50	27.2	36.12				7.50	4.20
64	59	M/S-3	11 41	62 41	27.0	36.20	0.18			7.50	5.00

Table 2 Sept-Oct Data

Sample i (depth)	Station	Latitude °N 'N	Longitude °W 'W	Temp °C	Salinity ppt	Silica µm/L	Ra-224 dpm/100L	Th-228 dpm/100L	ex Ra-224 dpm/100L	Ra-226 dpm/100L	Ra-228 dpm/100L
1											
2											
3	A/F	19 04	67 32	28.50	35.480		1.05	1.01	0.04	6.79	5.23
4	D/F-1	17 17	66 53	28.30	35.180		0.93	0.66	0.27	6.78	4.77
5	UW	16 48	66 35	28.60	34.780	1.83	0.96	0.95	0.01	7.75	5.91
6	UW	16 24	66 18	28.50	33.990	3.31	0.89	0.70	0.19	8.04	7.32
7	UW	16 05	66 02	28.50	33.460	5.02	0.49	0.99	-0.50	7.79	7.83
8	G/F-1	15 21	65 29	28.50	33.550		0.72	0.73	-0.01	7.42	7.09
9	Y/F-1	15 22	62 51	28.50	34.900		0.81	0.96	-0.15	7.53	3.97
10	UW	15 22	63 02	29.10	33.980	3.94	1.08	0.64	0.45	7.61	6.20
11	Z/F	15 13	63 04	29.00	33.045	6.31	1.06	0.98	0.08	8.01	8.52
12	L/F-1	12 56	63 45	29.00	32.858	6.58	0.62	0.12	0.12	8.36	9.83
13	UW	11 56	62 51	29.10	31.398	14.69	2.10	0.31	1.79	7.95	9.72
14	M/F-1	11 48	62 45	28.60	30.473	14.86	3.52	0.94	2.58	8.00	15.81
15	UW	11 43	62 53	29.00	30.700	14.86	2.85		3.20	8.00	16.18
16	UW	11 45	62 55	28.30	31.989	11.20	4.12		3.62	7.95	15.73
17	M/F-2	11 27	62 37	27.90	32.905	10.38	3.61		3.11	7.53	10.74
18	UW	11 03	62 03	28.20	34.575	5.66	2.54	0.51	2.03	7.69	8.62
19	UW	10 45	61 48	29.50	20.439	47.18	6.86		6.36	10.70	24.36
20	R6/F	09 31	60 36	29.15	11.430	111.47	47.36	1.33	46.03	11.52	22.75
21	R4/F-1	08 57	60 10		36.398	15.46	18.36	0.30	18.06	9.01	13.07
22	R4/F-1	08 57	60 10	31.08	5.339	132.50	50.51		50.01	11.92	19.67
23	R4/F-1	08 57	60 10	30.28	2.259	145.46	36.78		36.28	8.24	16.81
24	UW	08 50	60 18	29.69	9.711	116.82	74.07		73.57	17.93	35.02
25	UW	08 43	60 25	29.77	1.396	149.09	51.89		51.39	3.86	6.03
26	R5/F-1	08 35	60 30	29.10	0.027	152.27	8.15		7.65	3.81	5.65
27	UW	08 48	60 21	29.00	6.827	128.64	97.65		97.15	16.81	33.62
28	R4/F-2	08 57	60 10	29.32	19.569	78.64	37.38		36.88	11.35	30.04
29	R4/F-2	08 57	60 10	29.33	19.266	79.55	37.39		36.89	11.08	29.09
30	R3/F	09 10	59 50	29.88	34.752	10.46	4.04	1.14	2.90	7.97	14.59
31	R1/F	09 50	59 10	29.44	34.496	3.64	1.00	0.74	0.26	6.22	4.39
32	R1/F	09 48	59 09	29.43	34.403	4.09	0.88	0.83	0.05	6.74	4.46
33	UW	09 48	59 20	29.33	34.592	3.56	0.79	0.83	-0.04	6.97	6.12
34	R2/F	09 30	59 30	28.84	35.222	3.33	4.09	0.60	3.48	7.90	11.73
35	UW	09 22	59 43	28.87	35.100	8.00	5.17	0.39	4.77	8.13	19.15
36	UW	09 09	60 01	29.08	34.982	9.33	2.47	0.84	1.63	8.52	11.95
37	UW	09 04	60 07	28.77	35.178	7.33	2.77		2.27	6.34	10.35
38	UW	09 03	60 08	28.58	28.528	40.00	35.28		34.78	9.55	24.72
39	UW	09 02	60 10	28.87	24.944	49.78	42.42		41.92	11.40	33.07
40	UW	09 02	60 13	28.63	13.767	98.26	70.44		69.94	14.18	30.22
41	UW	09 02	60 13	28.45	13.479	100.00	95.99		95.49	15.55	25.60
42	UW	09 00	60 07	28.00	32.055	20.87	15.33		14.83	8.68	16.88

Table 2 Sept-Oct Data

Sample	Station	Latitude	Longitude	Temp	Salinity	Silica	Ra-224	Th-228	ex Ra-224	Ra-226	Ra-228
43	100	08 50	60 19	28.30	3.446		52.42		51.92	8.09	15.88
44	101	09 52	60 59	27.39	25.011	57.83	47.95		47.45	13.07	24.81
45	102	10 15	62 11	28.90	18.701	60.44	14.52		14.02	10.90	19.27
46	103	11 13	62 18	28.27	30.568	29.27	5.42		4.92	8.82	16.24
47	104	11 25	62 31	28.18	31.937	17.31	3.59		3.09	8.16	10.39
48	105	11 50	62 56	27.66	33.188	10.90	2.31	0.76	1.55	7.96	8.29
49	106	11 52	62 56	28.31	32.022	14.53	2.71	0.75	1.97	8.31	11.68
50	107	11 55	62 58	28.48	32.072	13.89	2.38	0.72	1.66	8.45	11.89
51	108	12 39	63 40	29.36	32.916		0.98	0.88	0.10	7.90	8.36
52	109	11 51	62 49		30.387	11.90	3.21	0.72	2.48	8.26	14.30
53	110	11 05	62 00		27.653	22.90	12.36	1.35	11.01	10.42	25.38
54	111(120)	11 05	62 00		36.520	6.30	2.87		2.37	7.34	2.99
55	113(20)	11 06	62 00		36.601	2.40	1.43		0.93	7.39	3.38
56	114	10 41	61 34		22.122	29.90	8.01		7.51	11.08	20.76
57	115	10 09	62 10		15.586	71.20	27.94		27.44	10.70	20.02
58	116	10 03	62 03		12.321	83.60	26.94		26.44	10.00	16.80
59	117	08 59	60 08		35.292	10.20	28.30	1.19	27.11	7.83	17.00
60	119	08 38	60 28		0.023	125.90	7.27		6.77	2.85	6.05
61	120	08 46	60 24		4.854	109.20	49.83		49.33	10.80	20.48
62	121	08 46	60 23		3.741	113.20	39.11		38.61	8.43	15.65
63	122	08 50	60 19		8.851	97.10	56.29		55.79	10.60	20.29
64	123	08 58	60 11		20.335	59.20	74.20		73.70	10.70	23.34
65	124	08 57	60 10		29.337	33.50	73.00		72.50	8.63	27.22
66	125	08 57	60 11		12.106	86.00	50.45		49.95	8.62	19.09
67	126	08 56	60 13		7.282	101.50	52.23		51.73	11.36	21.40
68	127	08 55	60 13		4.879	108.50	48.31		47.81	10.00	17.98
69	128	09 25	60 33		11.021	91.50	94.36		93.86	8.46	16.67
70	129	09 47	60 58		15.407	76.80	67.52		67.02	9.24	18.51
71	130	09 58	61 36		15.448	73.20	25.02		24.52	9.38	17.38
72	131	10 22	62 09		17.453	58.10	21.45		20.95	11.95	22.24
73	132	12 51	63 52		32.653	6.80	2.31	0.39	1.92	9.06	10.36
74	133	15 28	65 26		33.871	7.60	1.51		1.01	7.31	7.27
75	134	13 52	67 10		36.184	0.70	1.09	0.73	0.36	7.42	4.18
76	135	17 20	66 54		34.665	1.40	1.75	0.74	1.01	7.40	5.35

Table 3

April Caribbean Samples

Fri, Aug 3, 1990 12:13 PM

Sample	Salinity	f Paria	ex Ra-224	228/226	CH4	Distance	Age
	ppt		dpm/100L	A. R.	nM/kg	km	days
1							
2							
3							
4	2	36.45	0.00	0.87	0.540	2	740
5	3	36.12	0.00	0.14	0.560		556
6	4	36.15	0.00	0.15	0.510	3	520
7	5	36.19	0.05	0.56	0.720		400
8	6	36.20	0.00	0.24	0.550		360
9	7	36.22	0.04	0.73	0.680	2	260
10	8	36.02	0.09	0.62	0.850		105
11	41	35.45	1.00	10.49	2.159		2
12	42	36.29	0.40	0.93	1.524	9	10
13	43	35.93	1.00	4.34	2.350	18	20
14	44	35.92	0.22	3.27	1.190	8	30
15	45	36.21	0.03	0.47	0.660	6	45
16	46	36.16	0.07	0.55	0.780	5	65
17	47	35.94	0.55	2.83	1.740	11	102
18	48	36.14	0.17	0.75	1.077	4	105
19	49	36.18	0.09		0.840	4	105
20	50	36.23	0.14	1.40	0.990	9	105
21	51	36.22	0.05	0.66	0.730	9	105
22	52	36.53	0.03	1.01	0.660		270
23	53	36.88	0.03	0.31	0.670		295

Figure Captions

1. Profiles of Ra isotopes through the Orinoco estuary during April 1988.
2. Activity ratios of $^{228}\text{Ra}/^{226}\text{Ra}$ as a function of salinity for samples collected in the Atlantic Ocean adjacent to the Orinoco estuary in April 1988.
3. Activity ratios of $^{228}\text{Ra}/^{226}\text{Ra}$ as a function of salinity for samples collected in the Caribbean Sea during April 1988.
4. Ages of freshened waters as a function of distance from the Gulf of Paria for samples collected in April 1988. The lines represent current speeds required to transport the waters into the Caribbean.
5. Methane versus excess ^{224}Ra in samples collected in the Gulf of Paria and Caribbean during April 1988.
6. Silica versus salinity for samples collected from the Orinoco estuary in September and October 1988.
7. Profiles of ^{226}Ra through the Orinoco estuary for samples collected in September and October 1988.
8. Profiles of Ra isotopes through the Orinoco estuary during September 1988 and October 1988.
9. Activity ratios of $^{228}\text{Ra}/^{226}\text{Ra}$ as a function of salinity for samples collected in the Atlantic Ocean adjacent to the Orinoco estuary during the period September to October 1988.
10. Activity ratios of $^{228}\text{Ra}/^{226}\text{Ra}$ as a function of salinity for samples collected in the Caribbean Sea during the period September to October 1988.

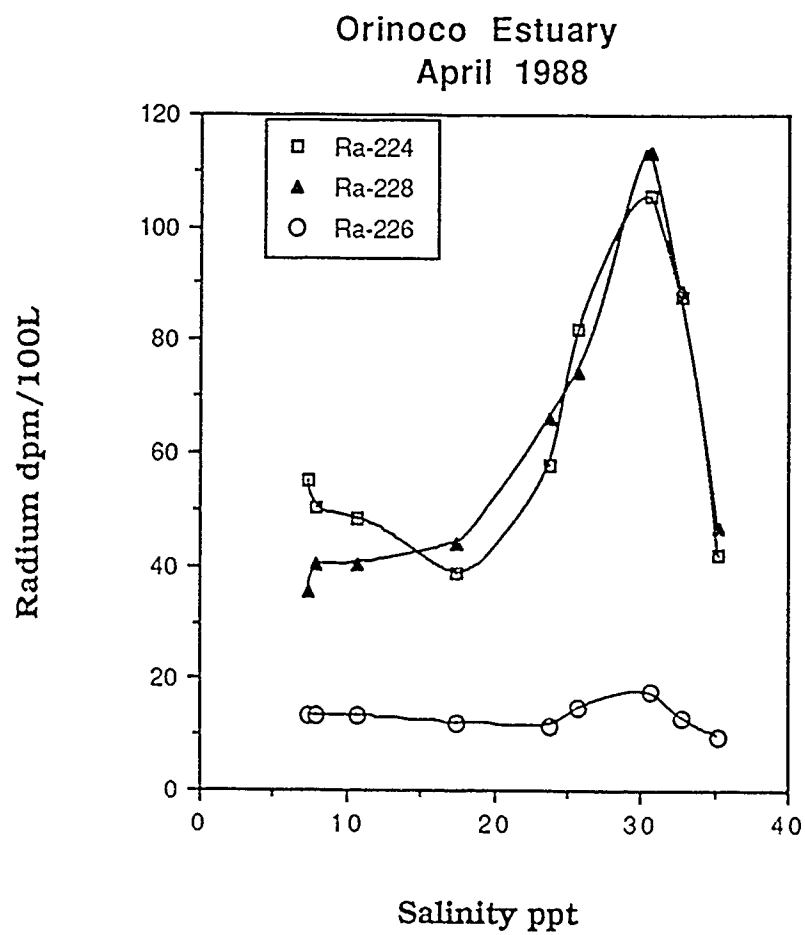


Figure 1

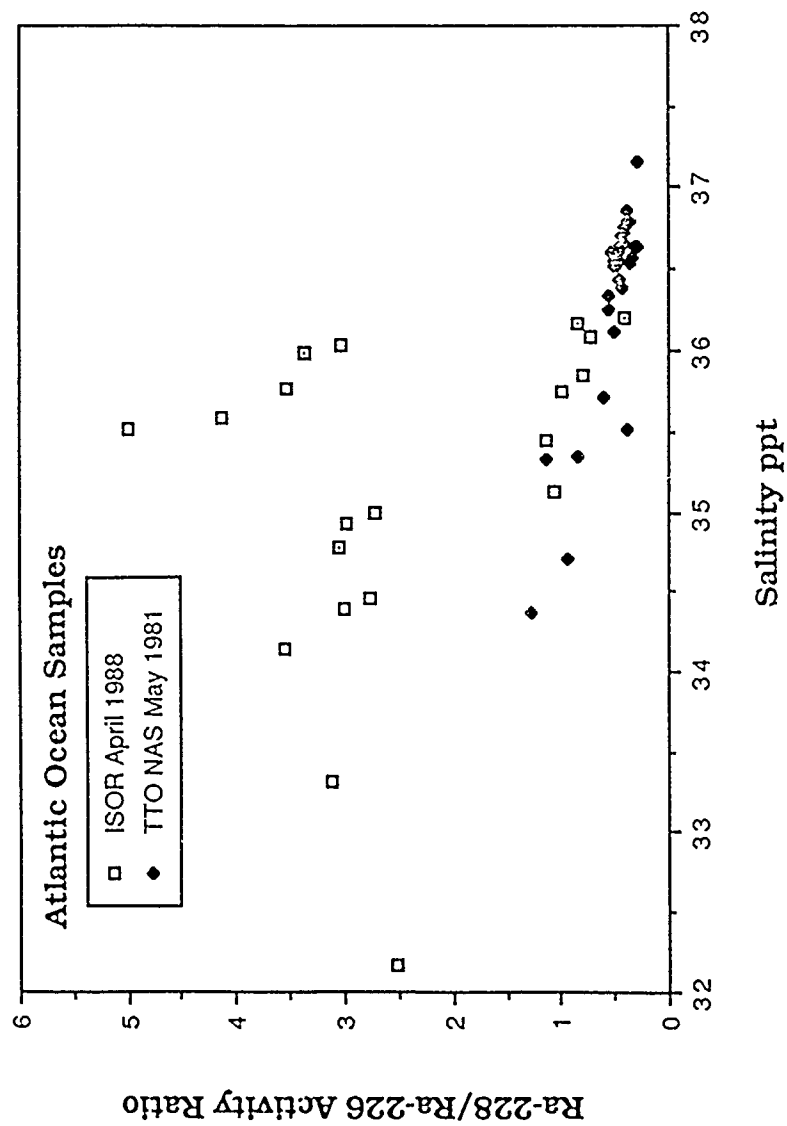


Figure 2

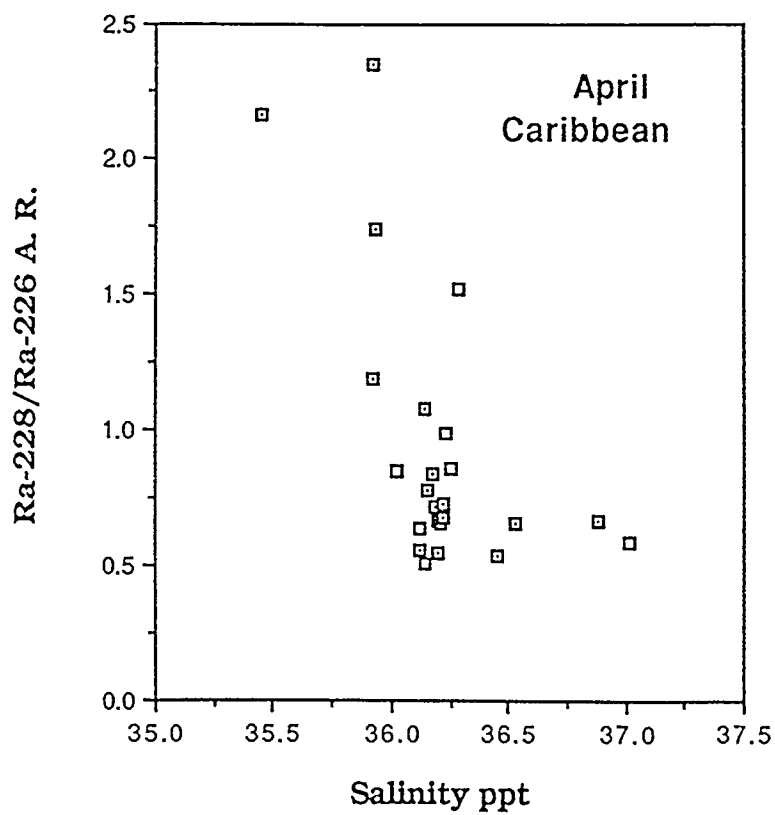


Figure 3

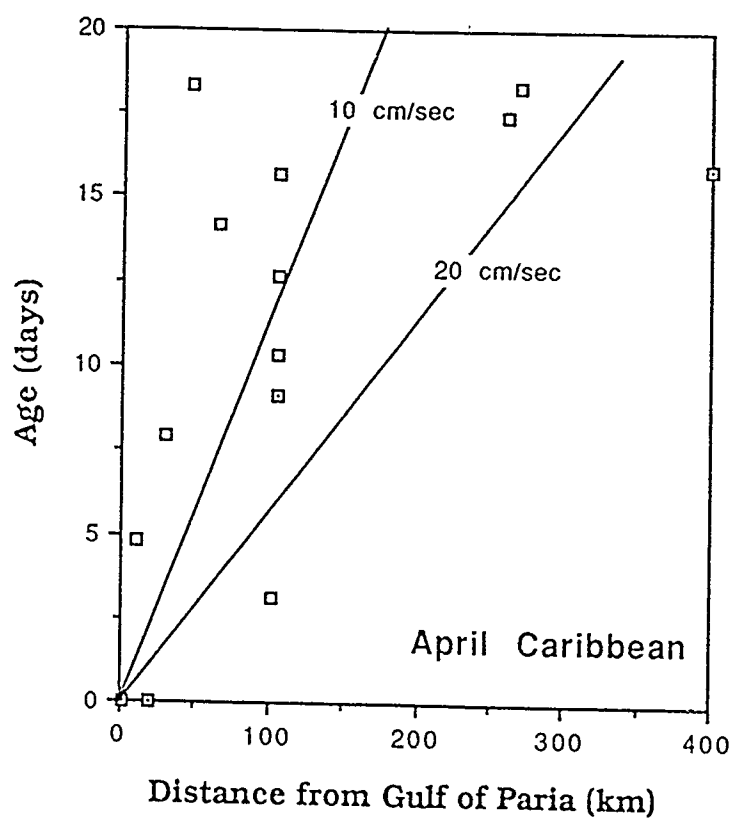


Figure 4

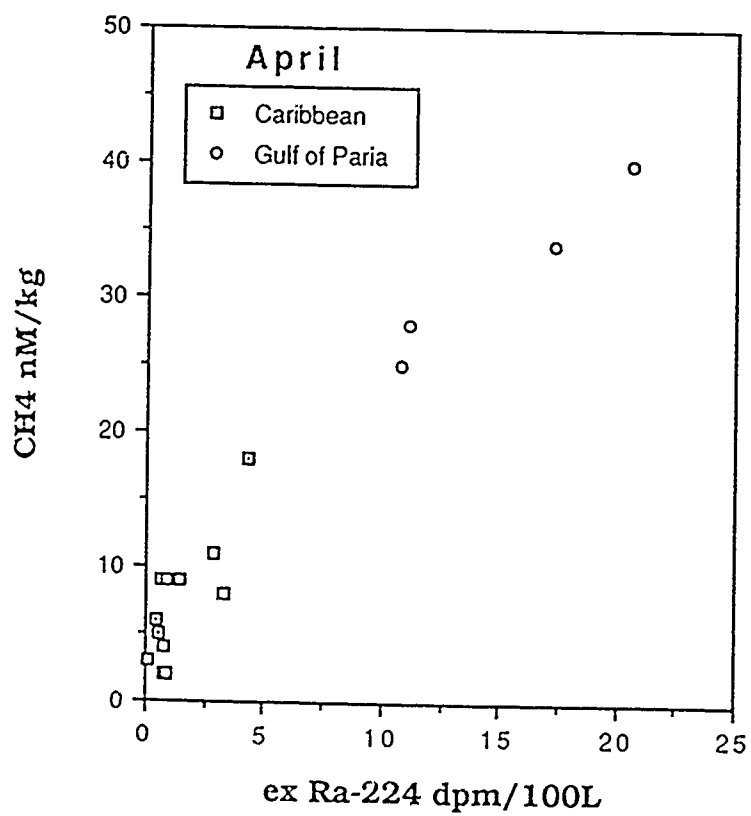


Figure 5

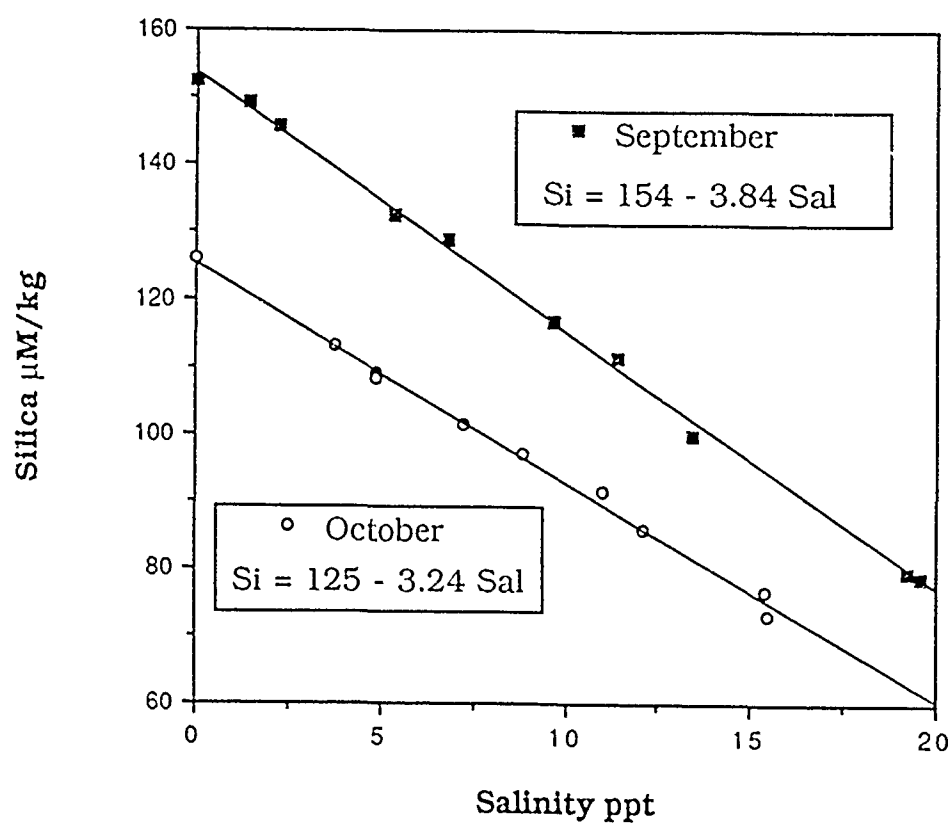


Figure 6

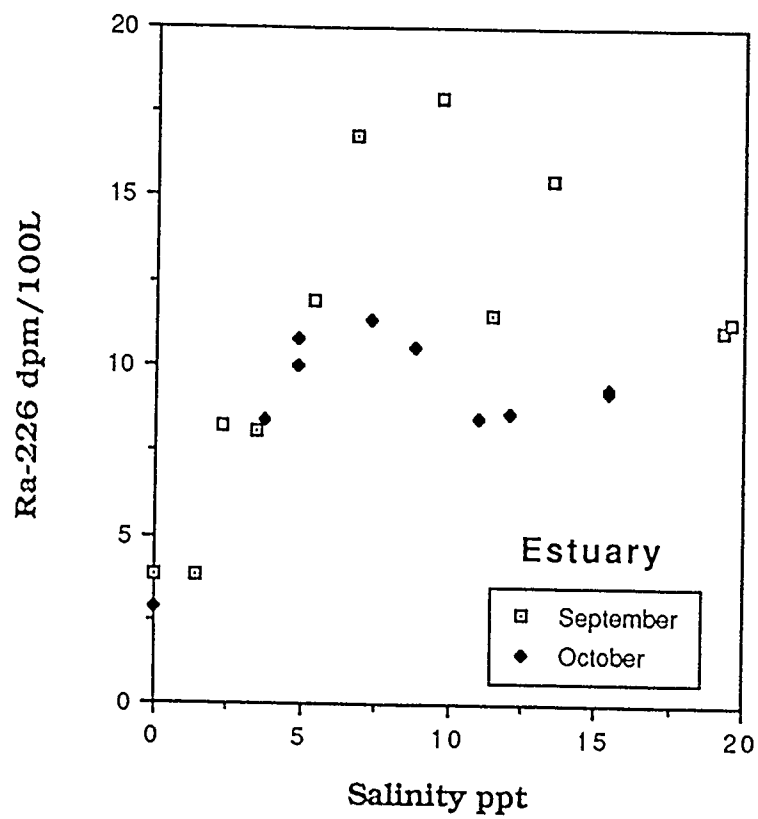


Figure 7

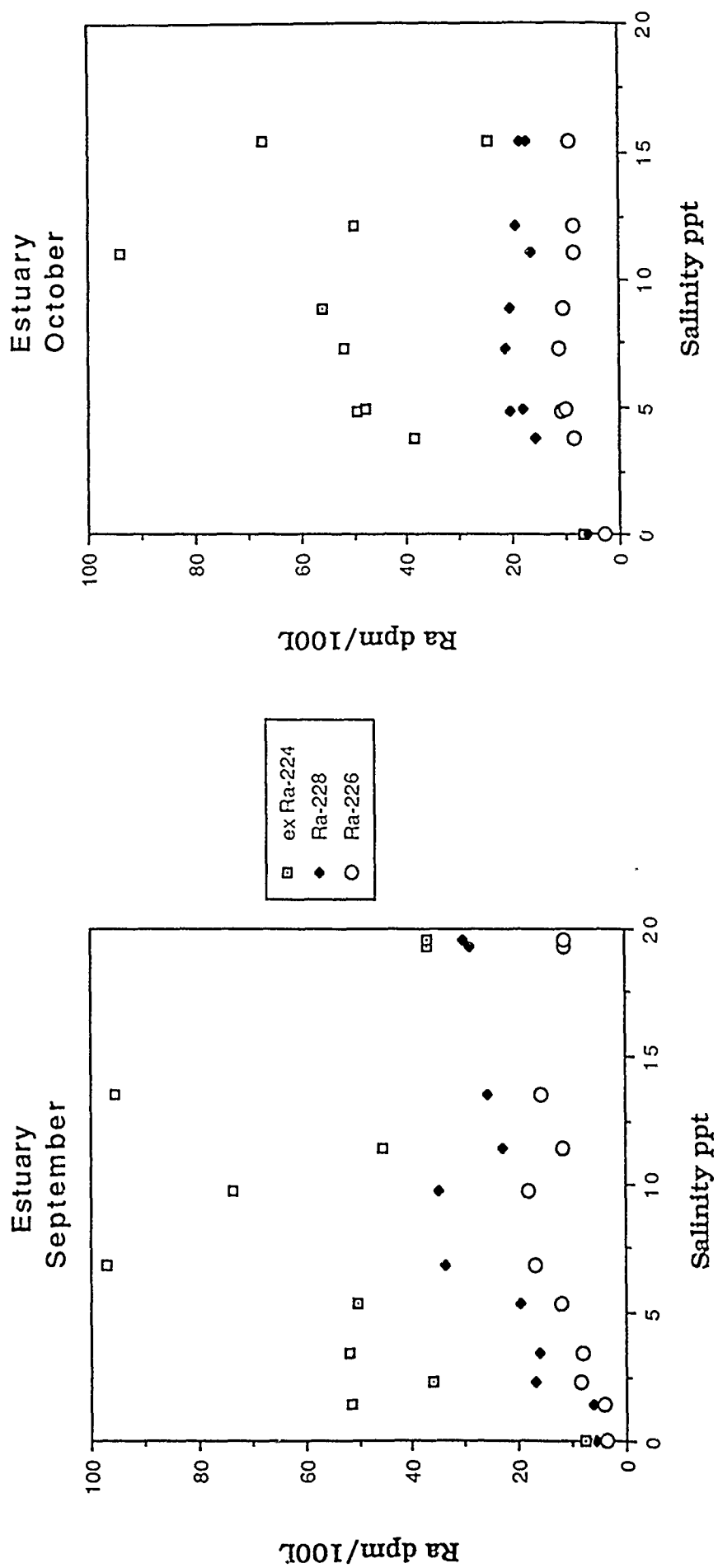


Figure 8

Atlantic Ocean Samples
September-October

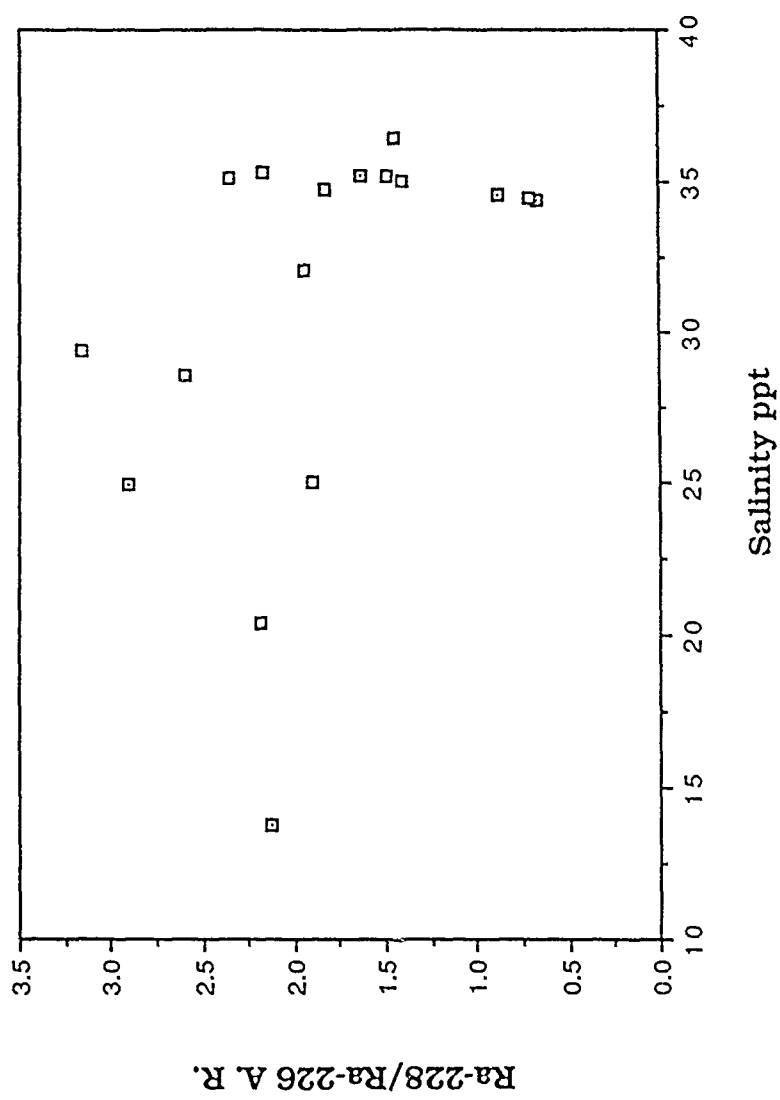


Figure 9

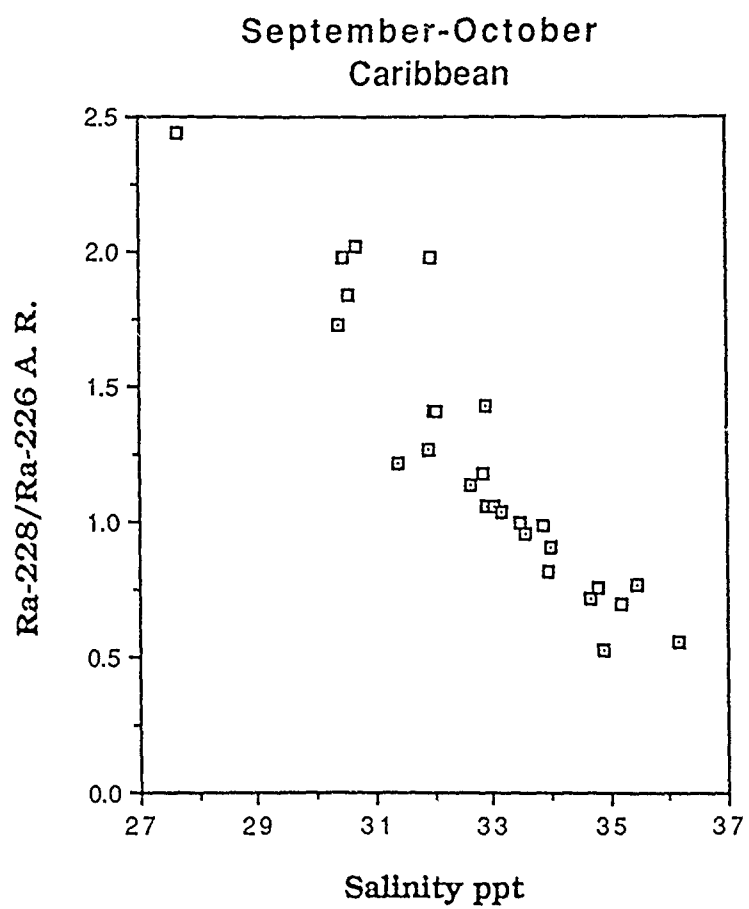


Figure 10