

# ISOTOPES OF URANIUM AND RADIUM IN INDIAN RIVERS

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## ABSTRACT

The concentrations of dissolved uranium and radium isotopes have been determined in the waters of several Indian rivers. The concentration of uranium (U-238) is found to vary between 0.01–7.0 micrograms/litre, depending primarily on the terrain through which the river flows. From the available data on the activity ratios, U-234/U-238, concentration of U-238 in rivers and the amount of uranium depositing on the ocean floor, a material balance calculation has been attempted. This shows that the supply of uranium to sea by rivers based on above measurements, is inadequate to explain the observed concentrations of U-238 and U-234 in the oceans.

The Ra-226 concentration of river waters is found to be of the order of 0.1 dpm/litre (varying between 0.05–0.2 dpm/litre). The activity ratios, Ra-228/Ra-226, range between 1 and 4. This large variation is clearly related to the fact that the concentrations of the parent nuclides of Ra-226 and Ra-228 are different. The observed ratios are consistent with those expected from leaching of clays/soils; however, analogous to the situation in the case of uranium, the observed concentrations of radium isotopes in the oceans are too large to be explained by their influx by rivers.

## 1. INTRODUCTION

It is well known that the significant radioactive disequilibria between the members of the three naturally occurring radioactive series, U-238, U-235 and Th-232 in the hydrosphere are related to their grossly different geochemical behaviours. The first noteworthy observation on disequilibrium between two isotopes in the same series was reported by Russian workers<sup>1</sup> who found that the concentration of U-234 in ground waters was in excess compared to that of its parent, U-238. Similar disequi-

tibrium between U-234 and U-238 has been reported in sea and river waters by several workers.<sup>2-7</sup>

During the last few years, more examples of disequilibria have been observed in natural waters.<sup>8-11</sup> Moore and Sackett<sup>8</sup> have shown that the Th-228/Th-232 activity ratios in sea-water are an order of magnitude higher than the expected secular equilibrium value. This result was later corroborated by the studies of Somayajulu *et al.*<sup>9</sup> Measurements of the two radium isotopes Ra-226 and Ra-228<sup>12-14</sup> in sea and river waters also show that they are not in equilibrium with their parents, Th-230 and Th-232 respectively.

The above studies are largely confined to the marine environment and data on the concentrations of uranium and radium isotopes in rivers are scanty.<sup>4, 15, 16</sup> Rivers are the primary agency for the introduction of a host of elements into the sea; eolian transportation of dust and subsequent leaching of trace elements from the dust during its transit through sea-water probably plays an important role in the input of certain elements.<sup>17</sup>

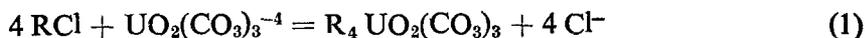
In this paper we report our measurements of the concentrations of U-238, U-234, Ra-228 and Ra-226 in some major Indian river waters. Seasonal variations in the concentrations of uranium have also been studied. A few ground water samples have been analysed for uranium in order to explore the possibilities of determining the extent of effluent seepage into the river waters.

## 2. EXPERIMENTAL METHODS

Uranium, which exists mainly as an anion complex in natural waters,<sup>18</sup> is usually pre-concentrated from large volumes of water by various methods, the most common being the ferric-hydroxide scavenging. To get a quantitative yield of uranium from water samples by the scavenging method, it is necessary to destroy the anion complex before scavenging and hence the use of an *in situ* scavenging method is limited. If, however, one uses an anion exchange resin to pick up the uranium complex directly, it can serve as a better *in situ* extraction method. Based on studies of the uptake of sulphate, phosphate, and carbonate complexes of uranium on anion exchange resins, some methods have been developed to extract uranium from leachates of ores and natural waters of high uranium content.<sup>19-21</sup>

In this work pre-concentration of uranium from large volumes of water at the sampling site was done using an anion exchange resin, after converting the uranium to a stable  $\text{UO}_2(\text{CO}_3)_3^{-4}$  complex by addition of suitable

amounts of ammonium carbonate to the water. The uptake mechanism probably is:



Water samples of 400–600 litres were collected in polyethylene drums from the mid-stream regions of the rivers. (For locations of samples, see Fig. 1.) The suspended matter was first removed by allowing it to settle for a period of about 10–12 hours (this step removes all particles of greater than about five micron size). To the clear decanted water, analar grade ammonium carbonate was added to make a concentration of about 0.08 M of the reagent. U-232 was added as a spike and the water was passed through an anion exchange resin (Dowex-1; 50–100 mesh; column volume, ca. 500 c.c.), in carbonate medium at a rate of about 20–30 column volumes per hour. The resin was later brought to the laboratory and the mud (if any) was separated by suspending the resin in distilled water. Uranium was eluted from the resin with 4–5 column volumes of 3N ammonium chloride, followed by 2–3 column volumes of hydrochloric acid at pH ~ 4. The ammonium salts were decomposed and the uranium was purified by the procedure of Bhat *et al.*<sup>11</sup>

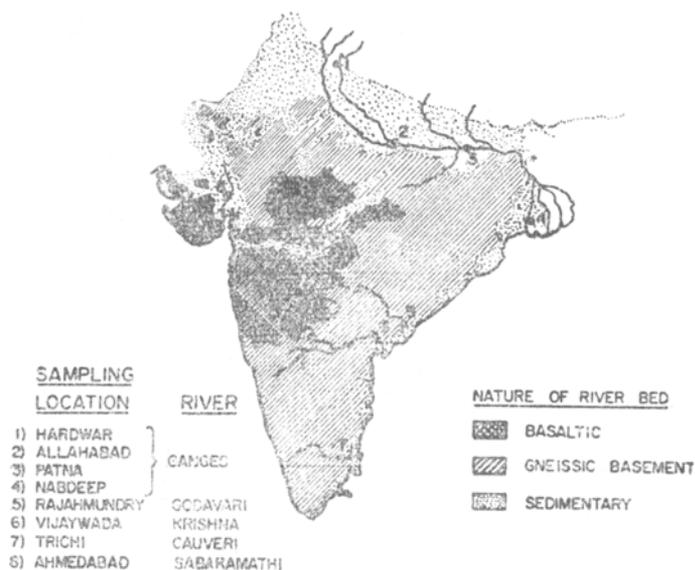


FIG. 1. Map of India showing the major petrological divisions and locations from where the river water samples were collected.

For studying the seasonal variations of uranium content and U-234/U-238 activity ratios, 20–50 litre samples were processed, using smaller amounts of resin.

After purification, an aliquot (less than 1%) of the sample was directly deposited on a platinum planchet, and the total alpha activity was measured using a zinc sulphide detector system.<sup>22</sup> Depending on the total alpha activity a suitable fraction of the sample was electroplated on a one inch diameter, 1 mil. thick, platinum disc.

The alpha activities of uranium isotopes were measured on a 128 channel alpha spectrometer employing a one inch diameter Nuclear Diode surface barrier detector.

The efficiency of pick-up of uranium, as determined by laboratory experiments using U-232 tracer was about 70–80%.

The column method used by us has the advantage that it provides a fast and efficient *in situ* pre-concentration step for uranium from large volumes of water. However this procedure has two limitations:

(1) It is applicable only if the uranium isotopes in the waters are present in the +6 valency state, only when the carbonate complex is formed. To check on this point, we evaporated three 5 liters mud free water samples to dryness, and processed them for uranium. The results are shown in Table I. The results are in good agreement, within statistical errors which are about 10 per cent.

TABLE I

*Concentrations of U-238 and U-234/U-238 activity ratios as determined by the column and evaporation methods*

Sample	Column method		Evaporation method	
	U-238 concentration (ppb)	U-234/U-238 (activity ratio)	U-238 concentration (ppb)	U-234/U-238 (activity ratio)
Ganges (Hardwar) ..	1.9±0.2	1.03±.03	1.8±0.1	1.08±.06
Ganges (Allahabad) ..	6.6±0.8	1.04±.02	6.9±0.4	1.09±.04
Sabarmati (Ahmedabad) ..	4.2±0.2	1.57±.04	3.6±0.1	1.51±.04

(2) The suspended matter should be separated from the water as efficiently as possible. As the gravitational settling of the particulate matter did not give a complete separation, the possibility of leaching uranium from mud by ammonium carbonate can arise. An estimate of the amount of leachable uranium was obtained by shaking about fifty grams of one of the river sediments with 0.1 M ammonium carbonate for about fifteen hours. It was found that the amount of uranium leached was 0.3 micrograms U-238/gm. of mud; the U-234/U-238 activity ratio was  $1.4 \pm 0.1$ . An upper limit on the contribution of uranium due to leaching of mud present in samples was estimated from the known particulate matter content of these river waters,<sup>22</sup> assuming a complete leaching of the normal uranium concentration—3 ppm. By this approach we estimate that not more than ten per cent of the total observed activity in the water can be due to leaching of uranium from the mud.

Thus we conclude that the measured uranium concentrations as well as U-234/U-238 activity ratios represent closely the actual values for dissolved uranium present in the waters.

Ra-226 was determined by measuring its daughter nuclide, Rn-222, essentially by the procedure described by Broecker,<sup>23</sup> from five litre water samples. We replaced the single U-tube used by Broecker by a coiled one, which traps the radon more efficiently. The trapped radon is released directly into the counter, which consists of a two-inch diameter stainless steel chamber coated inside with activated zinc sulphide. The method will be described in detail elsewhere.<sup>22</sup> The two short-lived daughter products of radon, *viz.*, Po-218 and Po-214 grow in the chamber and attain saturation value within about two hours. The scintillations produced by the alpha particles emitted by the three nuclides are recorded by photomultiplier assembly. The alpha counting yield for the above system, using a N.B.S. Ra-226 standard, was measured to be  $1.5 \pm 0.1$ . The background rates were about 3–4 counts per hour.

Blank runs with distilled water did not give any measurable activity above the background. The usual activity signals due to Ra-226 were in the region of 0.5–5 counts per minute.

The activity ratios, Ra-228/Ra-226, were determined from about 200–400 litres of water after the removal of suspended matter by gravitational settling. A preliminary concentration of radium isotopes was carried out at the sampling site by precipitating  $\text{CaCO}_3$ , which scavenges most of the radium.  $\text{CaCO}_3$  was dissolved in HCl, and Ra-226 concentration was first

determined by radon emanation method. The concentration of Ra-228 was obtained by milking its short-lived daughter Ac-228 (half-life = 6.1 hrs.) by Fe (OH)<sub>3</sub> scavenging. The beta activity of Ac-228 was assayed on a flat rectangular gas flow counter.<sup>24</sup> The activity levels of Ac-228 were about 5–10 counts per minute compared to background of 0.1–0.15 counts per minute of the counter.

The samples were remilked; duplicate analyses were in good agreement.

### 3. RESULTS

The uranium concentrations in micrograms/litre (ppb) and the U-234/U-238 activity ratios are given in Table II, along with the salinity (sum of the major dissolved constituents) data. Table III lists the variation in the uranium concentrations and U-234/U-238 activity ratios during different seasons for the three rivers Ganges, Cauvery and Sabarmati. The radium content (in dpm/litre) and the Ra-228/Ra-226 ratios are given in Table IV. (Since the activity levels of Ra-226 in five litre samples were low, the results are reliable only within  $\pm 15\%$ .)

TABLE II

*Concentrations of U-238 and activity ratios, U-234/U-238, in Indian Rivers*

River	Sample location	Date of collection	Nature of river bed	Total dissolved solids (ppm)	Uranium (ppb)	U-234/U-238 (activity ratio)
Ganges	.. Hardwar	Jan. 1968	Sedimentary	186	1.92 $\pm$ 0.2	1.03 $\pm$ 0.03
"	.. Allahabad	"	"	338	6.55 $\pm$ 0.80	1.04 $\pm$ 0.02
"	.. Patna	"	"	372	4.12 $\pm$ 0.50	1.12 $\pm$ 0.03
"	.. Nabdeep	"	"	373	1.55 $\pm$ 0.16	1.07 $\pm$ 0.03
Sabarmati	. Ahmedabad	Feb. 1968	"	470	3.50 $\pm$ 0.40	1.50 $\pm$ 0.03
Godavari	.. Rajahmundry	Oct. 1967	Basalts and gneissic basement	213	0.68 $\pm$ 0.04	1.35 $\pm$ 0.03
Krishna	.. Vijayawada	"	"	205	1.08 $\pm$ 0.12	1.58 $\pm$ 0.04
Cauvery	.. Trichi	"	Gneissic basement	310	0.58 $\pm$ 0.04	1.28 $\pm$ 0.03
Mutha	.. Khadakwasla	Sept. 1967	Basaltic	96	0.01 $\pm$ 0.001	1.44 $\pm$ 0.05
Ulhas	.. Ulhasnagar	"	"	66	0.01 $\pm$ 0.001	1.31 $\pm$ 0.12

## 4. DISCUSSIONS

(a) *Uranium*.—The uranium concentrations observed by us are in good agreement with the values reported earlier for Chambal river,<sup>25</sup> but are higher compared to many world rivers.<sup>4, 15</sup>

The river Ganges has been sampled at four stations along its course. This river originates in the Southern Himalayas and flows mainly through sedimentary rocks. Uranium concentration values are highest at Allahabad, 6.5 micrograms/litre, decreasing along its course to a value of about 1.5 micrograms/litre near the mouth of the river. This decrease in the concentrations could either be due to the dilution by the tributaries of Yamuna which meets Ganges at Allahabad. These tributaries (not shown in Fig. 1) flow mainly through basement complex. (Basement complex or gneissic basement consists of a mixture of igneous and sedimentary rocks.<sup>26</sup>) Another possible explanation involves the precipitation of uranium due to the presence of a reducing environment along its course. The available data are inadequate to choose between these alternatives. The low value at Nabdeep, near the mouth, cannot be explained in part as due to dilution by sea-water because of the observed low chloride content (8 ppm) of the water.

The three South Indian rivers, Cauvery, Godavari and Krishna which mainly flow through basement complex have nearly similar uranium concentrations, 0.6–1.0 ppb, considerably lower than northern rivers (Ganges and Sabarmati). The U-234/U-238 activity ratios of these rivers are however significantly higher compared to Ganges.

The two small rivers Ulhas and Mutha which flow through basaltic terrain (Deccan trap series), have the lowest uranium concentration. The U-234/U-238 activity ratios are higher compared to that of Ganges, but are similar to the ratios observed for the three South Indian rivers.

The river Sabarmati flows mainly through sedimentary deposits similar to that of Ganges. During summer, the water is fed mainly by effluent seepage from ground.<sup>27</sup> Both the uranium concentration and U-234/U-238 activity ratio are high (Table II). One ground water sample analysed from an area adjacent to the river was found to have a concentration of 10 micrograms U/litre with U-234/U-238 activity ratio of  $1.45 \pm 0.05$ . Therefore, it seems possible to delineate the extent of effluent seepage (dry discharge) from a study of U-238 concentration and U-234/U-238 activity ratios in ground waters and in river during different seasons, particularly in areas where these parameters are very different in the two cases.

From the data presented in Table II (*see* also Fig. 2), it is clear that rivers draining through sedimentary rocks are generally rich in uranium compared to the rivers flowing through igneous rocks, an observation similar to that reported earlier by Koczy.<sup>16</sup> Also, U-234/U-238 activity ratios in rivers flowing through igneous rocks are higher compared to those flowing through sedimentary rocks<sup>5</sup>—*i.e.*, an anti-correlation between uranium concentration and the activity ratios, U-234/U-238.

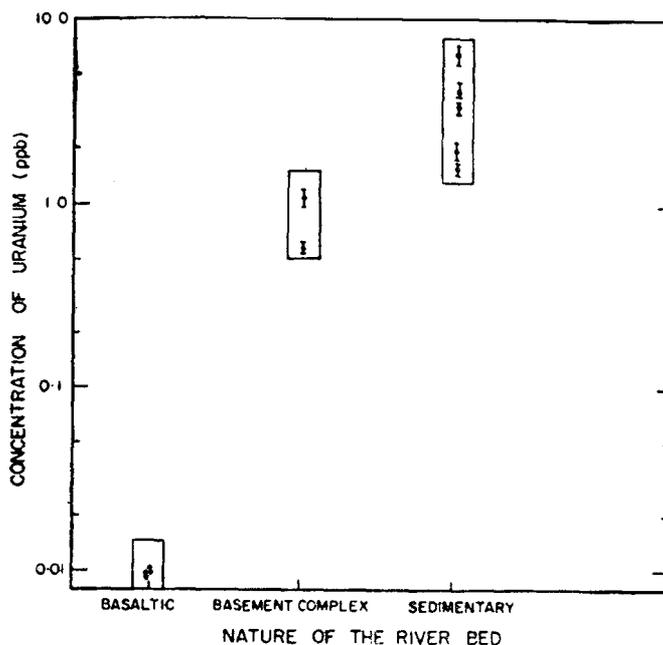


FIG. 2. Observed ranges in concentration of uranium in rivers and the nature of river bed.

In Fig. 3, we have plotted the uranium concentration values against the salinity (total dissolved salts) values from all the world rivers for which the data are available. A general positive correlation seems to be there which is not surprising since salinity values are an approximate measure of the weathering of the terrain through which the river flows.

Table III gives the measured concentrations of U-238 and activity ratios U-234/U-238, for three rivers during different seasons. There does not seem to be any marked variation in the U-234/U-238 activity ratios. The U-238 concentration however decreases significantly during the monsoon period.

From the available data on river discharge to sea,<sup>28</sup> we obtain a mean activity ratio  $U-234/U-238 = 1.2$  for the Indian rivers. This value is

TABLE III  
*Seasonal variation in the concentration of uranium and activity ratios  
 U-234/U-238*

River (location)	Date of collection	Uranium (ppb)	U-234/U-238 (activity ratio)
Sabarmati (Ahmedabad)	.. May 1967	2.5±0.40	1.45±0.07
	February 1968	3.5±0.40	1.49±0.03
	April 1968	4.2±0.20	1.57±0.04
	September 1968	2.4±0.12	1.58±0.04
	October 1968	3.0±0.11	1.51±0.02
	December 1968	3.8±0.11	1.47±0.04
Ganges (Allahabad)	.. May 1967	7.0±1.00	1.00±0.04
	January 1968	6.6±0.80	1.04±0.02
	July 1968	0.9±0.03	1.03±0.03
	October 1968	5.7±0.40	1.04±0.02
	November 1968	6.1±0.60	1.08±0.03
	January 1969	5.1±0.30	1.01±0.02
Cauvery (Trichi)	.. February 1967	0.7±0.09	1.30±0.03
	October 1967	0.6±0.04	1.28±0.03
	June 1968	0.3±0.01	1.31±0.04
	October 1968	0.2±0.01	1.29±0.09

close to the published values for principal world rivers, 1.2 for the Russian rivers,<sup>5</sup> 1.1 for Amazon, and 1.3 for Mississippi.<sup>4</sup> These rivers comprise about 25% of the world annual river discharge. We therefore estimate that the weighted mean activity ratio, U-234/U-235 for world rivers is about 1.2, with an upper limit of 1.3. As discussed below this result is not consistent with the observed activity ratio of U-234/U-238 in sea-water if one considers rivers to be the dominant source of uranium in ocean. In this case it can be shown that if steady state conditions exist in oceans for uranium:

$$T = \frac{A_r - A_o}{\lambda_4(A_o - 1) - A_r \lambda_3} \quad (2)$$

T is the residence time of uranium in sea-water,  $\lambda_3$  and  $\lambda_4$  are the disintegration constants of U-238 and U-234 respectively, A is the activity ratio of U-234/U-238 and subscripts *r* and *o* refer to river and ocean water

respectively. Two additional conditions imposed by the material balance of U-238 are

$$T = K_1 \left( \frac{C_o}{C_r} \right) \quad (3)$$

$$C_s = K_2 C_r \quad (4)$$

where  $C_r$  is the weighted average concentration U-238 per litre in river waters,  $C_o$  is the U-238 concentration in oceans,  $C_s$  is the concentration of authigenic U-238 in pelagic sediments (weight parts),  $S$  is the average

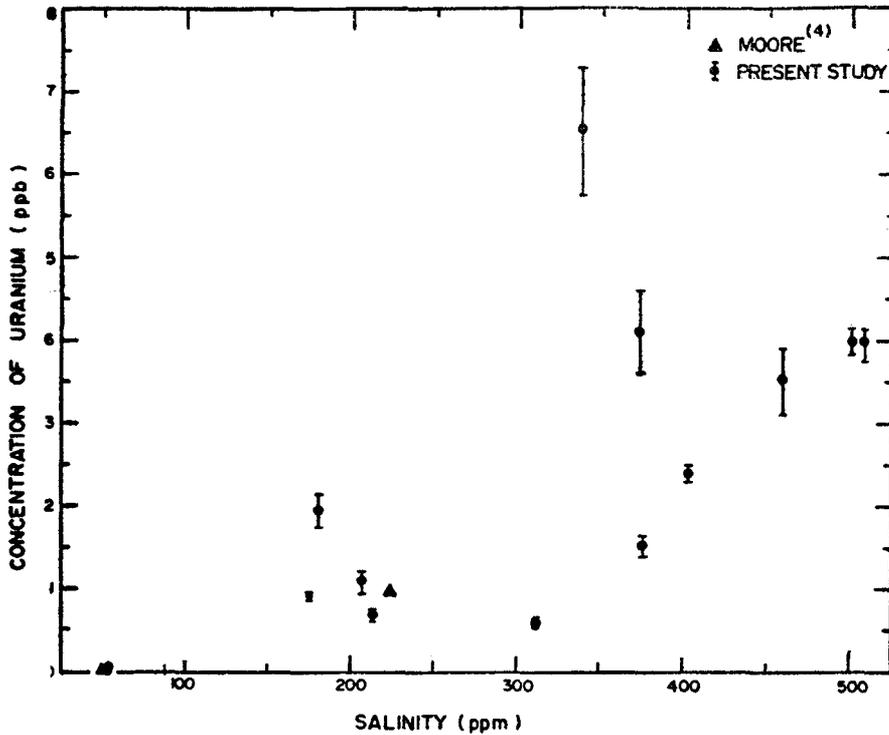


FIG. 3. Measured concentrations of uranium are plotted vs. salinity (total dissolved salt content). The salinity data for Amazon and Mississippi are taken from Livingstone,<sup>30</sup> those for Indian rivers are from present study.

sedimentation rate ( $\text{gm}/\text{cm}^2\text{-yr.}$ ). The value of the proportionality constant,  $K_1$  has been taken to be  $= 3.75 \times 10^4$  yrs, assuming the commonly adopted values of  $1.35 \times 10^{21}$  litres for the oceanic volume<sup>29</sup> and  $3.6 \times 10^{16}$  litres for the volume of global annual river run off.<sup>30</sup> The constant  $K_2$  is

calculated to be 10, assuming a mean oceanic depth of 3750 metres and world average sedimentation rate of  $1 \text{ gm/cm}^2 \cdot 10^3 \text{ yrs}$ .

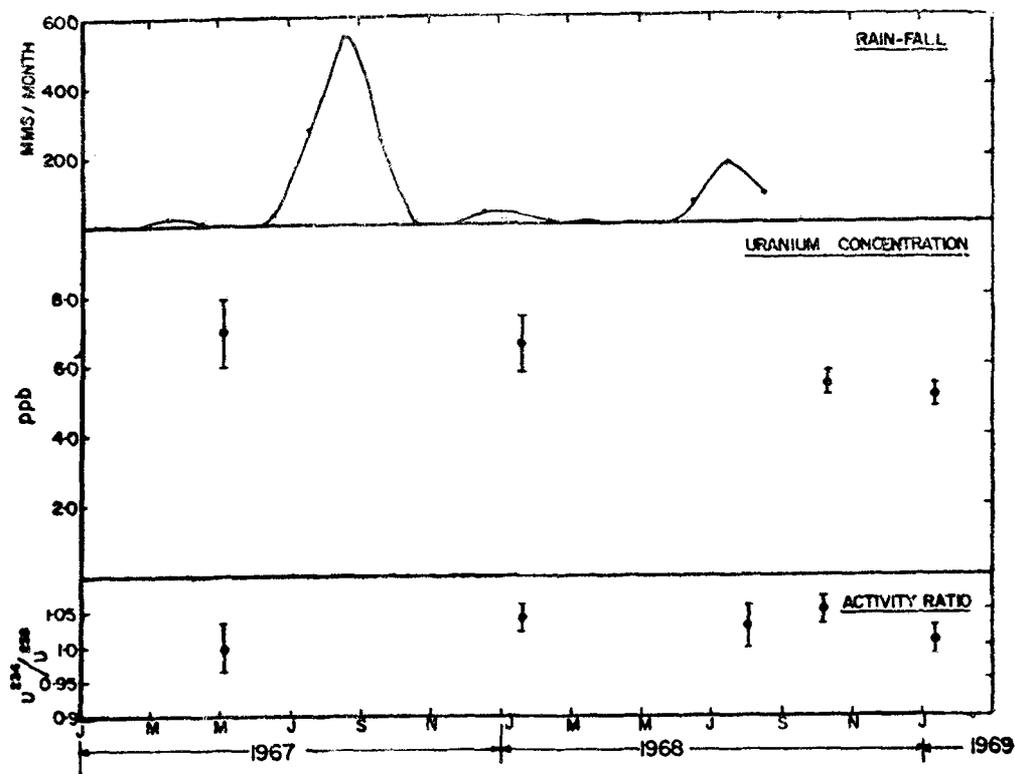


FIG. 4. Measured values of uranium concentration and activity ratios U-234/U-238, in Ganges river sample at Allahabad. The concentration of uranium is observed to decrease during the monsoon period of 1968. No samples were collected during 1967 monsoon.

Figure 5 shows the relation between equations (2), (3) and (4). The shaded bands on the axes give the ranges in the weighted mean of U-234/U-238 activity ratios for the world rivers, concentration of authigenic uranium present in deep sea sediments (based on the results of Ku<sup>31</sup>) and the weighted mean of U-238 concentration in rivers. From this figure it is clear that the amount of uranium depositing on the ocean floor could be explained by input from rivers, *i.e.*, conditions given by equations (3) and (4) are not necessarily violated. However, in order to explain the uranium concentrations observed in the sea by a model in which the input is only by rivers, it becomes necessary to have U-234/U-238 activity ratios at input to exceed 1.3 (considering the upper limit of average U-238 concentration

in rivers) and to exceed 1.6 (to explain the observed concentration of authigenic uranium in deep sea sediments).

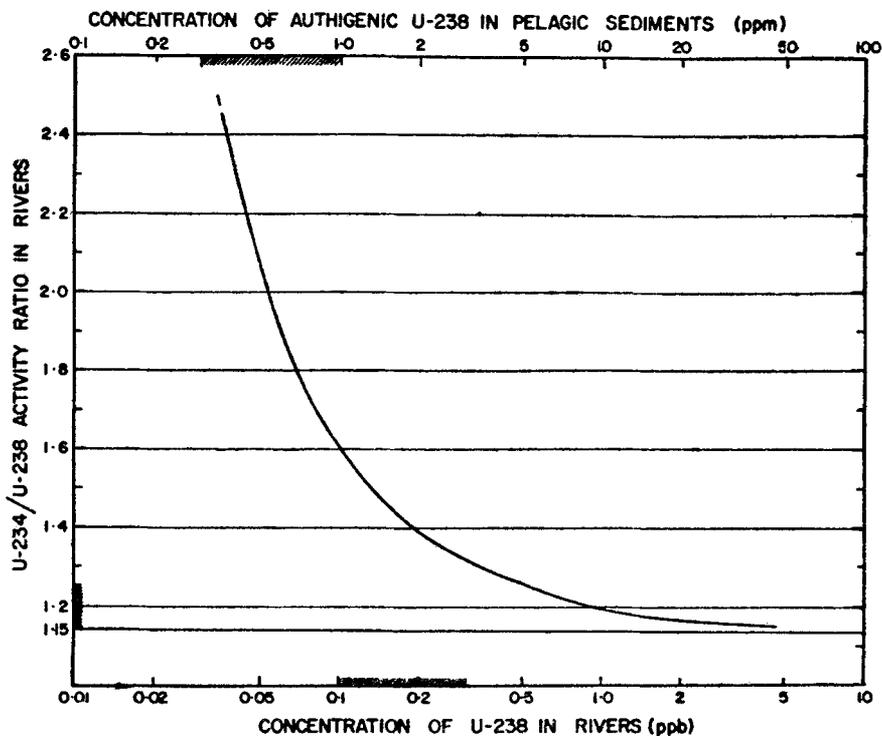


FIG. 5. Theoretically estimated correspondence between average weighted U-234/U-238 activity ratios and U-238 concentration in rivers, based on a steady state river-influx model (see equations 2 and 3). The corresponding expected authigenic U-238 concentrations in pelagic sediments are also given (cf. equation 4).

Hence there must be some other agency/mechanism which constitutes an important source of uranium in the oceans. Further in this source the U-234/U-238 activity ratio should be high ( $\sim 2$ ). One of the possible sources could be the migration of uranium from deep sea sediments as was suggested by Ku<sup>31</sup> which is supported by the observed deficiency of U-234 in the upper layers of the sediments.<sup>31, 32</sup> This may be due to the fact that there has been a preferential leaching of U-234 in sediments possibly due to the difference in the chemical nature of the two isotopes.

Another possible source is the *in situ* leaching of eolian dust and river suspended load in sea-water. The quantitative aspects of both these alternatives will be discussed in detail elsewhere.<sup>17</sup>

TABLE IV

*Concentrations\* of Ra-226 and Ra-228/Ra-226 activity ratios in Indian Rivers*

River	Location	Ra-226 (dpm/litre)	Ra-228/Ra-226 (activity ratio)
Ganges	.. Hardwar	0.20	0.78
do.	.. Nabdeep	..	1.95
Godavari	.. Rajahmundry	0.05	..
Krishna	.. Vijayawada	0.05	..
Sabarmati	.. Ahmedabad	0.09	3.40

\* The statistical errors in the determination of Ra-226 are  $\pm 15\%$  only 5 litre samples were analysed. The activity ratios, Ra-228/Ra-226 are reliable to  $\pm 5\%$  samples of 200-400 litres were analysed.

TABLE V

*Available data on the concentrations of Uranium and U-234/U-238 activity ratios for world rivers*

	Uranium (ppb)	Average U-234/U-238 activity ratio	Mean discharge* (cu.ft./sec.) $\times 10^3$	Reference
Indian rivers	.. 0.01-7.0	1.2 (1.00-1.58)	600†	Present study
Russian rivers	.. ..	1.2 (1.12-1.29)	4000	5
Amazon	.. 0.04	1.1	6000	4
Mississippi	.. 1.0	1.3	620	4

Figures in parentheses give the range.

\* Livingstone.<sup>80</sup>

† Private communication from the Officers-in-Charge, Central Water and Power Commission, New Delhi.

It may be added here that it is quite likely that a major portion of the uranium brought to the sea by rivers appears to be deposited in the shelf regions due to reducing conditions prevalent in the coastal regions.<sup>33</sup> If most of the riverborne uranium is deposited on the shelf, then it is required that the ratio in the additional source discussed above should have a value close to 1.5. However, if river influx effectively constitutes greater than 50% of the uranium input to open oceans, then it is necessary that the additional source should have a ratio exceeding a value of 2 or thereabouts.

(b) *Radium*.—The concentrations of Ra-226 in Indian rivers are similar to the published values for other world rivers<sup>4, 15</sup> while those of Ra-228 are at least an order of magnitude higher than the only reported value for Amazon.<sup>13</sup>

In Fig. 6, the Ra-226 concentrations are plotted against the  $(Ca+Mg)$ <sup>30</sup> concentrations. There seems to be a linear correlation between the

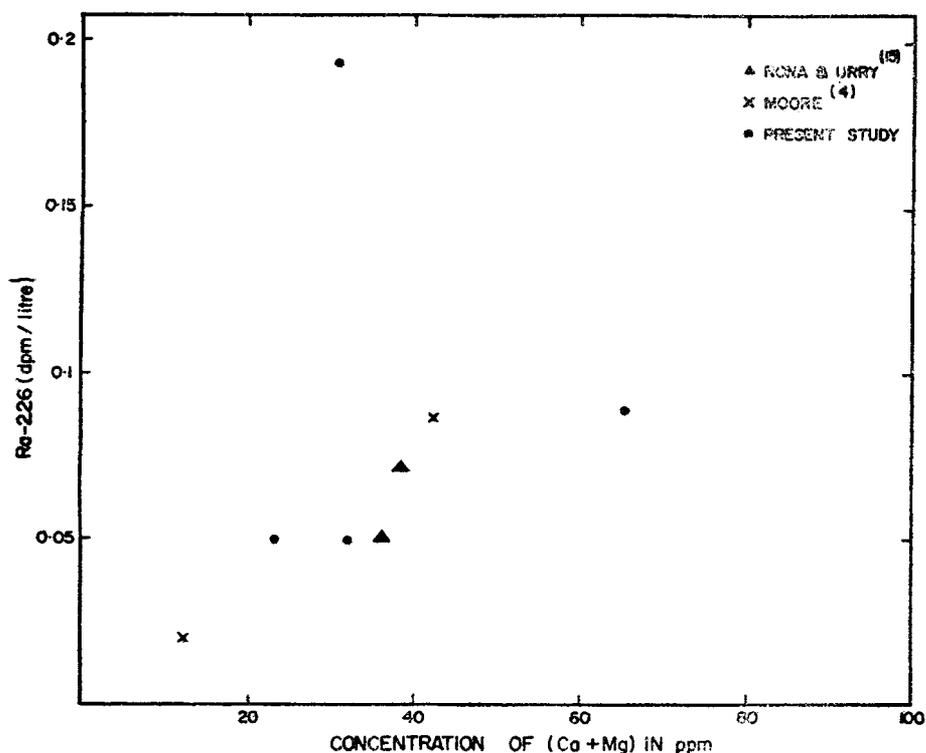


FIG. 6. Measured Ra-226 activity is plotted *versus*  $(Ca + Mg)$  concentration of river. The  $(Ca + Mg)$  data for Indian rivers are from present study, and for other world rivers, the data are from Livingstone.<sup>30</sup>

(Ca + Mg) and Ra concentrations which is not unexpected because of the similar behaviour of the alkaline earth elements.

Proceeding in a manner similar to that adopted for uranium and taking the respective mean lives of the nuclides, Ra-226 and Ra-228 as the upper limits for their residence times in the oceans, it is seen that the observed concentration of these isotopes in rivers fall short by factors of 30 and 10 respectively to explain their observed sea-water concentrations. Once again we note that one cannot satisfactorily make a material balance in these cases. For the long-lived Ra-226, the observed concentrations in sea-water could be explained away in terms of diffusion from deep sea sediments, a mechanism proposed by Koczy.<sup>34</sup> However, in the case of Ra-228 which has a short half-life, the observed concentrations in the upper layers of the oceans<sup>13</sup> cannot be due to diffusion from deep sea sediments.

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