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# Characterization of Minerals and Naturally Occurring Radionuclides in River Sediments

[1]V. Ramasamy, [1]G. Suresh, [2]V. Meenakshisundaram and [2]V. Gajendran
[1] Department of Physics, Annamalai University, Annamalainagar, Tamilnadu, India
[2]Health and Safety Division, IGCAR, Kalpakkam, Tamilnadu, India

**Abstract:** The minerals such as quartz, feldspar in different structure, kaolinite, calcite, gibbsite, montmorillonite, smectite, organic carbon and polygorskite are identified in 40 locations of Ponnaiyar river by Fourier Transform Infrared (FTIR) spectroscopic technique. The relative distribution of major minerals such as quartz, feldspar (orthoclase and Microcline) and kaolinite are determined by calculating extinction co-efficient. The naturally occurring radionuclides ($^{238}U$, $^{232}Th$ and $^{40}K$) and associated absorbed dose rate are determined by Gamma ray spectroscopic technique. The determined activity concentrations and average absorbed dose rate of all sites are fall within the typical world and Indian average values although some extreme values are determined. Correlation analysis is carried out for distributions of quartz, feldspar and kaolinite and naturally occurring radionuclides, with associated absorbed dose rate.

**Key words:** FTIR, minerals, radioactivity, river sediments and gamma ray spectroscopic technique

## INTRODUCTION

Sediments are detrital products of rocks and bear the mineralogical properties of the original rock formation. The principal constituents of most of the sediments are quartz, feldspar, carbonates and clay minerals. Of these, quartz is overwhelmingly the most abundant. Feldspar, though more abundant in parent igneous rock, is of intermediate durability and so runs second place to quartz in sediments. The others, though more durable than feldspar, are simply for less abundant in source materials (Dott and Batten, 1976). The mineralogical properties of sediments, reflects the geological history of transport and sorting process. The Fourier Transform Infrared (FTIR) absorption spectra of sediments contain more information about mineralogy (Ramasamy *et al.*, 2006).

Natural radioactivity is widespread in the earth environment and it exists in various geological formations such as earth crust, rocks, soils, plants, water and air. When rocks are disintegrated through natural process, radionuclides are carried to soil by rain and flows (Taskin *et al.*, 2009). The way minerals incorporate the radionuclide depend on several geological conditions, but is most strongly dependent on the mineral species and geological formation from which they originate. The radioactive nuclides uranium and thorium are not found to be associated in appreciable extent in the common mineral of the rock, but are strongly concentrated in accessory minerals.

The river sediments are used as major mixing material in building construction in India, especially in Tamilnadu (state). The characterization of minerals and radionuclides in river sediments are analyzed in an effort to better understand their spatial distributions and relations. Hence, the objective of this study is to characterize the various minerals and naturally occurring radionuclides in river sediments by FTIR spectroscopic technique and gamma ray spectroscopy respectively and also, to study the correlation between the minerals and radionuclides in sediments.

## MATERIALS AND METHODS

**Study Area:** In the present study, sediment samples were collected from various sites of the Ponnaiyar river. It is originated on the hills of Nandidrug in Kolar districts of Karnataka state, and flows south and then east for 400Km through Karnataka and Tamilnadu, and terminated at Cuddalore, Tamilnadu in Bay of Bengal. It is entered in Tamilnadu at Dharmapuri district. It covers four districts (Dharmapuri, Thiruvannamalai, Villupuram and Cuddalore) in Tamilnadu. A dam is constructed on this river at Satthanur, Chengam taluk, Thiruvannamalai district. Capacity of this dam nearly 4600 M CFT. The sediments of this river are excavated only for building constructions. The small hydraulic structure and barrages were constructed for drinking and agriculture purposes respectively on the study area. On both side of the bank of this river, so many living residents and some industries are situated. None of the industrials have proper and controlled outlet. The discharge wastes and toxic metals from such industries and living residents are directly let out in to the river. Also along the river, lot of agricultural lands is available, overuse of chemical fertilizers and pesticides are washed into the river. These are all main factors for enrichments of pollutants in the study area.

**Sample Collection and Preparation:** The present study area (Ponnaiyar river) covers a total length of 200 Km, from which 40 locations were selected. Each location is

**Corresponding Author:** V. Ramasamy, Department of Physics, Annamalai University, Annamalainagar, Tamilnadu, India

separated by a distance of 4-5 Km approximately. All sediment samples were collected at 0-10 cm depth during the summer season (April-May 2008). Each sample has a weight of 3-4 kg approximately. The collected samples were dried at room temperature in open air for two days and stored in black polythene bags.

**Sample preparation and techniques**

**FTIR:** Wet grinding was carried out by placing 30 to 50mg of the sample in an agate mortar along with 20 to 25 drops of ethanol. The ground samples were dried in a hot air oven at 110ºC to remove the moisture content and sieved to various grain sizes such as 74, 53 and 44μm. Using the KBr pellet technique, each grain sized sample was mixed with KBr at various ratios viz., 1:10, 1:20, 1:30, 1:40 and 1:50. The mixture was then pressed into a transparent disc in an evocable dye at sufficiently high pressure. The samples in the ratio 1:30 was taken for further analysis, since it gives rise to maximum transmittance and observable peaks (Ramasamy *et al.*, 2006). Using the Perkin Elmer RX1 FTIR spectrometer, the infrared spectra for all sediment samples were recorded in the region 4000-400 $cm^{-1}$. The resolution of the instruments is 0.001$cm^{-1}$ and the accuracy is 4 $cm^{-1}$.

**Gamma ray spectroscopic technique:** The samples were dried in an oven 110ºC till the constant dry weight was obtained, crushed and homogenized. The homogenized samples were packed in a 250 ml plastic container (9cm x 6.5cm: Height x Diameter) to its full volume with uniform mass. These containers shielded hermetically and also shielded externally to ensure that all daughter products of uranium and thorium, in particular, radon isotope formed, do not escape. A time of four weeks was allowed after packing to attain secular equilibrium between Ra-226 and its short-lived daughter products. The net weight of the sample was determined before counting.

The gamma ray spectrometer with NaI(TI) detector was used to determine the concentration of primordial radionuclides ($^{238}U$, $^{232}Th$ and $^{40}K$). The detector was shielded by 15 cm thick lead on all four sides and 10 cm thick on top. The energy resolution of 2.0 Kev and relative efficiency of 33% at 1.33Mev was achieved in the system with the counting time of 10000 seconds. The Standard International Atomic Energy Agency (IAEA) sources were used for calibration. From the counting spectra, the activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ was determined using computer program. The peak corresponds to 1460Kev (K-40) for $^{40}K$, 1764.5 Kev (Bi-214) for $^{238}U$ and 2614.5Kev (Ti-208) for $^{232}Th$ were considered in arriving at the activity levels (Bq/kg).

## RESULTS

The FTIR spectra were recorded for all samples. The observed absorption wave numbers are tabulated with corresponding minerals in Table 1. The minerals such as quartz, microcline feldspar, orthoclase feldspar, kaolinite, calcite, gibbsite, montmorillonite, smectite, organic carbon and polygorskite are identified by comparing the observed wave numbers with available literatures (Russell 1987 and Ramasamy *et al.*, 2004, 2005 & 2006).

Table 1: The observed absorption wave numbers and corresponding minerals from FTIR spectra

| S. No. | Mineral Name | Site number Observed | Wave numbers ($cm^{-1}$) |
|---|---|---|---|
| 1 | Quartz | $S_1$-$S_{40}$ | 460-464 |
| | | $S_1$-$S_{40}$ | 514-520 |
| | | $S_1$-$S_{40}$ | 693-694 |
| | | $S_1$-$S_{40}$ | 777-778 |
| | | $S_1$-$S_{40}$ | 794-796 |
| | | $S_1$-$S_{40}$ | 1080-1084 |
| | | $S_1$-$S_{40}$ | 1162-1164 |
| | | $S_4$-$S_{14}$, $S_{19}$-$S_{25}$, $S_{28}$, $S_{31}$, $S_{33}$-$S_{40}$ | 1870-1883 |
| 2 | Microcline Feldspar | $S_1$-$S_{40}$ | 583-587 |
| 3 | Orthoclase Feldspar | $S_1$-$S_{40}$ | 647-650 |
| | | $S_1$, $S_3$, $S_{15}$, $S_{21}$, $S_{23}$, $S_{29}$ | 532-537 |
| 4 | Kaolinite | $S_1$-$S_{40}$ | 1015-1019 |
| | | $S_1$, $S_3$, $S_{10}$, $S_{15}$ | 1030-1037 |
| | | $S_6$,$S_{10}$, $S_{15}$-$S_{19}$, $S_{24}$, $S_{32}$, $S_{35}$-$S_{36}$ | 3618-3622 |
| | | $S_1$, $S_2$, $S_{23}$, $S_{33}$, $S_{29}$, $S_{34}$ | 3690-3691 |
| 5 | Gibbsite | $S_1$-$S_{40}$ | 662-670 |
| 6 | Calcite | $S_2$, $S_3$, $S_4$,$S_{33}$, $S_{36}$ | 1420-1438 |
| 7 | Montmorillonite | $S_{36}$ | 878 |
| 8 | Polygorskite | $S_1$, $S_7$, $S_{21}$, $S_{33}$, $S_{29}$, $S_{36}$ | 3611-3615 |
| 9 | Organic Carbon | $S_1$-$S_{40}$ | 2923-2929 |
| | | $S_1$-$S_{40}$ | 2852-2865 |
| 10 | Smectite | $S_{27}$ | 523 |

Table 2: The Extinction-coefficient of Quartz, Microcline Feldspar, Orthoclase Feldspar and Kaolinite of Ponnaiyar river sediments

| Site Number | Extinction Co-efficient | | | |
|---|---|---|---|---|
| | Quartz | Microcline Feldspar | Orthoclase Feldspar | Kaolinite |
| $S_1$ | 92.65 | 26.65 | 8.39 | 1.10 |
| $S_2$ | 40.91 | 6.73 | 3.38 | 1.19 |
| $S_3$ | 171.82 | 26.83 | 21.11 | 2.29 |
| $S_4$ | 156.53 | 6.65 | 5.09 | 2.52 |
| $S_5$ | 77.25 | 13.55 | 5.16 | 2.87 |
| $S_6$ | 137.30 | 18.92 | 12.27 | 3.05 |
| $S_7$ | 107.01 | 15.58 | 8.6 | 3.10 |
| $S_8$ | 139.57 | 17.15 | 10.42 | 3.08 |
| $S_9$ | 95.70 | 18.69 | 6.85 | 3.10 |
| $S_{10}$ | 147.14 | 34.99 | 19.41 | 3.16 |
| $S_{11}$ | 110.38 | 6.81 | 8.65 | 3.22 |
| $S_{12}$ | 107.01 | 17.04 | 10.28 | 3.30 |
| $S_{13}$ | 92.31 | 15.29 | 8.6 | 3.39 |
| $S_{14}$ | 62.06 | 8.39 | 6.81 | 3.17 |
| $S_{15}$ | 153.10 | 29.83 | 17.48 | 3.73 |
| $S_{16}$ | 127.93 | 6.65 | 15.58 | 3.82 |
| $S_{17}$ | 49.35 | 20.58 | 5.09 | 3.22 |
| $S_{18}$ | 112.09 | 46.43 | 10.35 | 3.92 |
| $S_{19}$ | 210.10 | 24.02 | 23.09 | 4.88 |
| $S_{20}$ | 106.21 | 37.10 | 10.35 | 4.25 |
| $S_{21}$ | 165.43 | 27.45 | 20.84 | 4.01 |
| $S_{22}$ | 111.24 | 8.39 | 11.96 | 4.03 |
| $S_{23}$ | 70.63 | 11.82 | 5.92 | 4.03 |
| $S_{24}$ | 132.11 | 42.84 | 10.55 | 4.09 |
| $S_{25}$ | 275.75 | 8.33 | 27.36 | 4.19 |
| $S_{26}$ | 47.67 | 20.45 | 5.06 | 4.24 |
| $S_{27}$ | 127.23 | 29.64 | 13.89 | 4.25 |
| $S_{28}$ | 141.86 | 6.65 | 13.72 | 4.32 |
| $S_{29}$ | 45.98 | 22.23 | 5.09 | 4.33 |
| $S_{30}$ | 112.93 | 4.99 | 12.04 | 4.42 |
| $S_{31}$ | 171.82 | 8.44 | 15.68 | 4.48 |
| $S_{32}$ | 71.64 | 25.82 | 5.12 | 4.57 |
| $S_{33}$ | 159.21 | 5.19 | 13.98 | 4.68 |
| $S_{34}$ | 108.66 | 24.17 | 10.42 | 4.69 |
| $S_{35}$ | 181.5 | 717.26 | 17.93 | 4.79 |
| $S_{36}$ | 35.68 | 10.03 | 5.09 | 5.01 |
| $S_{37}$ | 88.88 | 13.38 | 8.65 | 4.95 |
| $S_{38}$ | 71.13 | 8.28 | 6.81 | 4.97 |
| $S_{39}$ | 66.3 | 16.65 | 5.09 | 5.07 |
| $S_{40}$ | 78.36 | 10.25 | 6.85 | 5.16 |

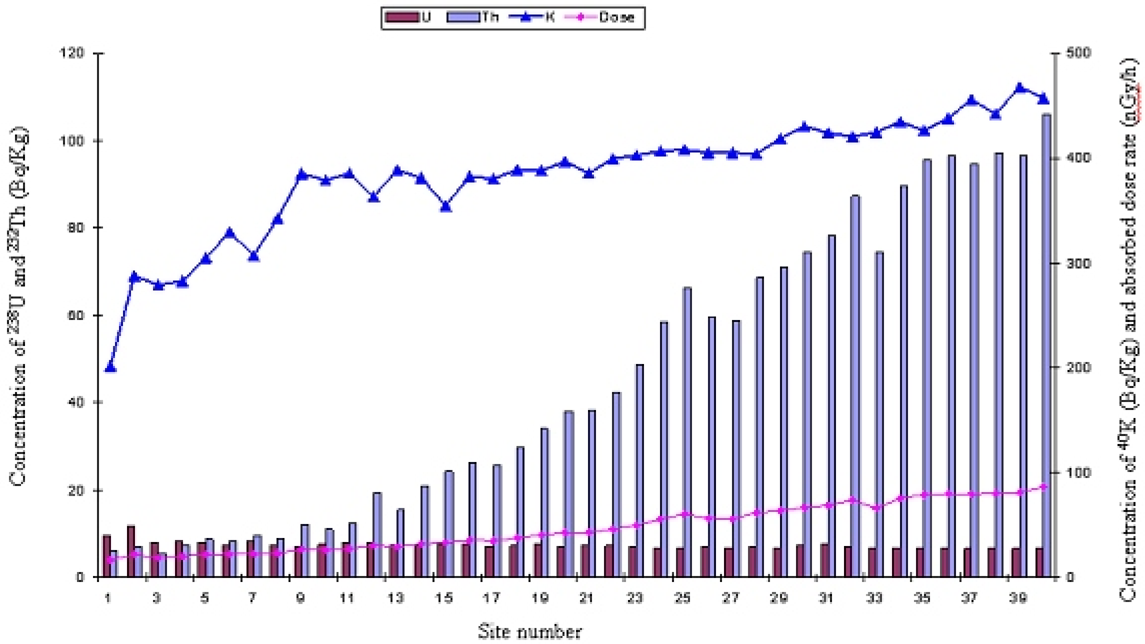


Fig 1: Concentration of $^{238}U$, $^{232}Th$, $^{40}U$ and Absorbed dose rate of all sites

**Extinction co-efficient of Quartz, Feldspar and Kaolinite:** With reference to the number of peaks and intensity, the minerals such as quartz, feldspar and kaolinite are consider as major. The other minerals are accessory. The relative distribution can be quantified by calculating the extinction co-efficient for the characteristic peaks quartz, orthoclase feldspar, microcline feldspar and kaolinite at around 778, 647, 585 and 1015 $cm^{-1}$ respectively for all sites using the formula (Ramasamy *et al.*, 2006). From the calculated values (Table 2), maximum extinction co-efficient values for quartz, orthoclase feldspar, microcline feldspar and kaolinite are 275.75, 46.43 and 5.16 in the site no. $S_{25}$, $S_{18}$ and $S_{40}$ respectively. In the same way, minimum values are 35.68, 6.65 and 1.10 are in the site no. $S_{36}$, $S_{28}$ and $S_1$ respectively.

**Activity Concentration of $^{238}U$, $^{232}Th$ and $^{40}K$:** The activity concentration of natural radionuclides ($^{238}U$, $^{232}Th$ and $^{40}K$) for all samples is determined and is shown in Fig. 1. It varies from site to site, because river bottoms can exhibit large variation in chemical and mineralogical properties (Krmar *et al.*, 2009). The mean activity concentration ranges for $^{238}U$, $^{232}Th$ and $^{40}K$ are BDL - 11.60±6.13Bq/kg with an average 7.31 ± 3.41Bq/kg, BDL - 106.11 ± 9.20Bq/kg with an average 46.85 ± 5.25Bq/kg and 201.23 ± 19.90 - 467.71 ± 34.34Bq/kg with an average 384.03±26.82Bq/kg respectively.

**Absorbed Dose rate:** The mean activity concentrations of $^{238}U$, $^{232}Th$ and $^{40}K$ are converted in to dose rate based on the conversion factor given by UNSCEAR (2000) and El-Gamal *et al.,* (2007)(Fig. 1).

$$D = (0.462C_U + 0.604\ C_{Th} + 0.0417\ C_K)\ nGyh^{-1}$$

Where D is the absorbed dose rate ($nGyh^{-1}$), $C_U$, $C_{Th}$ and $C_K$ are the activity concentrations (Bq/kg) of $^{238}U$, $^{232}Th$ and $^{40}K$ in river sediments respectively. The range of absorbed dose rates is from 14.72nGy/h to 88.95nGy/h with average of 47.07nGy/h.

## DISCUSSION

Quartz is common and invariably present in all samples. The presence of quartz in the samples can be explained by Si-O asymmetrical bending vibrations around 464 $cm^{-1}$, Si-O symmetrical bending vibrations around 694 $cm^{-1}$, Si-O symmetrical stretching vibrations at around 778 and 796 $cm^{-1}$, while the 1082 and 1162 $cm^{-1}$ absorption region arises from Si-O asymmetrical stretching vibrations due to low Al for Si substitution. These assignments are in good agreement with the observation on the quartz mineral obtained by Ramasamy *et al.*, 2006.

Feldspar is also frequent constituents in sediments. The peak corresponding the range 583-587 $cm^{-1}$ is due to the O-Si-(Al)-O bending vibration for microcline and in the ranges 647-650 $cm^{-1}$ are due to the Al-O coordination vibrations indicates the presence of orthoclase feldspar.

Kaolinite is the clay mineral. The presence of band near 3620 $cm^{-1}$ arises from internal OH group and that near 3690 $cm^{-1}$ arises from internal surface OH group (Russell 1987; Ramasamy *et al.*, 2006). The band near 1015 $cm^{-1}$ is present in all the sites indicates the presence of Kaolinite (Russell 1987).

Calcite is the most common carbonate mineral in sediments. From the existence of a peak in the range 1420-1438 $cm^{-1}$ it is easily recognized that the calcite is present in site no. $S_2$-$S_4$, $S_{33}$ and $S_{36}$. The existence of a peak near 665, 878, 523, 2925 &2855 and 3612 $cm^{-1}$ respectively indicate that presence of gibbsite, montmorillonite, smectite, organic Carbon and polygorskite. From extinction co-efficient values, the amount of kaolinite is very much lesser than quartz and lesser than orthoclase and microcline feldspar.

In all sampling sites, mean activity concentration of naturally occurring radio nuclides is of the order $^{238}U < ^{232}Th < ^{40}K$. In particular $S_2$, the activity concentration of $^{238}U$ is high, which may be due to the solubility and mobility of U (VI)$O_2^{2+}$ (Powell *et al.*, 2007). However, the $S_{40}$ is having high activity concentrations. Increasing concentration of $^{232}Th$ and $^{40}K$ may be due to the high content of monazite (Orgun *et al.*, 2007). The increasing trend of $^{40}K$ is due to presence of clay sediments (El-Gamal *et al.*, 2007)

The concentration of $^{238}U$, $^{232}Th$ and $^{40}K$ for all measured samples are below the world and Indian average values (World average value of $^{238}U$, $^{232}Th$ and $^{40}K$ is 33Bq/kg, 45 Bq/kg and 420Bq/kg respectively. Indian average value of $^{238}U$, $^{232}Th$ and $^{40}K$ is 28.67 Bq/kg, 63.83 Bq/kg and 327.6 Bq/kg respectively). However in some sampling sites, concentration of $^{232}Th$ is higher than world average value, indicating that clay mineral may exist at that sampling site. Ramasamy *et al.*, 2004a & 2006a reported the values of Palaru and Cauvery rivers, which are higher than the present values. Average absorbed dose rate for all samples are lower than the world average value (55nGy/h) (UNSCEAR 2000).

**Correlation analysis:** In order to determine inter- relation between the minerals and concentrations of radionuclides in the sediments, Pearson correlation analysis is carried out using SPSS for windows 16.0 software, and is shown in Table 3 as the linear correlation matrix. From the correlation analysis, the distribution of quartz and feldspar are poorly correlated with individual activities of $^{238}U$, $^{232}Th$ and $^{40}K$ and absorbed dose rate (Table 3). But the distribution of Kaolinite (clay minerals) is well positively correlated with individual activities of $^{232}Th$ and $^{40}K$ and absorbed dose rate (Table 3).

.Table 3: Correlation analysis of Extinction coefficient of Quartz, Microcline Feldspar, Orthoclase Feldspar and Kaolinite and concentration of Radionuclides of Ponnaiyar river sediments

| | $^{238}U$ | $^{232}Th$ | $^{40}K$ | ADR | E-Q | E-MCF | E-OCF | E-KA |
|---|---|---|---|---|---|---|---|---|
| $^{238}U$ | 1 | | | | | | | |
| $^{232}Th$ | -0.623 | 1 | | | | | | |
| $^{40}K$ | -0.769 | 0.818 | 1 | | | | | |
| ADR | -0.638 | 0.999 | 0.847 | 1 | | | | |
| E-Q | -0.107 | -0.089 | -0.076 | -0.093 | 1 | | | |
| E-MCF | -0.098 | -0.118 | -0.061 | -0.118 | 0.116 | 1 | | |
| E-OCF | -0.132 | -0.046 | 0.013 | -0.044 | 0.90 | 10.221 | 1 | |
| E-KA | -0.822 | 0.852 | 0.922 | 0.869 | 0.075 | -0.0050. | 137 | 1 |

$^{238}U$, $^{232}Th$ and $^{40}K$ are the concentrations of natural radionuclides (Bq/Kg). ADR – Absorbed Dose rate (nGy/h). E-Q - Extinction co-efficient of Quartz. E-MCF - Extinction co-efficient of Microcline Feldspar. E- OCF- Extinction co-efficient of Orthoclase Feldspar and E-KA - Extinction co-efficient Kaolinite.

## CONCLUSION

From the FTIR study, the minerals such as quartz, feldspar (orthoclase and feldspar) and kaolinite are as major minerals. Others such as calcite, gibbsite, smectite, montmorillonite, polygorskite and organic carbons are as accessory minerals in Ponnaiyar river sediments. The calculated extinction co-efficient values show that, the amount of quartz is greater than feldspar (orthoclase and feldspar) and very much greater than kaolinite in all the sites. The average activity concentrations of Ponnaiyar river sediments were within the world and Indian average value, although some extreme values have been determined. The Ponnaiyar river sediments have insignificant radiological hazards. The obtained positive correlation (r = 0.87) between the distribution of kaolinite(clay minerals) and absorbed dose rate shows that, clay minerals like kaolinite may act as a booster of level of radioactivity.

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