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High fluxes of radium and barium from the mouth of the Ganges–Brahmaputra River during low river discharge suggest a large groundwater source

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Abstract

The annual flux of sediment from the Ganges–Brahmaputra River is among the highest in the world. The desorption of ^{226}Ra and Ba from this sediment produces a major point source of ^{226}Ra and Ba input to the ocean. Highest ^{226}Ra and Ba fluxes are expected to occur during high river flow (June–September) when most of the sediment is discharged. Surprisingly, during low discharge of the Ganges–Brahmaputra River, fluxes of ^{226}Ra and Ba to the northern Bay of Bengal are comparable to expected river-derived fluxes during peak river discharge. A large non-riverine source of Ra and Ba is required to explain the high fluxes during low river discharge. I suggest that this source is submarine discharge of groundwater containing high concentrations of ^{226}Ra and Ba. Total annual fluxes of ^{226}Ra and Ba to the ocean from this system are significantly greater than estimated previously. © 1997 Elsevier Science B.V.

Keywords: radium; barium; ground water; rivers and streams; Bay of Bengal

1. Introduction

The variability of ^{226}Ra and Ba concentrations in surface ocean water is rather small except in coastal regions. When plotted with respect to salinity, dissolved Ra and Ba concentrations fall above the conservative mixing line between rivers and the open ocean. The accepted reason for these higher concentrations in coastal waters is the desorption of these elements from sediment delivered by rivers. Li et al. [1] first demonstrated ^{226}Ra enrichments in the Hudson estuary and explained the nonconservative increase by the desorption of ^{226}Ra from riverine particles. Numerous additional studies [2–4] have confirmed that the desorption of ^{226}Ra and Ba from riverine sediments is a significant source of these elements to the ocean. Because the annual flux of sediment from the Ganges–Brahmaputra rivers is among the highest in the world, this system should be one of the major point sources of Ba and ^{226}Ra input to the ocean. Highest ^{226}Ra and Ba fluxes are expected to occur during high river flow (June–September) when most of the sediment is discharged.

Veeh et al. [5] measured activities of ^{226}Ra in Spencer Gulf, Australia, that exceeded oceanic activities even though there is negligible surface fresh water/sediment input to the Gulf. After normalizing to offshore salinity due to evaporation within the Gulf, some samples contained double the offshore ^{226}Ra concentration. They concluded that an additional source was required to explain the high ^{226}Ra activities, and speculated that direct input of subma-

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rine groundwater containing high ^{226}Ra activities was this source.

Moore [6] demonstrated that brackish groundwater was the primary source of ^{226}Ra enrichment in the South Atlantic Bight. He hypothesized that progressive movement of a salt front into an aquifer and discharge of the brackish water into the near shore zone would provide a source of ^{226}Ra to the coastal ocean. This input would be important as long as the salt front advanced into the aquifer and there was net advection of brackish groundwater into the ocean. Shallow aquifers along this coast have been impacted by groundwater mining, sea level rise and dredging. Shaw et al. [7] concluded that high Ba concentrations in the South Atlantic Bight result from the input of groundwater containing elevated Ba concentrations.

The realization that groundwater input of Ra and Ba plays a role in the supply of these elements to the ocean requires a re-evaluation of the global input of these elements. Are groundwater inputs restricted to a few regions, or are they important globally? Data from other regions are required to answer this question.

This paper presents new data on Ra isotopes and Ba in the Ganges–Brahmaputra River mixing zone. These require a re-evaluation of the sources of these elements to the Bay of Bengal. An earlier study [4] during low river discharge in January–February 1987 concluded that the measured fluxes of ^{226}Ra and Ba were an order of magnitude higher than could be explained by river discharge and immediate desorption of these elements from suspended particles. Here, I propose the high fluxes from this system are sustained by submarine groundwater input.

2. Measurements

In March 1991, 100 l surface samples for Ra analyses were pumped from the Bay of Bengal into plastic drums. Additional small volume samples for Ba analysis were taken by a plastic bucket cast. Subsamples from the drums or buckets were immediately filtered through a 0.45 μm filter cartridge. One subsample was reserved for salinity measurement, another was reserved for Ba analysis. The Ba samples were acidified with nitric acid approximately 2 months after collection. The large volume samples were passed through a 0.45 μm filter car-

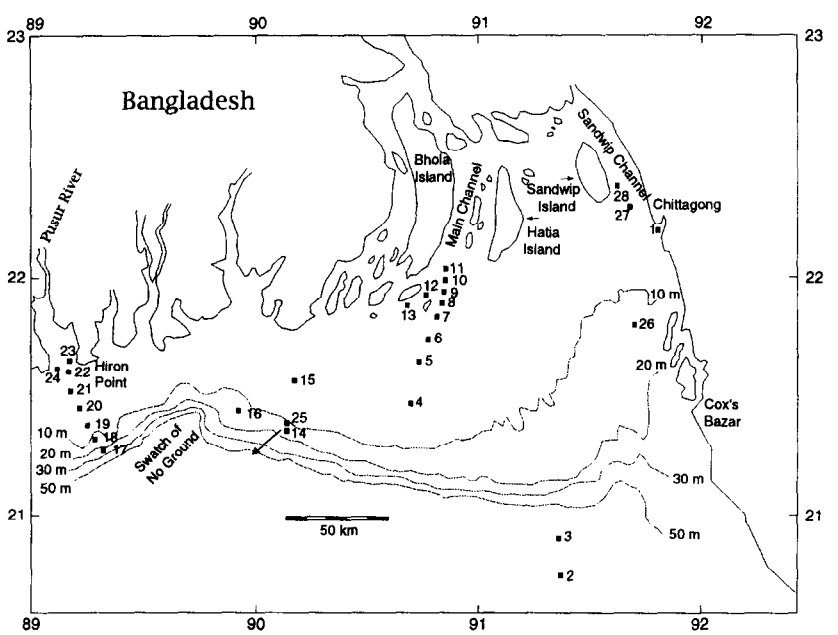

Fig. 1. Index map of the study area showing sampling locations for the March 1991 cruise and depth contours. An arrow near station 14 indicates a residual current of 0.52 m s^{-1} at this station.

tridge and through a column of Mn fiber to remove Ra isotopes [8].

In the laboratory each Mn fiber sample was partially dried and placed in an air circulation system described by Rama et al. [9]. Helium was circulated over the Mn fiber to sweep the decay product of ^{224}Ra, ^{220}Rn, through a 0.1 l scintillation cell where alpha particles from the decay of Rn and daughters were recorded by a photomultiplier tube (PMT) attached to the scintillation cell. Most of the activity within the detector resulted from the decay of ^{220}Rn and its daughter, ^{216}Po. A small contribution was from ^{219}Rn and its daughter, ^{215}Po. The samples were measured again within 2 weeks to assess the activity of ^{228}Th, the parent of ^{224}Ra on the Mn fiber. The activity of ^{224}Ra was decay-corrected to the time of collection, 5–8 days earlier, and corrected for the contribution of ^{228}Th.

After completing the ^{224}Ra and ^{228}Th measurements, the Mn fibers were leached with HCl to remove the long lived Ra isotopes. The Ra was precipitated with $BaSO_4$. The precipitant was aged 2 weeks to allow ^{222}Rn and its daughters to equilibrate with ^{226}Ra. The samples were measured in a gamma-ray spectrometer to assess the activities of ^{226}Ra and ^{228}Ra [10].

The Ba samples were analyzed using isotope dilution–inductively coupled plasma mass spectrometry at the Skiddaway Institute of Oceanography following the procedure of Klinkhammer and Chan [11]. A ^{135}Ba-enriched spike was added and equilibrated with the acidified sample before injection.

Table 1
March 1991 samples from the Bay of Bengal

Sample	Date (1991)	Time	Salinity (ppt)	Ba (nM)	^{226}Ra	^{228}Ra	^{224}Ra
1	11 March	2200	18.62	465	70	157	133
2	12 March	1345	31.19	79	12	25	2
3	12 March	1900	29.61	90	15	34	13
4	13 March	0700	30.15	144	24	45	74
5	13 March	0900	28.40	202	36	74	83
6	13 March	1000	20.93	405			
7	13 March	1135	18.93	459			
8	13 March	1200	19.75	448	63	146	113
9	13 March	1230	15.64	548			
10	13 March	1300	12.16	668			
11	13 March	1330	10.85	698	110	232	222
12	13 March	1440	11.07	728			
13	13 March	1505	11.65	743	106	230	148
14	14 March	0900	31.83	82			
15	14 March	1305	30.80	92			
16	14 March	1840	30.50	111	25	60	35
17	15 March	0900	31.09	117			
18	15 March	0930	29.01	183			
19	15 March	0945	28.19	215			
20	15 March	1000	27.11	262	59	118	106
21	15 March	1015	25.07	358			
22	15 March	1030	24.48	384			
23	15 March	1045	23.18	450			
24	15 March	1110	22.81	472			
25	16 March	0900	31.64	100			
26	17 March	0615	27.76	258	38	90	161
27	17 March	1415	23.22	454			
28	17 March	1555	19.22	649	114	265	376
Riv-1	25 March		0.00	110			
Riv-2	25 March		0.00	131			

Ra data are in dpm/100 l.

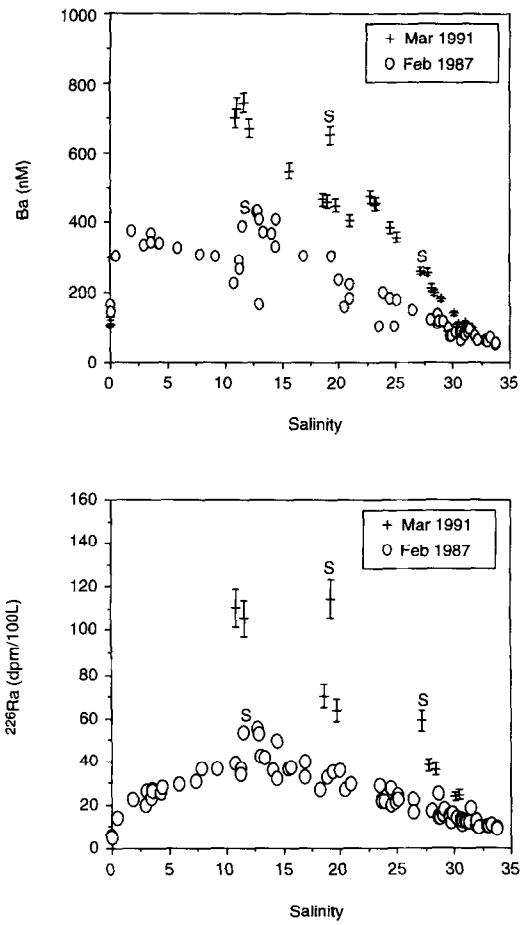

Fig. 2. Ba and ^{226}Ra concentrations obtained in March 1991 (crosses) compared with data reported by Carroll et al. [4] for samples collected in January–February 1987 (circles). Samples from the Sandwip Channel in the northeastern corner of the region are indicated with an 'S' near the data points on both graphs.

3. Results

Fig. 1 shows the 1991 sampling locations. Data for the 1991 samples are presented in Table 1. Fig. 2 compares the ^{226}Ra and Ba concentrations obtained in 1991 with data reported by Carroll et al. [4] for samples collected in January–February 1987. Samples from the Sandwip Channel in the northeastern corner of the region are indicated with an 'S' near the data points on each graph. Concentrations of Ba and ^{226}Ra are considerably greater in the 1991 samples compared to the 1987 samples at similar salinities (Fig. 2). The samples from Sandwip channel

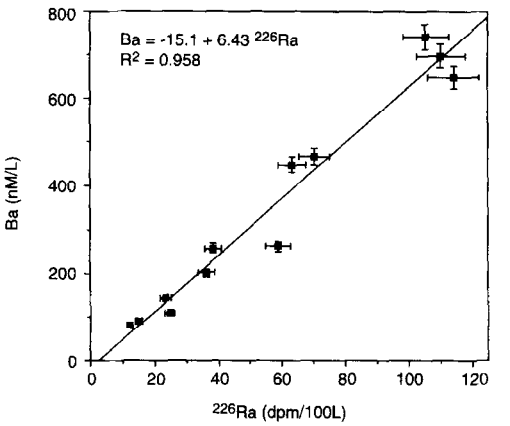

Fig. 3. Plots of Ba vs. ^{226}Ra for March 1991 samples revealing strong correlations. The Ba/^{226}Ra slope is 640 nM/dpm, the same as Carroll et al. [4] observed for the 1987 data. This suggests a similar source and behavior for each element during both studies.

from both studies have distinctly higher concentrations of Ba and ^{226}Ra compared to other samples at similar salinities.

Strong correlations occur between Ba and ^{226}Ra over the entire range of concentrations (Fig. 3), suggesting similar inputs and behavior for each. The Ba/^{226}Ra slope is 640 nM dpm^{-1}, exactly the same as Carroll et al. [4] observed for the 1987 data. Correlations also occur for ^{226}Ra and ^{228}Ra (Fig. 4). The high correlation ($R^2 = 0.992$) again suggests two

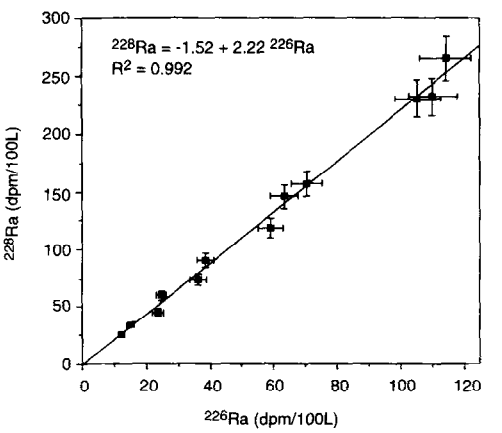

Fig. 4. Correlations between ^{226}Ra and ^{228}Ra for the March 1991 samples are also strong. The high correlation ($R^2 = 0.992$) suggests a similar source for each isotope and no decay of ^{228}Ra.

end-member mixing and shelf flushing rates that are too rapid to discern decay of ^{228}Ra.

The very high Ba concentrations measured in the mixing zone require comment. Falkner et al. [12] reviewed the solubility of pure barite in sea water and noted that the saturation concentration was exceeded by at least a factor of 2 in the deep waters of the Black Sea. In spite of the supersaturated conditions, barite falling through the water column appeared to dissolve in Black Sea deep waters, perhaps because the phase produced in the surface waters was not pure barite [12]. At a salinity of 18 ppt, 25°C, 1 bar, the Ba saturation concentration is 204 nM kg^{-1} [12]. Some samples reported here from the Bay of Bengal are supersaturated with respect to $BaSO_4$ by at least a factor of 3. Some of the 1987 samples were also supersaturated with respect to $BaSO_4$. Shaw et al. [7] have reported Ba concentrations in brackish groundwater that exceed $BaSO_4$ solubility by factors of 4–16.

4. Discussion

4.1. Strategy to determine the fluxes of Ra and Ba

The distribution of a chemical tracer in the coastal ocean depends on the flux of the tracer and the time available for it to accumulate. If the system is at steady state and the tracer is not removed except by mixing, the flux of the tracer offshore must be balanced by the flux to the coastal ocean. Using the distribution of Ra and Ba tracers on the shelf, along with an estimate of the residence time of shelf waters, I will calculate the offshore fluxes of Ra and Ba. This information will then be used to determine the fluxes necessary to sustain the measured signals and evaluate possible sources for these tracers. I will only consider the shelf inland of the 10 m isobath. The region under consideration is about 300 km long and extends offshore a distance of about 60 km. The total area is 18000 km^2, mean depth is 7 m and volume is 1.3×10^{11} m^3.

I realize that there are potential pitfalls to this approach. The system may not be at steady state. The estimated residence times may be incorrect. The measured tracer distribution may not be representative. In this analysis I will use a range of estimates of residence time of coastal waters to demonstrate that, in addition to river input, another large source of Ra and Ba is required to explain the distribution of these tracers on the Bengal shelf.

4.2. Riverine and coastal processes and coastal residence times

On an annual basis the Ganges–Brahmaputra River system ranks first in the supply of sediment to the ocean [13]. Most of the sediment is discharged during high river flow in June–September. At peak flow in August, the rivers contribute 86000 m^3 s^{-1} of water and 1.3×10^5 kg s^{-1} of sediment. This decreases to an average of 5800 m^3 s^{-1} of water and 1.5×10^3 kg s^{-1} of sediment during lowest flow in March [14]. During March 1991, the river discharge was 6500 m^3 s^{-1} and the sediment discharge was 1400 kg s^{-1}.

Most of the river flow during low discharge is through the main or Shahbazpur Channel between Bhola and Hatia Islands. The shelf in this eastern section of the delta is broad and shallow, averaging 7 m depth for a distance of 50–100 km from shore (Fig. 1). Beyond the 10 m isobath depths increase abruptly over most of the shelf. In the western section a deep submarine canyon, the Swatch of No Ground, is incised into the shelf.

The tidal range along the coast varies from 4–6 m in the Sandwip Channel to 2–3 m west of the river mouth. The high tidal range and strong currents effectively mix the water column on the shallow shelf during low river discharge. The maximum depth-averaged flood tide velocity in Sandwip Channel (west of Sandwip Island) may exceed 3 m s^{-1} during spring tides [15]. Tidal bores 1–1.5 m high often occur during spring tide in Sandwip Channel [16]. In Hatia Channel, between Hatia and Sandwip Islands, maximum tidal velocities occur during ebb tide [16]. Thus, tidal exchange effectively flushes water north through Sandwip Channel and south through Hatia Channel.

The height of the tide relative to the depth of the water may be used to estimate the time required to replace the water on the shelf. This tidal prism model assumes that each turn of the tide replaces a volume of water equivalent to the tide range and mixes it with offshore waters. With an average diurnal tide

range of 2–6 m, the replacement time for water shoreward of the 10 m isobath (average depth = 7 m) is only 1–2 days. This model is probably very effective in Sandwip and Hatia Channels, but less effective to the west because some of the water moved offshore during low tide is not mixed into the coastal ocean but returns during the next high tide. Therefore, predictions using this model are a minimum.

Another way to estimate coastal residence times is to use residual currents measured over a complete tidal cycle. For the period November 1989 to March 1991, Barua et al. [17] reported 23 measurements of residual currents in this study area. The highest currents, averaging 1–2 m s^{-1}, were measured in August 1990. During October–March, the residual currents ranged from 0.1 to 0.9 m s^{-1} with a mean of 0.38 m s^{-1}. During the 1991 study period, a residual current of 0.52 m s^{-1} to the south-southeast was measured near station 14 [17]. This is indicated by an arrow on Fig. 1. These residual currents provide another estimate of the residence time of water on the shelf. If the March 1991 measurement is representative of the current regimen over the shelf during the 1991 cruise, the entire volume of shelf water to a depth of 10 m could be replaced in 4–5 days.

The short lived ^{224}Ra also provides an indication of residence time. Fig. 5 is a plot of ^{224}Ra against ^{228}Ra for samples collected east of 90°E. The highest $^{224}Ra/^{228}Ra$ AR values are from the Sandwip Channel, where they exceed 1.4. The line on this graph is drawn with the assumption that the high activity Sandwip Channel sample is one end-member and the offshore samples are another. This assumption is consistent with the other data, which indicate that the Sandwip Channel has the highest Ra and Ba concentrations and represents a major source of Ra input to the system. The samples that fall below this mixing line do so by a factor of 2 or less with respect to predicted ^{224}Ra, suggesting that the residence time is of the order of one half live of ^{224}Ra, about 4 days. If sedimentary input of regenerated ^{224}Ra contributes significantly, this estimate will be too low.

A fourth way to estimate residence times of coastal waters is to use fresh water as a tracer and assume this fresh water only enters the system through river discharge. In this case the total volume of fresh

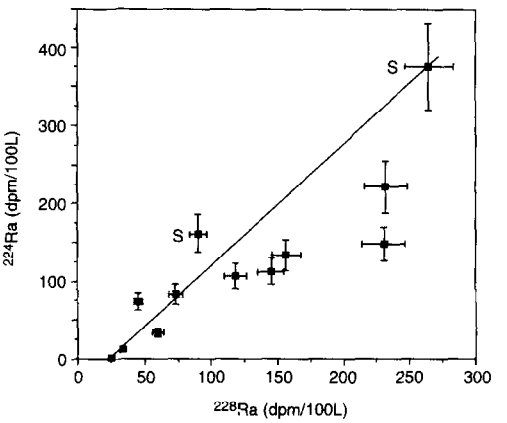

Fig. 5. A plot of ^{224}Ra vs. ^{228}Ra for March 1991 samples shows the highest $^{224}Ra/^{228}Ra$ AR values are from the Sandwip Channel, where they exceed 1.4. The line on this graph is drawn with the assumption that the high activity Sandwip sample is one end-member and the offshore samples are another.

water on the shelf coupled with the river discharge provides an estimate of the time available to accumulate fresh water. Carroll et al. [4] used this fresh water replacement model to estimate a residence time of 16 days for water near the river mouth in January–February 1987. Because this model assumes freshening is caused entirely by river discharge, any additional source of ungauged freshened water will cause this estimate to be too long. During the 1991 study we did not conduct a comprehensive survey that would be necessary to quantify the amount of fresh water on the shelf or even to contour the distribution. Nevertheless, we can place some limits on the range of salinity on the shelf and the corresponding replacement times. We have 20 measurements of salinity for water inshore of the 10 m isobath. The average of these measurements is 21 ppt. Considering the sampling density is greatest in the main channel, this figure probably underestimates the average salinity over the entire region. But, even eliminating the main channel samples, the remaining samples average 25 ppt. It is clear that the shallow water is significantly freshened. If we use an average salinity of 25 ppt for water inshore of the 10 m isobath and 31 ppt for offshore water, the residence time for the shallow water is 45 days. This estimate is an order of magnitude greater than the physical and radiochemical estimates. To bring the fresh water replacement estimate in line with the

Table 2
Estimates of residence times and Ra and Ba fluxes using different methods

Method	Residence time (d)	Ba flux (mol d^{-1})	^{226}Ra flux (dpm d^{-1})
Tidal prism	1–2	3×10^{7}	6×10^{13}
Residual current	4–5	1×10^{7}	2×10^{13}
^{224}Ra	4–5	1×10^{7}	2×10^{13}
Fresh water replacement	45	1×10^{6}	2×10^{12}

other estimates of 4–5 days, the average salinity for water inshore of the 10 m isobath would have to be within 1 ppt of the offshore water (i.e. 30 ppt). Clearly, there is a difference in the residence time estimates using the fresh water replacement model. Estimates of residence times using these different techniques are given in Table 2.

It is physically unreasonable to suggest that water can remain within the 10 m isobath for ninety 2–6 m tidal cycles when there are well documented residual currents that could remove it in a few days. There must be an additional source of fresh water augmenting the river flow. Nevertheless, estimates of Ra and Ba fluxes using the long residence time reach the same conclusion regarding Ra and Ba sources. For the following discussions I will use residence time estimates of 5 and 45 days to demonstrate that, even using the long residence time, the rivers cannot support the fluxes of Ba and Ra.

4.3. Riverine and oceanic supply of Ba and Ra

Dissolved concentrations of ^{226}Ra in the Ganges (10 dpm 100 l^{-1}) and Brahmaputra (7 dpm 100 l^{-1}) reported by Sarin et al. [18] for November 1983 are somewhat higher than measurements made by Carroll et al. [4] in January 1987 (5.3 dpm 100 l^{-1}). Concentrations of ^{226}Ra in Indian Ocean surface waters are 9–10 dpm 100 l^{-1} [19,20]. Carroll et al. [4] measured dissolved concentrations of Ba in the Ganges–Brahmaputra River of 147–167 nM l^{-1}. These results are slightly higher than results from March 1991, 110–131 nM l^{-1} (Table 1). Surface Indian Ocean Ba concentrations are 43–57 nM l^{-1} [20].

The March sediment flux of 1400 kg s^{-1} is expected to desorb 0.6 dpm ^{226}Ra g^{-1} and 250 nM Ba g^{-1} [4]. During the study period desorption from these sediments could support a ^{226}Ra flux of 7×10^{10} dpm d^{-1} and a Ba flux of 3×10^{4} mol d^{-1}. The total riverine flux (dissolved + desorbed) for ^{226}Ra is 1×10^{11} dpm d^{-1} and for Ba it is 1×10^{5} mol d^{-1}.

4.4. Offshore fluxes of Ba and ^{226}Ra

During low river discharge ^{226}Ra concentrations in the mixing zone increase rapidly and exceed by at least a factor of 5–10 the concentrations of the end-members (Fig. 2). Ba behaves in a similar manner, reaching concentrations 4–7 times greater than the river end-member (Fig. 2). Additional sources of ^{226}Ra and Ba are required to explain these nonconservative increases of both elements.

To estimate the total flux of ^{226}Ra and Ba which leaves the shelf, I will only consider the shelf inland of the 10 m isobath. The region under consideration has a volume of 1.3×10^{11} m^{3} which must be replaced every 5–45 days. The average ^{226}Ra concentration is 730 dpm m^{-3} and the average Ba concentration is 380 nM l^{-1}. Assuming the system is at steady state, a ^{226}Ra flux of $2–20\times10^{12}$ dpm d^{-1} and a Ba flux of $1–10\times10^{6}$ mol d^{-1} are required (Table 2). These calculated Ra and Ba fluxes exceed the total river fluxes by factors of 20–200 and 10–100, respectively.

4.5. Sources of Ba and ^{226}Ra

Carroll et al. [4] also demonstrated that only a small fraction of the measured concentrations of Ba and ^{226}Ra at low discharge could be produced by desorption from sediments delivered during or immediately preceding the study period. They suggested the high Ba and ^{226}Ra concentrations were due to the storage of sediments in fresh water sections of the river mouth during high discharge, followed by reworking and desorption of Ba and ^{226}Ra from these sediments during low discharge. This hypothesis is an extension of an idea formulated by Elsinger and Moore [3], who suggested that high ^{226}Ra concentrations in Winyah Bay during low river flow might be explained by up-river advancement of the salt front. Reworking of sediments deposited in

fresh water could release ^{226}Ra and temporarily enhance concentrations in the estuary. Carroll et al. [4] also suggested that salt water encroachment into the low-lying mangrove regions to the west and erosion of island sediments could further augment the supply of Ra and Ba to the shelf.

The 1991 data demonstrate that stored sediment is not an adequate source of Ba and ^{226}Ra. After peak flow in August, salt water quickly intrudes into the river mixing zone where waves and tides effectively mix the sediments. By mid March the sediments supplied during the previous peak discharge would have had 8 months of reworking to desorb Ba and ^{226}Ra. Yet the coastal water concentrations and offshore fluxes in March 1991 are even greater than in January–February 1987. To appreciate the magnitude of the March 1991 fluxes, compare them to the maximum expected fluxes during peak sediment discharge. This calculation assumes the sediments release 0.6 dpm ^{226}Ra g^{-1} and 250 nM Ba g^{-1} [4] and none of the sediments are stored in the system before desorbing Ra and Ba. The average peak sediment flux is 1.3×10^5 kg s^{-1} [13]. If ^{226}Ra immediately desorbs, the desorbed flux should be 6.6×10^{12} dpm d^{-1}. The peak dissolved ^{226}Ra flux is 6×10^{11} dpm d^{-1}, giving a maximum river-supported flux of 7×10^{12} dpm d^{-1}. This is only 3.5 times greater than the estimated flux during low river discharge using the long residence time (Table 2). At peak discharge the desorbed Ba flux is expected to be 2.7×10^6 mol d^{-1} and the dissolved Ba flux is 1×10^6 mol d^{-1}. This yields a total Ba flux of 4×10^6 mol d^{-1}, 4 times greater than the estimated flux at low discharge (Table 2). If these high fluxes during low discharge are due to sediment storage, a substantial fraction of the sediment supplied during high discharge would have to be stored for months in fresh water regions to be available for desorption during March. This is physically unreasonable. Additionally, any such storage would reduce the total fluxes during high discharge and lead to the unlikely case of desorption fluxes during low discharge exceeding desorption fluxes during high discharge. I conclude that sediment storage cannot play a major role in supplying Ra and Ba during the waning months of low river discharge.

These calculations demonstrate conclusively that, during low discharge, the fluxes of Ra and Ba from the Ganges–Brahmaputra delta cannot be sustained by river input. The next most likely source that can supply the necessary flux is the discharge of submarine groundwater highly enriched in Ra and Ba. Although we do not have Ra and Ba analyses from groundwaters in the region, we know that salty groundwaters are commonly encountered in shallow wells (D. Barua, pers. commun., 1996). All of the salty (5–35 ppt) groundwaters we have measured in South Carolina are significantly enriched in Ra and Ba. This is probably the case in Bangladesh as well.

I hypothesize the following cycle of Ba and Ra in response to changes in river discharge: During high river discharge, coastal aquifers are recharged with fresh water. Under these conditions the distribution coefficient (K_d) for Ba and ^{226}Ra on the aquifer solid is high and Ra and Ba adsorb to solids (and possibly Fe and Mn oxide coatings). During low river discharge, the penetration of salt water into the coastal aquifer causes K_d to decrease and facilitates the desorption of Ba and Ra from particles in the aquifer. Salty groundwater transports the dissolved elements to the sea. These processes are modulated by the tidal cycle. Highest penetration of salt water into the aquifer should occur during low river flow on a spring tide. Thus, the outflow of salty groundwater during falling water on a spring tide should contain the highest concentrations of Ba and Ra. The March 1991 sample from Sandwip Channel (sample 28, Table 1) was recovered 3 h after high tide near the peak of a 6.1 m spring tide. This hypothesis predicts that submarine groundwater discharge (SGWD) is the primary source function of Ba and ^{226}Ra to the Bay of Bengal during low river discharge.

The ^{228}Ra data also support the idea that the sample from Sandwip Channel is representative of the source of ^{228}Ra and ^{226}Ra to the system. All of the 1991 data fall along a tight mixing line of ^{228}Ra versus ^{226}Ra (Fig. 4) with the Sandwip Channel sample anchoring one end-member position. The Ba and ^{226}Ra correlation (Fig. 3) also requires an end-member of constant ^{226}Ra/Ba and no preferential removal of either element. The simplest explanation for these data (both 1991 and 1987) is that almost all of the excess Ba, ^{226}Ra and ^{228}Ra at low river flow are supplied by discharge of salty submarine groundwater.

4.6. How much groundwater is required?

If we knew the average concentration of Ba and Ra in the groundwater, estimating the groundwater flux based on the Ba and Ra fluxes would be trivial. However, at present, there are no data on these elements in brackish groundwaters from the region. However, the Sandwip sample (#28) suggests that the emerging groundwater is highly enriched in Ba. In this sample, Ba is over 3 times the saturation value with respect to barite. This in itself is not too troubling; surface sea water is 5 times supersaturated with respect to calcite. But how high can the groundwater supersaturation go? Shaw et al. [7] report Ba concentrations of 1000–3300 nM in groundwaters having salinity in the range 4–34 ppt, factors of 4–16 times the saturation concentration. If we assume the groundwater can reach 25-fold supersaturation with respect to barite, 5000 nM Ba at 19 ppt salinity, the groundwater flux would have to be 2300–23000 $m^3\ s^{-1}$, or 0.35–3.5 times the river flow. There are several possibilities for producing such high Ba concentrations. The groundwater salinity may be less than 19 ppt. This would reduce the corresponding degree of supersaturation with respect to barite, but it would increase the amount of fresh water entering the system through SGWD. Alternatively, the groundwater could have a low sulfate/chloride ratio. This could result from extensive reduction of sulfate within the surfical aquifer or there could be leakage of deep connate water into the system. Kraemer and Kharaka [21] report Ba concentrations of 15 μM to 5 mM in deep geothermal aquifers of the U.S. Gulf Coast.

5. Conclusions

(1) During peak flow, the Ganges–Brahmaputra River system is expected to contribute more Ra and Ba to the ocean than any other river system. Remarkably, the fluxes of Ra and Ba during low river discharge are comparable to fluxes expected during peak river discharge.

(2) During low river discharge, fluxes of ^{226}Ra, ^{228}Ra and Ba to the northern Bay of Bengal are most likely controlled by the submarine discharge of groundwater highly enriched in Ra and Ba.

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