Geochemical cycling in the Hooghly estuary, India

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Abstract

U–Th decay series isotopes, δ¹⁸O and Si measurements in the river estuarine waters and sediments of the polluted Hooghly estuary as well as the surface waters of the Bay of Bengal, its high salinity end member, are reported. Dissolved Si indicates that there are probably two mixing regimes, dissolved U behaviour is nonconservative and δ¹⁸O behaves conservatively in the overall estuarine region. Isotopes of reactive elements, viz. ²³⁴Th and ²¹⁰Po, are removed from the estuarine waters in < 2 days and < 1 month, respectively, which is due to high suspended matter (30–301 mg l⁻¹). ²²⁸Ra and ²²⁶Ra are profusely released into the estuarine waters in the low to mid-salinity regions.

As expected, the opposite trend is observed in the case of estuarine sediments and suspended matter. Reactive isotopes of Th, ²¹⁰Pb and ²¹⁰Po are enriched, whereas Ra isotopes are depleted with respect to their parent nuclides in the estuarine sediments and suspended matter. ²³²Th/Al ratio appears well suited to study the distribution and mixing of the bed load sediments of the Ganga–Brahmaputra (G–B) and the Hooghly rivers with those from other rivers on the Bay of Bengal floor.

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Keywords: Hooghly estuary; U–Th series nuclides; δ¹⁸O; Suspended matter; Sediments

1. Introduction

The Bay of Bengal is characterized by deltas of seven large rivers, the largest being that of the Ganga–Brahmaputra (G–B) river systems, which join in Bangladesh to form the Meghna river. The Hooghly river, a smaller channel of the Ganga, flows through the state of West Bengal, India and enters into the Bay of Bengal ~ 100 km south of the metropolis of Calcutta. It is made more polluted by sewage of humans and cattle than by industrial wastes (Basu et al., 1970; Gopalakrishnan et al., 1973). Compositionally, surface water and sediments of the Hooghly river resemble those of the Ganga river before the latter enters Bangladesh and merges with the Brahmaputra river. During the non-monsoon (summer) season, the surface waters of all peninsular rivers have salinity ~ 28 near their mouths, whereas the Hooghly and the main G–B river channels remain relatively fresh (salinity < 15). The real seawater end member (salinity ~ 35) of the Hooghly and G–B estuaries is the southernmost region of the Bay of Bengal (~ 5°N).

The G–B river system is the largest supplier of suspended matter to the oceans, 1.7 × 10¹⁵ g dispersed in 1 × 10¹⁵ l of water (Milliman and Meade,
In view of the large suspended matter transport, one expects rapid removal of reactive elements from the water (Broecker and Peng, 1982) and release of large amount of Ra isotopes, $\sim 10^{15}$ dpm year$^{-1}$ of $^{226}$Ra as well as $^{228}$Ra (Carroll, 1990; Carroll et al., 1993). $^{228}$Ra has been used as a tracer for surface-water mixing in the Bay of Bengal (Rengarajan et al., 2002). Such a release can also be due to groundwater, (Moore, 1997) and Indian rivers during non-monsoon season have a significant component of subsurface waters (Sarin, 1983; Ray et al., 1984). U and Th series nuclides serve as excellent tracers for understanding the nature and time scales of geochemical cycling involving adsorption of reactive elements onto particles from water, release/desorption of passive elements from suspended matter and estuarine sediments, and mixing of water, within the deltaic region and beyond, in the Bay of Bengal. Here, we report results of measurements of Si, $\delta^{18}O$, U, Th and Ra isotopes, $^{210}$Pb and $^{210}$Po in the estuarine waters and sediments, and those from the Bay of Bengal.

2. Materials and methods

2.1. Geohydrology

The Hooghly river is a small branch of the Ganga, which originates downstream of the Farakka barrage, about 300 km north of Calcutta (Fig. 1). The Kumaun Himalaya on the north and the Vidhyans on the south bind the drainage basin of the Ganga. Alluvial plains characterize the lower reaches. The Hooghly flows mainly over alluvium between the Farakka barrage and the Bay of Bengal (Rao, 1975; Sarin, 1983; Biswas, 1986).

Both water and sediment samples were collected in the river–estuarine regions of Hooghly during

![Fig. 1. Location map of the sampling sites in the Hooghly estuary. H-13 and E-13 are water sampling stations of 1991, and 34G is the location of gravity core collected close to the mouth of Hooghly river.](image)
December 1991, between Kalna (north of Hgy-14, Fig. 1) and the mouth of the Hooghly. Downstream of Kalpi (Fig. 1), the Hooghly river branches off into two channels; the smaller one, known as Mooriganga, was sampled in this investigation. Even at the mouth of the Hooghly river, salinity was 14.9. The estuarine region, i.e. the Bay of Bengal, was sampled during 1991 and January–February of 1997 and 1999. It should be pointed out that all sampling of the Hooghly estuarine and the Bay of Bengal waters was done during the winter (December–February). Based on the studies hitherto conducted, it can be said that except for the monsoon (high flow during June–September) when the concentrations of elements and/or U–Th series isotopes are low, there is not much change in the chemical characteristics during the rest of the year (Sarin, 1983; Ray et al., 1984).

2.2. Sampling and processing

During the low discharge season, water samples were collected on board launches close to the mid-stream, in clean plastic buckets and transferred to precleaned polyethylene containers after rinsing. For particulate matter determination, surface waters were collected directly into 1-l plastic bottles. Separate 30-ml samples were collected for \( \delta^{18}O \) measurement. Twenty-litre samples were collected separately for U and \( ^{234} \)Th measurements. After acidification with 50% HNO\(_3\), spike-\( ^{233} \)U for U and \( ^{230} \)Th for \( ^{234} \)Th) and Fe-carrier additions, and rigorous stirring (to destroy the carbonate complex in the case of U), U and Th were scavenged by Fe(OH)\(_3\) precipitation and the precipitates were taken to Ahmedabad for further analysis. Ra was extracted from 20-l water by passing it through two columns of MnO\(_2\)-coated acrylan fibre (Moore, 1976). Chlorinity (Cl, \( \%_0 \)) was determined at the field station, to a precision of 3%, by AgNO\(_3\) titration from which salinity values were deduced using the Knudsen equation (Strickland and Parsons, 1972).

Salinity = 0.030 + 1.8050 × Cl (\( \%_0 \))

2.3. Analysis

For water samples, radiochemical separation and purification of \( ^{234} \)Th, U isotopes and \( ^{210} \)Po were done following the procedures of Sarin et al. (1992). \( ^{234} \)Th chemistry was completed within 2 weeks from the date of sample collection. For dissolved Ra, manganese, along with Ra from the fibre sample, was dissolved by taking it to near boiling in 2 M HCl. \( ^{226} \)Ra was measured from this solution by the Rn emanation method (Mathieu et al., 1988).

Si in waters was measured colorimetrically (Strickland and Parsons, 1972) to a precision of < ±5%. Suspended matter content was determined by drying the sample along with preweighed Millipore membrane filter at ~ 75 °C to constant weight.

In the case of sediment samples, U, Th isotopes and \( ^{210} \)Po were measured after radiochemistry, followed by alpha spectroscopy using Si-surface barrier detectors having sensitive area of 450 mm\(^2\), connected to a multichannel analyzer (Krishnaswami and Sarin, 1976; Sarin et al., 1992), whereas \( ^{210} \)Pb, \( ^{228} \)Ra and \( ^{226} \)Ra were measured by gamma spectroscopy using Canberra Model GCW 2523 coaxial, well-type, low-background intrinsic HPGe detector (Moore, 1984; Somayajulu et al., 1999).

For oxygen isotopes, 2 ml of water sample was equilibrated with CO\(_2\) in tank at 25 °C for 48 h (Epstein and Mayeda, 1953). The equilibrated CO\(_2\) was
extracted and the oxygen isotopic composition ($\delta^{18}$O) was measured using a VG Micromass 602D mass spectrometer with an overall precision of $\pm 0.2\%$.

### Table 1

Dissolved Si, suspended matter and $\delta^{18}$O in the Hooghly estuarine waters

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Salinity (µM)</th>
<th>Suspended matter (µM)</th>
<th>Si (µM)</th>
<th>$\delta^{18}$O (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hgy-15</td>
<td>0.053</td>
<td>92.9</td>
<td>224.6</td>
<td>−7.97</td>
</tr>
<tr>
<td>Hgy-14</td>
<td>0.055</td>
<td>301.0</td>
<td>217.5</td>
<td>−7.87</td>
</tr>
<tr>
<td>Hgy-1</td>
<td>0.055</td>
<td>33.5</td>
<td>234.3</td>
<td>−7.72</td>
</tr>
</tbody>
</table>

The values are reported relative to V-SMOW (Vienna Standard Mean Ocean Water; Gonfiantini, 1981).

### 3. Results

Salinity, suspended matter, Si and $\delta^{18}$O results from the Hooghly river–estuarine waters are given in Table 1. After making appropriate decay and growth corrections for radioisotope activities, measured chemical and radioisotopic composition of water and sediment samples are listed in Tables 2–4, and Activity ratios of radionuclides are denoted by [ ]. Mention should be made of the $^{234}$Th activity estimate. It was not possible to do the U–Th separation soon after $^{234}$Th coprecipitation with Fe(OH)$_3$ at site. As mentioned in Section 2.3, Th radiochemistry was done in the Ahmedabad Laboratory and the samples were beta-counted at regular intervals to check for $^{234}$Th decay. The total $^{234}$Th activities ranged from 0.10 to 0.29 dpm l$^{-1}$ of which 0.05–0.17 dpm l$^{-1}$ could be accounted for $^{234}$Th ingrowth due to $^{238}$U that was coprecipitated with Th isotopes during Fe(OH)$_3$ scavenging.

Only two estuarine samples, Hgy-7 and Hgy-11 (Table 2), were analyzed for $^{210}$Po. From the date of

### Table 2

U, Th, Ra and Po nuclides in the Hooghly estuarine waters

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Salinity (µM)</th>
<th>$^{238}$U (dpm l$^{-1}$)$^a$</th>
<th>$^{234}$Th (dpm l$^{-1}$)$^b$</th>
<th>$^{226}$Ra (dpm/100 l)$^c$</th>
<th>$^{210}$Po (dpm/100 l)$^d$</th>
<th>$^{234}$Th/$^{238}$U</th>
<th>$^{210}$Po/$^{226}$Ra</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hgy-15</td>
<td>0.053</td>
<td>−</td>
<td>−</td>
<td>−</td>
<td>−</td>
<td>−</td>
<td>−</td>
</tr>
<tr>
<td>Hgy-1</td>
<td>0.055</td>
<td>2.66 ± 0.08</td>
<td>0.12 ± 0.02</td>
<td>11.0 ± 0.5</td>
<td>−</td>
<td>0.05 ± 0.01</td>
<td>−</td>
</tr>
<tr>
<td>Hgy-3</td>
<td>0.062</td>
<td>2.69 ± 0.08</td>
<td>0.17 ± 0.02</td>
<td>12.2 ± 0.5</td>
<td>−</td>
<td>0.06 ± 0.01</td>
<td>−</td>
</tr>
<tr>
<td>Hgy-4</td>
<td>0.072</td>
<td>2.63 ± 0.07</td>
<td>0.28 ± 0.03</td>
<td>11.1 ± 0.5</td>
<td>−</td>
<td>0.11 ± 0.01</td>
<td>−</td>
</tr>
<tr>
<td>Hgy-5</td>
<td>1.655</td>
<td>2.38 ± 0.08</td>
<td>−</td>
<td>10.7 ± 0.5</td>
<td>−</td>
<td>−</td>
<td>−</td>
</tr>
<tr>
<td>Hgy-7</td>
<td>6.239</td>
<td>2.39 ± 0.09</td>
<td>0.10 ± 0.04</td>
<td>27.2 ± 1.1</td>
<td>0.82 ± 0.10$^d$</td>
<td>0.04 ± 0.02</td>
<td>0.03 ± 0.01</td>
</tr>
<tr>
<td>Hgy-8</td>
<td>7.469</td>
<td>1.93 ± 0.06</td>
<td>−</td>
<td>30.6 ± 1.3</td>
<td>−</td>
<td>−</td>
<td>−</td>
</tr>
<tr>
<td>Hgy-10</td>
<td>10.806</td>
<td>2.14 ± 0.06</td>
<td>0.17 ± 0.02</td>
<td>31.2 ± 1.2</td>
<td>−</td>
<td>0.08 ± 0.01</td>
<td>−</td>
</tr>
<tr>
<td>Hgy-11</td>
<td>13.026</td>
<td>2.08 ± 0.07</td>
<td>0.15 ± 0.02</td>
<td>32.3 ± 1.3</td>
<td>2.3 ± 0.1$^d$</td>
<td>0.07 ± 0.01</td>
<td>0.07 ± 0.01</td>
</tr>
<tr>
<td>Hgy-13</td>
<td>14.849</td>
<td>1.72 ± 0.04</td>
<td>−0.07 ± 0.03</td>
<td>26.7 ± 1.0</td>
<td>−</td>
<td>−0.06 ± 0.02</td>
<td>−</td>
</tr>
<tr>
<td>F 11/5$^e$</td>
<td>32.100</td>
<td>2.22 ± 0.06</td>
<td>1.71 ± 0.04</td>
<td>12.2 ± 0.5</td>
<td>3.7 ± 0.1</td>
<td>0.77 ± 0.04</td>
<td>0.3 ± 0.1</td>
</tr>
<tr>
<td>H 13/5$^e$</td>
<td>32.18</td>
<td>2.27 ± 0.06</td>
<td>1.69 ± 0.07</td>
<td>12.2 ± 0.5</td>
<td>3.7 ± 0.1</td>
<td>0.74 ± 0.04</td>
<td>0.3 ± 0.1</td>
</tr>
<tr>
<td>E 13/5$^e$</td>
<td>32.25</td>
<td>2.27 ± 0.06</td>
<td>1.62 ± 0.08</td>
<td>10.7 ± 0.7</td>
<td>3.7 ± 0.1</td>
<td>0.74 ± 0.04</td>
<td>0.3 ± 0.1</td>
</tr>
<tr>
<td>A 12/2$^e$</td>
<td>33.528</td>
<td>2.20 ± 0.06</td>
<td>−</td>
<td>3.7 ± 0.1</td>
<td>−</td>
<td>−0.21 ± 0.1</td>
<td>−</td>
</tr>
</tbody>
</table>

$^a$: Activity ratio.

$^b$: Activity ratio.

$^c$: Activity ratio.

$^d$: Activity ratio.

$^e$: Activity ratio.

$^f$: Activity ratio.

$^g$: Activity ratio.

$^h$: Activity ratio.

$^i$: Activity ratio.

$^j$: Activity ratio.

$^k$: Activity ratio.

$^l$: Activity ratio.

$^m$: Activity ratio.

$^n$: Activity ratio.

$^o$: Activity ratio.

$^p$: Activity ratio.

$^q$: Activity ratio.

$^r$: Activity ratio.

$^s$: Activity ratio.

$^t$: Activity ratio.

$^u$: Activity ratio.

$^v$: Activity ratio.

$^w$: Activity ratio.

$^x$: Activity ratio.

$^y$: Activity ratio.

$^z$: Activity ratio.

$^AA$: Activity ratio.

$^AB$: Activity ratio.

$^AC$: Activity ratio.

$^AD$: Activity ratio.

$^AE$: Activity ratio.

$^AF$: Activity ratio.

$^AG$: Activity ratio.

$^AH$: Activity ratio.

$^AI$: Activity ratio.

$^AJ$: Activity ratio.

$^AK$: Activity ratio.

$^AL$: Activity ratio.

$^AM$: Activity ratio.

$^AN$: Activity ratio.

$^AO$: Activity ratio.

$^AP$: Activity ratio.

$^AQ$: Activity ratio.

$^AR$: Activity ratio.

$^AS$: Activity ratio.

$^AT$: Activity ratio.

$^AU$: Activity ratio.

$^AV$: Activity ratio.

$^AW$: Activity ratio.

$^AX$: Activity ratio.

$^AY$: Activity ratio.

$^AZ$: Activity ratio.

$^BA$: Activity ratio.

$^BB$: Activity ratio.

$^BC$: Activity ratio.

$^BD$: Activity ratio.

$^BE$: Activity ratio.

$^BF$: Activity ratio.

$^BG$: Activity ratio.

$^BH$: Activity ratio.

$^BI$: Activity ratio.

$^BJ$: Activity ratio.

$^BK$: Activity ratio.

$^BL$: Activity ratio.

$^BM$: Activity ratio.

$^BN$: Activity ratio.

$^BO$: Activity ratio.

$^BP$: Activity ratio.

$^BQ$: Activity ratio.

$^BR$: Activity ratio.

$^BS$: Activity ratio.

$^BT$: Activity ratio.

$^BU$: Activity ratio.

$^BV$: Activity ratio.

$^BW$: Activity ratio.

$^BX$: Activity ratio.

$^BY$: Activity ratio.

$^BZ$: Activity ratio.
Table 3
U and Th series nuclide concentrations in the Hooghly estuarine sediments and suspended matter

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Salinity (%)</th>
<th>U-238 (dpm g⁻¹)</th>
<th>Th-230 (dpm g⁻¹)</th>
<th>Th-232 (dpm g⁻¹)</th>
<th>Ra-226 (dpm g⁻¹)</th>
<th>Pb-210 (dpm g⁻¹)</th>
<th>Th/Al (dpm mg⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.055</td>
<td>9.43</td>
<td>2.09 ± 0.06</td>
<td>3.32 ± 0.07</td>
<td>4.62 ± 0.23</td>
<td>2.35 ± 0.17</td>
<td>1.93 ± 0.42</td>
</tr>
<tr>
<td>14</td>
<td>0.055</td>
<td>6.50</td>
<td>1.76 ± 0.03</td>
<td>2.91 ± 0.15</td>
<td>4.85 ± 0.24</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>14P</td>
<td>0.055</td>
<td>7.43</td>
<td>1.16 ± 0.03</td>
<td>1.81 ± 0.07</td>
<td>2.92 ± 0.11</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>4</td>
<td>0.072</td>
<td>8.54</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>2.48 ± 0.22</td>
<td>1.54 ± 0.35</td>
</tr>
<tr>
<td>5</td>
<td>1.655</td>
<td>8.97</td>
<td>2.08 ± 0.06</td>
<td>4.94 ± 0.36</td>
<td>6.41 ± 0.47</td>
<td>2.53 ± 0.17</td>
<td>1.52 ± 0.37</td>
</tr>
<tr>
<td>5P</td>
<td>1.653</td>
<td>6.25</td>
<td>2.79 ± 0.09</td>
<td>4.43 ± 0.23</td>
<td>6.73 ± 0.33</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>8</td>
<td>7.467</td>
<td>7.56</td>
<td>1.76 ± 0.05</td>
<td>2.81 ± 0.16</td>
<td>4.47 ± 0.25</td>
<td>1.90 ± 0.17</td>
<td>2.45 ± 0.48</td>
</tr>
<tr>
<td>10</td>
<td>10.806</td>
<td>6.14</td>
<td>1.81 ± 0.05</td>
<td>3.77 ± 0.21</td>
<td>5.77 ± 0.32</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>12</td>
<td>14.578</td>
<td>9.44</td>
<td>1.90 ± 0.05</td>
<td>3.40 ± 0.12</td>
<td>4.95 ± 0.17</td>
<td>2.06 ± 0.17</td>
<td>2.34 ± 0.05</td>
</tr>
<tr>
<td>12R</td>
<td>14.578</td>
<td>9.44</td>
<td>1.83 ± 0.06</td>
<td>3.11 ± 0.16</td>
<td>4.57 ± 0.23</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>34c</td>
<td>–</td>
<td>4.02</td>
<td>1.0 ± 0.1</td>
<td>1.78 ± 0.06</td>
<td>3.41 ± 0.08</td>
<td>–</td>
<td>–</td>
</tr>
</tbody>
</table>

P—suspended matter; R—repeat measurement.

4. Discussion

We first discuss the behaviour of δ¹⁸O and Si in the Hooghly estuary followed by the radioisotope distributions.

4.1. δ¹⁸O

The δ¹⁸O values of the Hooghly estuarine waters are shown in Table 1 and plotted as a function of salinity in Fig. 2. Five samples collected from the Bay

Table 4
U, Th and Ra isotope activity ratios [%] in the Hooghly estuarine sediments and suspended matter

<table>
<thead>
<tr>
<th>Sample code</th>
<th>²³⁴U/²³⁸U [%]</th>
<th>²²⁸Ra/²³²Th [%]</th>
<th>²³⁰Th/²³⁴U [%]</th>
<th>²³⁰Th/²³²Th [%]</th>
<th>²²⁸Ra/²³⁰Th [%]</th>
<th>²¹⁰Pb/²²⁶Ra [%]</th>
<th>²²⁶Ra/²²²Ra [%]</th>
<th>²³²Th/²³⁸U [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.10 ± 0.03</td>
<td>0.42 ± 0.09</td>
<td>1.44 ± 0.08</td>
<td>0.72 ± 0.02</td>
<td>0.71 ± 0.06</td>
<td>1.45 ± 0.21</td>
<td>0.82 ± 0.18</td>
<td>2.21 ± 0.12</td>
</tr>
<tr>
<td>14</td>
<td>1.01 ± 0.02</td>
<td>–</td>
<td>1.64 ± 0.09</td>
<td>0.60 ± 0.01</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>14P</td>
<td>1.02 ± 0.03</td>
<td>–</td>
<td>1.51 ± 0.07</td>
<td>0.62 ± 0.02</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>4b</td>
<td>–</td>
<td>0.33 ± 0.08</td>
<td>1.44 ± 0.08</td>
<td>0.72 ± 0.02</td>
<td>0.75 ± 0.06</td>
<td>1.13 ± 0.20</td>
<td>0.62 ± 0.15</td>
<td>2.21 ± 0.12</td>
</tr>
<tr>
<td>5</td>
<td>1.06 ± 0.03</td>
<td>0.24 ± 0.06</td>
<td>2.20 ± 0.17</td>
<td>0.77 ± 0.02</td>
<td>0.51 ± 0.05</td>
<td>1.25 ± 0.19</td>
<td>0.60 ± 0.15</td>
<td>3.08 ± 0.24</td>
</tr>
<tr>
<td>5P</td>
<td>1.01 ± 0.03</td>
<td>–</td>
<td>1.57 ± 0.10</td>
<td>0.66 ± 0.02</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>8</td>
<td>1.02 ± 0.02</td>
<td>0.55 ± 0.11</td>
<td>1.56 ± 0.11</td>
<td>0.63 ± 0.01</td>
<td>0.68 ± 0.09</td>
<td>1.12 ± 0.20</td>
<td>1.29 ± 0.27</td>
<td>2.54 ± 0.16</td>
</tr>
<tr>
<td>10</td>
<td>1.03 ± 0.02</td>
<td>–</td>
<td>2.02 ± 0.13</td>
<td>0.65 ± 0.01</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>12</td>
<td>1.04 ± 0.02</td>
<td>0.47 ± 0.04</td>
<td>1.72 ± 0.07</td>
<td>0.69 ± 0.01</td>
<td>0.61 ± 0.05</td>
<td>1.45 ± 0.24</td>
<td>1.14 ± 0.10</td>
<td>2.61 ± 0.11</td>
</tr>
<tr>
<td>12R</td>
<td>0.99 ± 0.03</td>
<td>–</td>
<td>1.70 ± 0.08</td>
<td>0.68 ± 0.02</td>
<td>–</td>
<td>–</td>
<td>–</td>
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</tr>
<tr>
<td>34c</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>0.52 ± 0.02</td>
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<td>3.51 ± 0.23</td>
</tr>
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P—suspended matter; R—repeat measurement.

a Activities of individual radionuclides are given in Table 3.
b Data of sample 1 are used, see Fig. 1.
c Sarin et al. (1979).
of Bengal in the high salinity end member of the Hooghly estuary during the same season are also included in the $\delta^{18}O$–salinity plot, which shows a linear relationship:

$$\delta^{18}O \text{ (‰)} = 0.227 \times \text{salinity} - 7.56 \quad (2)$$

with a correlation coefficient of 0.993, significant to 0.01 level. These data indicate that the waters of the Hooghly estuary display a perfect mixing behaviour with the seawater, with a freshwater end member of $\delta^{18}O = -7.5$‰ and a seawater end member of $\delta^{18}O = 0.4$‰ (at assumed salinity of 35). It is interesting to note that the Yellow river, which flows from the Chinese side of the Himalaya, has similar $\delta^{18}O$ in the freshwater end member, viz. $-8.0$‰ to $-7.0$‰ during both summer and winter months of 1986 (Zhang et al., 1990). In addition, the $\delta^{18}O$ vs. salinity plot has an almost identical relationship (Eq. (2)).

Martin and Letolle (1979) have studied a number of estuaries in France and Africa, including the Godavari estuary in South India. The slope of the $\delta^{18}O$–chlorinity relationship in their study ranged from 0.03 for Godavari to 0.25 for the Rhone estuary. Our value, 0.227 for the Hooghly estuary, falls in this range. It is interesting to compare the two Indian estuaries, Godavari and Hooghly. Godavari had a slope of 0.03 for samples collected in December 1978, i.e. winter season (Martin and Letolle, 1979) and a slope in the $\delta D$–chlorinity pair of 0.58 for samples collected in May–June 1981 (summer) (Sarin et al., 1985). The latter can be translated into a $\delta^{18}O$–salinity slope of 0.11 by using the relationship of $\delta D \text{ (‰)} = 8 \times \delta^{18}O \text{ (‰)} + 10$ for freshwaters derived from precipitation (Craig, 1961; Dansgard, 1964; Yurtsever and Gat, 1981). The difference in the slope between Godavari and Hooghly arises mainly because the freshwater end members in the two cases have different isotopic compositions. Whereas the Godavari receives its water mainly from the summer monsoon (June–September), the Hooghly receives water throughout the year from both monsoon rainfall and snowmelt from the Himalayan glaciers through the Ganga river. Ramesh and Sarin (1992) have reported $\delta^{18}O$ values for Ganga near Patna ranging from $-3.8$‰ in March (lean-flow season) to $-10.6$‰ in September (high-flow season). Similarly, they have reported $\delta D$ values of the Brahmaputra river at Goalpara ranging from $-37$‰ in April to $-65$‰ in December. These values are equivalent to $\delta^{18}O$ values of $-5.9$‰ to $-9.3$‰, respectively.

On the other hand, the freshwater end member for the Godavari has a value of $-2$‰ in December (winter) (Martin and Letolle, 1979) and $-2.8$‰ in May–June (summer) (Sarin et al., 1985). Assuming a salinity of 35 for the seawater end member, these values lead to an estimated $\delta^{18}O$–salinity slope of 0.07 for the Godavari and 0.29 for the Hooghly estuaries. Considering the analytical uncertainties and seasonal variations of the isotopic ratios in precipitation, these values are in reasonable agreement with the measured values of the slope. It is to be noted that while in the case of Godavari, the slope does not display any significant variation, in the case of the Hooghly, there can be a significant seasonal change in the slope from a value of 0.29 in winter (estimated) to 0.14 in summer. Further, the intensity of mixing is also a function of the discharge. During the monsoon months (June–September), the discharge in the Ganga is in the order of $1.2 \times 10^{15} l$, about seven times more than that during the winter months (November–February) (UNESCO, 1971). This has two effects: (i) the waters are transported so rapidly that there may not be complete mixing; and (ii) the freshwater regime is pushed more towards the sea, and as a consequence, the mixing zone is displaced towards the sea. If the measurements are repeated during the monsoon, by collecting the samples from the same locations, one might not see much variation in $\delta^{18}O$ vs. salinity. One may have to collect samples further towards the sea (southern Bay of Bengal) in order to obtain the mixing trend.
4.2. Suspended matter

The suspended matter contents of the Hooghly estuarine waters ranged from 13 to 301 mg l\(^{-1}\) (Table 1), its variation as a function of salinity is shown in Fig. 3. While sampling, it was visibly clear that the Hooghly waters looked muddy almost up to the mouth of the Mooriganga channel (Hgy-10, salinity = 10.8, Fig. 1 and Table 1) and the suspended matter contents were high, 33.5–301 mg l\(^{-1}\). Beyond Hgy-10, i.e. salinity >13.0, the particulates were low, 13–23.8 mg l\(^{-1}\). The suspended matter in the freshwater end member (≤ 0.075) varies by about a factor of 6 (49.6–301 mg l\(^{-1}\), Table 1). The highest particulate contents were encountered at Outramghat (Hgy-14), where a lot of human activity by way of boat and launch movements and washing on the banks were most prevalent. Despite these activities, the particulate contents of the Hooghly estuarine waters are comparable to the values reported for Meghna estuary during the month of February, viz. 200 ± 20 mg l\(^{-1}\) (Barua, 1990) and also for those of Mahanadi and Godavari during the non-monsoon periods (Ray et al., 1984; Sarin et al., 1985). It should be noted here that December being a non-monsoon month, wherein the suspended matter, like in all Indian rivers, is much lower compared to the monsoon months, viz. July–October, when the values can be in the range of several g l\(^{-1}\) especially during peak floods. In the case of G–B estuarine waters, the suspended matter concentrations during monsoon periods reach 1.4–1.6 g l\(^{-1}\) (Barua, 1990).

4.3. Silicon

The dissolved silicon contents of the Hooghly estuarine waters ranged from 18.6 to 234.3 μM, its variation as a function of salinity is shown in Fig. 3. Though no data are available beyond the mouth of the Hooghly, which has a salinity of 14.85, the open-ocean Bay of Bengal waters at salinity of 35 are taken to have relatively small Si concentrations (Wyrtki, 1971), and the seawater value is joined by a straight line with the mean Si value of the freshwaters. The Hooghly freshwaters (salinity < 0.08) are slightly enriched (by ~ 20%) in Si over the Ganga waters collected in 1982 upstream of the Farakka barrage, i.e. near Patna (Sarin et al., 1983). If the freshwater end member Si concentration is joined by straight line with the near-negligible seawater end member value (Fig. 4a; Table 1), it appears that Si behaves nonconservatively. In all Indian estuaries (excluding the Ganga–Brahmaputra estuary, Carroll, 1990), mean seawater salinities exist at the mouths during non-monsoon seasons. In the present case, Si behaves nearly conservatively between the freshwater end member and the river mouth (Hgy-13, Fig. 1). There appears to be a different mixing trend between Hgy-13 and the open Bay of Bengal waters. Due to absence of data between salinities 14.8 and 32.1 (Table 1), it is not possible to resolve this problem with the present data set.

4.4. U–Th decay series nuclides

The U-isotope distribution in the Hooghly estuarine waters was discussed by Somayajulu (1994), who noted that it behaved nonconservatively and estimated U removal to an extent of ~ 25%. Fig. 4b shows the distribution of U against salinity. Between the fresh-
water end member and river mouth, $^{238}$U behaves fairly conservatively. At salinity = 14.85, the $^{238}$U concentration is 1.72 dpm l$^{-1}$, which is lower than either the freshwater value or that of seawater end member. Considering the observation of Carroll and Moore (1994) in the Ganga–Brahmaputra mixing zone that U is being removed, it is reasonable to argue for U removal in the estuarine region of Hooghly, too (beyond its mouth). U and Si trends are similar in the Hooghly as borne out from the U vs. Si plot (Fig 4c). The U/Si ratio near the river mouth, as is seen for U alone, is lower than either end members. It is of considerable interest to study the behaviour of its daughters. The U content of sediments in the mid-estuary varied from 1.76 to 2.09 dpm g$^{-1}$ (Table 3), which is about a factor of 2 higher than the sediment $\sim$ 100 km away from the Hooghly mouth.

4.4.1. $^{234}$Th

One of the most particle-reactive elements in the ocean is Th. Based on the $[^{234}$Th/$^{238}$U] in surface layers which range from 0.4 to 0.8, its scavenging residence time is evaluated to be in the range of $\sim$ 30–100 days (Bhat et al., 1969; Sarin et al., 1994). In the shallow waters of the New York Bight (depth < 100 m) with suspended matter concentrations ranging from $\sim$ 60 to 600 mg l$^{-1}$, the removal rates of $^{234}$Th and $^{228}$Th ($t_{234}$ and $t_{228}$, respectively) are found to range from $\sim$ 10 days in nearshore waters to $\sim$ 70 days at the shelf-break (Kaufman et al., 1981). All used the relation:

$$t_{234} = \left[ \frac{R}{1 - R} \right] \tau_{234} \tag{3}$$

where $R$ = $[^{234}$Th/$^{238}$U] and $\tau_{234}$ is the mean life of $^{234}$Th = 34.8 days.

In the Hooghly estuarine region where $R$ ranged from $-0.06 \pm 0.02$ to $0.11 \pm 0.02$ averaging 0.04 ± 0.01, $t_{234}$ is $< 2$ days. In the high salinity end member, viz. the Bay of Bengal, $R$ is 0.75 (Table 2) which yields $t_{234}$ of $\sim$ 100 days (Sarin et al., 1994). The increase in $t_{234}$ is clearly due to the decreasing particulate content of the Bay of Bengal surface waters by more than two orders of magnitude.

4.4.2. $^{226}$Ra and $^{228}$Ra

Radium is an element known for its desorption from sediments, especially in the salinity region $\sim$ 2–20 (Elsigner and Moore, 1984; Moore and Scott, 1986; Carroll et al., 1993). This is also evident from the Hooghly estuary Ra data. $^{226}$Ra concentration increased from $\sim$ 10 dpm/100 l, at salinity = 0.06, to $\sim$ 30 dpm/100 l, at salinity = 15, and then decreased $\sim$ 10 dpm/100 l in the seawater end member. Similarly, $^{228}$Ra concentration increased from 18.5 dpm/100 l, at salinity = 0.06, to 120 dpm/100 l, at salinity = 15, and then decreased 10 dpm/100 l in the seawater end member (Table 2; Fig. 5a). Within 100 km off the Hooghly river mouth, the $^{228}$Ra concentrations come down from 120 to $\sim$ 20 dpm/100 l and decrease to $\sim$ 10 dpm/100 l in the southern Bay of Bengal (salinity $\sim$ 34) in a distance of $\sim$ 1300 km. Since Ra remains in solution, one can use $^{228}$Ra (half-life = 5.75 years) to determine rapid...
horizontal mixing rates in surface waters (Rengarajan et al., 2002).

4.4.3. $^{210}$Po

Only two estuarine samples were measured for $^{210}$Po (Table 2). Unfortunately, no $^{210}$Pb measurements could be made. The $^{210}$Po and $[^{210}$Po/$^{226}$Ra] data are plotted in Fig. 5b. It is seen that $^{210}$Po concentration is 0.81 dpm/100 kg at a salinity = 6.23 and increases to 2.3 dpm/100 kg at salinity = 13.03 and 3.7 dpm/100 kg at salinity = 32.2; the $[^{210}$Po/$^{226}$Ra] correspondingly increases with salinity, from 0.03 to 0.07 and 0.3 (Table 2). The high salinity data are from Sarin et al. (1994), who measured $^{210}$Po, $^{210}$Pb and $^{226}$Ra, and estimated that $[^{210}$Po/$^{210}$Pb] in surface waters of the Bay of Bengal is ~ 0.3 and that the scavenging residence time for $^{210}$Po is ~ 140 days. With the Hooghly estuarine waters having a concentration lower by a factor ~ 4, the $^{210}$Po removal time, $t_{^{210}}$, turns out to be in the order of ~ 30 days. This is a lower limit because the correction for $^{210}$Pb ingrowth will be smaller as the $^{210}$Pb
concentration in estuarine surface waters is expected to be lower than that of its oceanic counterparts.

4.5. U–Th decay series nuclides in estuarine suspended matter and sediments

The Hooghly sediments and suspended matter are enriched in both $^{232}\text{Th}$ and $^{238}\text{U}$. The Th/U weight ratio is $8 \pm 3$ (Fig. 6), which is about a factor 2 higher than the average value of shale (Rogers and Adams, 1969). In fact, the sediments of the Ganga drainage basin before the inception of the Hooghly are also enriched in Th and U (Sarin et al., 1990). Since the $^{238}\text{U}$ concentrations of the river and estuarine waters of the Ganga have been high, any $^{230}\text{Th}$ produced during its transit and residence in reservoirs, etc. would be effectively removed to sediments. As a result, one expects the Ganga and the Hooghly River sediments to be enriched in $^{230}\text{Th}$. This is borne out by the data in Tables 3 and 4; there is excess $^{230}\text{Th}$. Though it is estimated that $\sim 25\%$ U is removed from estuarine waters, it is difficult to precisely calculate $^{230}\text{Th}_{\text{excess}}$ in the underlying sediments. As a gross approximation, we subtract $^{234}\text{U}$ activity (dpm g$^{-1}$) from the total $^{230}\text{Th}$ activity measured (dpm g$^{-1}$). This is a lower limit for the $^{230}\text{Th}_{\text{excess}}$, which ranges from 0.6 to 2.7 dpm g$^{-1}$ with the $[^{230}\text{Th}_{\text{excess}}/^{232}\text{Th}]$ ranging from $\sim 0.2$ to 0.4, except in the high salinity region where it goes up to $\sim 1.5$ (Fig. 6).

![Fig. 6. $[^{230}\text{Th}_{\text{excess}}/^{232}\text{Th}]$ and Th/U wt. ratio in the estuarine sediments as a function of salinity. Whereas Th/U wt. ratio is uniform (average $= 8 \pm 3$), the $[^{230}\text{Th}_{\text{excess}}/^{232}\text{Th}]$ increases in the high salinity region of the Bay of Bengal. Note that $^{230}\text{Th}_{\text{excess}}$ is a lower limit. See text for discussion.](image)

It is clearly seen from Table 4 and Fig. 6 and taking into account the behaviour exhibited by Th, U, Ra and Po in the Hooghly estuarine waters that reactive elements Th, Po and Pb are enriched over their passive (and some, like Ra, leachable in saline solutions) parents. The leachable ones are depleted over their particle-reactive parents. Examples are $[^{230}\text{Th}/^{234}\text{U}]$ and $[^{210}\text{Pb}/^{226}\text{Ra}]$ of the Hooghly estuarine sediments and suspended matter which are $> 1.0$, whereas $[^{228}\text{Ra}/^{230}\text{Th}]$ is $< 1.0$ (Fig. 7). Similarly, the $[^{228}\text{Ra}/^{232}\text{Th}]$ is $< 1.0$ and $[^{228}\text{Ra}/^{226}\text{Ra}]$ ranged from 0.60 to $\sim 1.0$. The $[^{232}\text{Th}/^{238}\text{U}]$ ranged from 2.2 to 3.5, whereas the $[^{228}\text{Ra}/^{232}\text{Th}]/[^{232}\text{Th}/^{238}\text{U}]$ is $< 0.5$ (Fig. 8). This implies that $^{228}\text{Ra}$ and $^{226}\text{Ra}$ are leached.
out of sediments instead of being in equilibrium with their parents. The samples collected from the islands in G–B estuary have $\frac{^{228}\text{Ra}}{^{226}\text{Ra}}$ of 1.8 ± 0.1 and $\frac{^{228}\text{Ra}}{^{226}\text{Ra}}/\frac{^{232}\text{Th}}{^{238}\text{U}}$, close to equilibrium at 0.86 ± 0.10 (Fig. 8, Carroll, 1990).

The $^{232}\text{Th}/\text{Al}$ ratios in the Hooghly estuarine sediments and particulates range from 0.039 to 0.107, averaging 0.066 dpm mg$^{-1}$ (Table 3). The G–B estuarine sediments (Carroll, 1990; Galy, 1999) and those from Ganga river sediments (Sarin, 1983) yielded similar values. The $^{232}\text{Th}/\text{Al}$ data from the sediment trap collections in the Bay of Bengal, even in the northernmost trap at 15°N, have all yielded $^{232}\text{Th}/\text{Al}$ values (mean) < 0.04 dpm mg$^{-1}$ (Sarin et al., 2000). This can be due to (i) contribution/dilution from the suspended lithogenic material from the peninsular rivers of India (Sarin et al., 2000) and Irrawaddy from Burma, and (ii) that a significant fraction of the G–B river system discharged suspended sediment is slumping down the region of entrance and is not suspended in the water. The only sediment core close to the Hooghly river mouth that is analyzed for $^{232}\text{Th}$ and Al has $^{232}\text{Th}/\text{Al}$ ratio of 0.085 (Sarin et al., 1979). Other analyzed cores are far away from the G–B and have contributions from the peninsular rivers, and hence, the $^{232}\text{Th}/\text{Al}$ ratios are lower ranging from 0.023 to 0.055. Th/Al ratio appears well suited to study G–B (bed load) sediment distribution on the Bay of Bengal floor.

### 4.6. $^{228}\text{Ra}/^{232}\text{Th}$ in estuarine and island sediments

From Figs. 7b and 8b, as well as data in Table 4, it is clear that $\frac{^{228}\text{Ra}}{^{232}\text{Th}}$ and $\frac{^{226}\text{Ra}}{^{238}\text{U}}$ in the Hooghly estuarine sediments range from 0.24 to 0.55 (mean = 0.40) and 0.51 to 0.75 (mean = 0.65), respectively. The sediment samples from the islands in the estuarine regions however, have both the activity ratios at unity (Carroll, 1990), indicating that the $^{228}\text{Ra}$ (half-life = 5.75 years) and $^{226}\text{Ra}$ (half-life = 1600 years) have reached secular equilibrium with their respective parents, viz. $^{232}\text{Th}$ and $^{230}\text{Th}$.

### 5. Conclusions

Si appears to indicate two mixing regimes in the Hooghly estuarine waters, whereas U behaves non-conservatively and 25% U are removed in the estuarine region. $\delta^{18}\text{O}$ behaves conservatively.

In the estuarine waters with higher (13–30 l mg$^{-1}$) particulate concentrations compared to the open Bay of Bengal surface waters, reactive elements like $^{234}\text{Th}$ and $^{210}\text{Po}$ are removed in a matter of < 2 days and < 1 month, respectively. This is supported by the enrichment of Th isotopes and $^{210}\text{Po}$ (over their parental radioactivities) in estuarine sediments.

Ra isotopes are leached in the mid-salinity regions and $^{228}\text{Ra}$ is useful for studying mixing in the Bay of Bengal surface waters, between the G–B and Hooghly river mouths and the seawater end member of the southern Bay. $^{228}\text{Ra}$ and $^{226}\text{Ra}$ are depleted (relative to their parents, $^{232}\text{Th}$ and $^{230}\text{Th}$) in estuarine sediments and particulate matter.
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