1. INTRODUCTION

1.1 General

Exposure from natural sources of ionizing radiation is a continuous and unavoidable feature of life on earth. The major sources responsible for this exposure are the naturally occurring radionuclides presents in the earth's crust. External irradiation from radionuclides naturally presents in the environment or released from man-made practices or events is usually an important component of the exposure of human populations. Exposures from natural sources are due to external exposure, internal exposure from radionuclides taken to the body through ingestion of food and inhalation of radon. In addition, some of the technological activities such as production of electricity from coal based thermal power station and the production and use of phosphate fertilizers may enhance the natural radiation level of the environment.

Exposures due to natural radiation are of particular importance because it accounts for the largest contributor to the total collective radiation dose to the world population (UNSCEAR 2000). Many radionuclides occur naturally in terrestrial soils and rocks and in building materials derived from them. Upon decay, these radionuclides produce an external radiation field to which all human beings are exposed. In terms of dose, the principal radionuclides are $^{232}$Th, $^{238}$U and $^{40}$K. Both $^{232}$Th and $^{238}$U head series of radionuclides that produce significant
exposures. The decay of naturally occurring radionuclides in soil produce a gamma-beta radiation field in soil that also crosses the soil-air interface to produce exposures to humans. The widespread interest in the subject of environment has resulted in acceleration of research on the behavior of trace substances including radioactive materials in the environment. Radionuclides are generally released in trace quantities to the environment. They are then physically transported in the air or water media in which they are located. A more extensive and systematic investigations are needed to obtain a clear understanding of the basic dynamics of radioactivity in the environment.

The natural radiation level varies from place to place depending on the local geology of the region. However, there are regions in the world where the radiation level can be exceptionally high from the normal variability ranges leading to the formation of what are known as high background radiation areas. The high background areas are of special interest because they present anomalous conditions in their geological and geochemical features and, consequently, in the background radiation levels. One of the prime sources for high background radiation level is the presence of radioactive monazite and the presence of these mineral deposits on the coastal areas of south west coast of India has been reported by several investigators (Sunta et al., 1982, Nambi et al., 1986, Radhakrishna et al., 1993). Though the presence of monazite deposit in these areas has been well established, systematic studies on the sources and geochemical processes leading to the formation of monazite deposits area are sparse.
Limited information in literature indicates that the monazite deposits on the coastal areas of Kerala and Tamilnadu are formed due to the weathering of rocks in Nilgiri Hills and Western Ghats (Mahadevan et al., 1956, Selvasekarapandian et al., 2000). In view of this, systematic studies on the transport of radionuclides from western ghats to Arabian sea through major rivers of coastal Karnataka, viz Kali, Sharavathi and Netravathi have been carried out to trace the sources and processes that are responsible for the formation of low level monazite deposit near Ullal beach area, on the south west coast of India. These rivers originate in different locations of Western Ghats and join Arabian sea at different locations that are distantly separated. Hence, a comparative study of radionuclides concentration in soil, sediment, rock and water samples at different locations of these rivers would throw light on the dynamics of the environment in terms of transport of radionuclides.

1.2 Sources of background radiation

The knowledge of distribution of radionuclides, their transport and radiation levels in the environment is important for understanding the dynamics of the environment and for assessing the effects of radiation exposure due to both extraterrestrial and terrestrial sources. While terrestrial component originates from primordial radionuclides in the earth crust, the extraterrestrial radiation originates from outer space as primary cosmic rays. For most of the world’s population, the variation of individual doses from natural sources is considered to be rather narrow. However, there are regions in the world where external exposure from
natural sources may substantially exceed from the normal variability range. Apart from the natural sources, modern scientific and technological activities also contribute to the radiation level of the environment, which would be significant if proper precautions are not taken and technologically enhanced radionuclide reflects a growing awareness of the radiological significance of natural radionuclides.

1.2.1 Extra terrestrial sources

Extra terrestrial sources due to cosmic ray radiation consists of protons (85 %), alpha particles (14%) and about 1% from nuclei of atomic number between 4 and 26. These particles are highly penetrating and have high energy. Attenuation in the atmosphere decreases the flux of the cosmic rays on the earth's surface. As a result of this, the cosmic ray exposure becomes double for every 1500 meters above the earth's surface. When these primary cosmic ray particles interact with earth's atmosphere, they produce quite a variety of radionuclides and sub atomic particles like mesons, muons and electrons. The sub atomic particles thus produced are known as secondary cosmic rays. At sea level the mesons account for 80% of the cosmic radiation and electrons account for about 20%. The dose rate at sea level due to cosmic rays is found to be about 32 nGy h\(^{-1}\) (Nambi et al., 1986). The cosmic ray component is usually very stable at the earth's surface, but it does vary with the geometric latitude and to a greater extent, it increases with altitude above sea level.
1.2.2 Terrestrial sources

Terrestrial radiation is due to radioactive nuclides present in varying amounts in soil, building materials, water, rocks and contribution due to the inhalation of $^{220}\text{Rn}$ and its short-lived decay products can equal or even exceed that of $^{222}\text{Rn}$ and its progeny.

The natural occurring terrestrial radionuclides can be divided into those that occur singly and those that are components of three chains of radioactive elements, namely the uranium series which originates with $^{238}\text{U}$, the thorium series which originates with $^{232}\text{Th}$ and the actinium series which originates with $^{235}\text{U}$. Among singly occurring radionuclides of terrestrial origin, $^{40}\text{K}$ is important. The natural background radiation level varies from place to place due to variations in the distribution of these radionuclides in the environment. It is important to understand the behaviour of natural radionuclides in the environment (distribution pathways, mobility and transfers) because the information can be used as a natural analogue for the long-term behaviour of materials and processes in developing and testing models, and in obtaining the associated values appropriate for radiological performance assessments (Thiessen et al., 1999).

1.3 Technologically enhanced natural radiation

Some of the present day technological activities involve enhanced natural radiation exposure to human population. The major pathways which lead to the enhanced radiation exposure are burning of coal in thermal power stations and the use of phosphate fertilizers in agriculture. Coal contains trace levels of natural
Introduction

radionuclides and its combustion results in their release into the environment mainly due to the escape of fly-ash to the atmosphere.

The release of these radionuclides causes an enhancement in the ambient radiation fields in the immediate human environment. On the global scale it is estimated that about 4,000 tons of uranium and 8,000 tons of thorium are distributed along with their daughter products solely by coal combustion (Fremlin and Wilson, 1979). The main pathways through which the population living around coal-fired power plants is exposed to enhanced level of natural radionuclides are: inhalation during the passage of the plume, external exposure and inhalation and ingestion resulting from the radionuclides deposited on the ground (UNSCEAR, 1982).

The phosphate rock is used as a source of phosphorous fertilizer. It contains trace amounts of \(^{238}\text{U}\), \(^{226}\text{Ra}\), \(^{232}\text{Th}\) and \(^{40}\text{K}\) which are redistributed to the environment in the course of rock’s industrial processing and use. This comes about through effluent discharges, the agricultural use of fertilizers and the utilization of by products and waste material for other purposes (UNSCEAR, 1982).

1.4 Artificial radionuclides

The use of radioisotopes in medicine, open air nuclear weapon tests and operation of nuclear power reactors are the major sources of artificial radioactivity. Among the several hundred radionuclides produced by nuclear explosions, only \(^{90}\text{Sr}\) and \(^{137}\text{Cs}\) contribute significantly to human exposure as they stay for longer
time in the environment due to their relatively long half lives. Strontium - 90 is a pure beta emitter with average energy of 195.8 keV and decays with half life of 28.6 years (Kocher, 1977). Cesium is also a beta emitter with average beta energy of 170.8 keV and decays with a half life of 30.2 years. The deposition of $^{90}\text{Sr}$ on land and the transfer to humans by ingestion is the most important pathway for human exposure. Strontium is chemically similar to calcium and therefore enters the human body following its path and concentrates in the skeleton causing internal exposure. On the other hand, $^{137}\text{Cs}$ is chemically similar to potassium and follows its path to get into the human body. After entering the human body it becomes relatively uniformly distributed in soft tissues (UNSCEAR, 1982).

1.5 Literature survey

Extensive studies have been reported on radiation level and distribution of radionuclides in different environs of the world. A brief survey of some of these important investigations is presented in this section.

1.5.1 Gamma dose rate

Surveys on gamma dose rates in air have been carried out by a number of investigators. Lowder et al., (1964); Spiers et al., (1964); Sansoni (1982); Schmier et al., (1982); Myrick et al., (1983); Pan Ziquiang et al., (1988); Carreiro et al., (1988); Megumi et al., (1988); Joshi (1989); Lakshmi (1989); Simon and Ibrahim (1990); Leung et al., (1990); Dailing et al., (1990); Wollenberg and Smith (1990);
Markkanene and Arvela (1992); McAulay and Marsh (1992); Fernandez Aldecoa et al., (1992); Amaral et al., (1992); Nambi et al., (1993); Labrecuqe (1994); Schuch et al., (1994); Quindos et al., (1994); Baeza et al., (1995); Weng (1996); Anagnostakis et al., (1996); Alberto Malanca et al., (1996); Bellia et al., (1997); Shenber (1997); Alam Butt et al., (1998); Bygaart and Protz (1999); Papastefanou et al., (1999); Karahan and Bayulken (2000); Al-Jundi et al., (2003); Freitas and Alencar (2004); Ramli et al., (2005); Nasim Akthar et al., (2005) Vassas et al., (2006) and Merdanoglu and Altinsoy (2006) are some of those who have reported the ambient gamma radiation level for different environs of the world. The reported world range (UNSCEAR, 2000) was 18 - 93 nGy h\(^{-1}\) with a median value of 57 nGy h\(^{-1}\).

The surveys were conducted using different methods and type of instrumentation. In an extensive nationwide survey using TL dosimeters, Nambi et al., (1987) have projected a national average value of 775 μGy y\(^{-1}\). The TLD results in general seem to be consistent with the various geological units of the Indian land mass. The Deccan plateau, consisting mostly of basalts of very Low primordial radioactivity has shown the lowest radiation levels in ambient air.

In India, several investigators have carried out gamma radiation survey. Kamath et al., (1996) have reported the gamma dose rate in the range 7.3 - 80.1 nGy h\(^{-1}\) for Indian environ from a nation wide survey. Mistry et al., 1965; Iyengar et al., (1980); Rajan et al., 1980; Lalit et al., (1987); Nambi et al., 1987; Lakshmi (1989); Radhakrishna et al., (1993); Nambi et al., (1993); Nagaiah et al., (1995); Anandaram et al., (1998); Manjunatha et al., (1998); Selvasekarapandian et al., (2000); Narayana et al., (2001); Kannan et al., (2002) and Sadasivan et al.,
(2003) have also reported gamma dose rate in air for different environs of India which vary in the range 44 - 500 nGy h⁻¹.

1.5.2 Activity of natural radionuclides

**Potassium - 40**

Potassium - 40 emits gamma radiation of energy 1.46 MeV and the activity of $^{40}$K in environmental samples can be conveniently measured by gamma spectrometry. Several investigators have reported the activities of $^{40}$K in soils of different environs. Myrick et al., (1983); Pan Ziquiang et al., (1988); Megumi et al., (1988); McAulay and Moran (1988); Leung et al., (1990); Simon and Ibrahim (1990); Klemola et al., (1991); Fernandez-Aldecoa et al., (1992); McAulay and Marsh (1992); Quindos et al., (1994); Labrecque (1994); Schuch et al., (1994); Baeza et al., (1994); Alberto Malanca et al., (1996); Anagnostakis et al., (1996); Weng (1996); Bellia et al., (1997); Shenber (1997); Alam Butt et al., (1998) and Bygaart and Protz (1999); Al-Jundi et al., (2003) and Nasim Akthar et al., (2005) are some of the investigators who have reported $^{40}$K activities in soil.

Mishra and Sadasivan (1971); Rao et al., (1983); Kamath et al., (1996); Lakshmi et al., (1997); Anandaram et al., (1998); Majunatha et al., (1998); Selvasekarapandian et al., (1999); Narayana et al., (2001) and Sadasivan et al., (2003) have reported the activity of $^{40}$K for Indian soil. The reported values of $^{40}$K in the Indian environs ranges from 9 - 572 Bq kg⁻¹ in areas of normal background and 24 - 1362 Bq kg⁻¹ in high background areas. The values reported for rest of the countries are in the range 29 - 1036 Bq kg⁻¹. From the analysis of numerous
data from different parts of the world United Nations Scientific Committee on Effect of Atomic Radiation (UNSCEAR, 2000) has come out with a world wide population – weighted average value of 420 Bq kg\(^{-1}\). As per UNSCEAR (2000) Survey of Natural Radiation Exposures, the \(^{40}\)K concentration for Indian soils ranges from 38 - 760 Bq kg\(^{-1}\) with a mean value of 400 Bq kg\(^{-1}\).

**Radium – 226**

Since \(^{226}\)Ra and its daughters are responsible for a major fraction of the dose received by the human population, concentration of \(^{226}\)Ra in different environmental matrices were determined by large number of investigators (Vinogradrov, 1959; Hallden and Fisenne 1961; Marsden, 1964; Kamath et al., 1964; Smith and Watson 1964; Bortoli and Gaglione 1969, 1972; Mishra and Sadasivan 1971; Khademi and Alemi 1980; Iyengar et al., 1980; Kodaira et al., 1980; Lalit and Shukla 1982; Schuttelkopf and Kiefer 1982; Kljaic et al., 1982; Garzon et al., 1982; Myrick et al., 1983; Rao et al., 1983; Delauene et al., 1986; Chung-keung Man et al., 1987; McAualy and Moran 1988; Paul Linsalata et al., 1989; Maul and O Hara 1989; Nageshwara Rao 1989, 1990; Mollah 1989; Wollenberg and Revzan 1990; Menon et al., 1992; Alam et al., (1999); Narayana et al., 2001; Navas et al., 2002; Topcuoglu et al., (2003); Nasim Akthar et al., 2005); Vukovic et al., (2006) and Bikit et al., (2006).

Kamath et al., (1996); Lakshmi et al., (1997); Anandaram et al., (1998); Manjunatha et al., (1998); Selvasekarapandian et al., (1999) and Narayana et al., (2001) have reported the activities of these radionculides for different parts of
India. From the analysis of numerous data from different parts of the world United Nations Scientific Committee on Effect of Atomic Radiation (UNSCEAR, 2000) has come out with a population weighted world average value of 32 Bq kg\(^{-1}\) in soil. As per UNSCEAR (2000) Survey of Natural Radiation Exposures, the \(^{226}\)Ra concentration for Indian soils ranges from 7 - 81 Bq kg\(^{-1}\) with a mean value of 29 Bq kg\(^{-1}\).

**Lead – 210**

Studies reported on \(^{210}\)Pb activities in environmental samples are quite sparse. Jawaorowski (1967); Parfenov (1974); Kodaira et al., (1980); Schuettekof and Kiefer (1982); Ibrahim and Whicker (1987); Santos et al., (1990); Amaral et al., (1992); Aguirre and Leon (1997) and Ljudmila Benedik and Polona Vrecek (2001) have reported \(^{210}\)Pb content of soils from different parts of the world. Iyengar et al., (1980) have measured \(^{210}\)Pb activity in soil and sand samples of Kalpakkam. Aguirre and Leon (1997) have reported \(^{210}\)Pb activity in soils of Spain which ranges from 15.4 to 820 Bq kg\(^{-1}\). Avadhani et al., (2001) have studied the distribution of \(^{210}\)Pb for Goa region, India and reported mean value was 119.9 Bq kg\(^{-1}\). The \(^{210}\)Pb activity reported for coastal Karnataka and Mangalore environs range from 3.6 to 45.2 Bq kg\(^{-1}\) (Radhakrishna et al., 1993).

**Polonium – 210**

Many investigators have reported on \(^{210}\)Po concentration in soil. Iyengar et al., (1980) measured \(^{210}\)Po activity in soils of Kalpakkam environ. In the US
Atomic Energy Commission report (1980) is presented $^{210}$Po content in various types of soil. In humic soil $^{210}$Po content is found to be approximately 3 times higher than in mineral soils (Berger et al., 1965).

Schuttelkopf and Kiefer (1982) have measured $^{210}$Po activity in soil samples from Black Forest in the south western part of the republic of Germany, a reported high background area. The Polonium - 210 activities of these samples is reported to vary from 33.3 Bq kg$^{-1}$ to 297.1 Bq kg$^{-1}$. Santos et al., (1990) have studied the $^{210}$Po activity in several soil samples from the environment of Pocos de Caldas Plateau (CPIC) in the Minas Gerais State of Brazil, a uranium mining and milling station. For comparison purposes soil samples were also analysed from the control region of Joinville. The activity reported for samples from the surrounding regions of CPIC ranged from 45.9 Bq kg$^{-1}$ to 70.0 Bq kg$^{-1}$ whereas the activity of samples from the control region ranged from 27.3 to 38.3 Bq kg$^{-1}$. Ljudmila Benedik and Polona Vrecek (2001) reported $^{210}$Po in different environmental samples.

Several investigators have reported the activity of $^{210}$Po for Indian soil. Siddappa et al., (1994); Narayana et al., (1995) and Radhakrishna et al., (1996) have studied the $^{210}$Po content in soils of coastal Karnataka and Kaiga and reported the values range from 1.3 Bq kg$^{-1}$ to 45.2 Bq kg$^{-1}$. The $^{210}$Po activity of soil samples of Kalpakkam is reported to be 44.4 Bq kg$^{-1}$ (Iyengar et al., 1980) and of Mysore region to be 7.6 - 37.3 Bq kg$^{-1}$ (Nagaiah et al., 1995). Avadhani et al., (2001) have studied distribution and behaviour of $^{210}$Po in soil samples of Goa, south west coast of India. The activity of $^{210}$Po in surface soil varies from 3.2 Bq kg$^{-1}$ to 186.2 Bq kg$^{-1}$ with a mean value 57.4 Bq kg$^{-1}$. 
Thorium - 232

Mishra and Sadasivan (1971); Myrick et al., (1983); Delaune et al., (1986); Yu-Ming Lin et al., (1987); McAulay and Moran (1988); Descamps and Foulquier (1988); Kalyani et al., (1990); Sutherland and de Jong (1990); Bari et al., (1992); Menon et al., (1992); Rekhakutty et al., (1993); Radhakrishna et al., (1993); Ibrahiem et al., (1993); Selvasekarapandian et al., (1999); Narayana et al., (2001); Navas et al., (2002); Topcuoglu et al., (2003); Ya-xin Yang et al., (2005); Ramli et al., (2005); Nasim Akthar et al., (2005) and Vukovic et al., (2006) have measured the concentration of $^{232}\text{Th}$ in different environmental matrices.

Rao et al., (1983) have reported the activity in marine sediments of Bombay region to be 53.6 Bq kg$^{-1}$. Londhe et al., (1984) have reported 9.3 Bq kg$^{-1}$ in sand samples of Bombay. Descamps and Foulquier (1988) have measured the concentration of $^{232}\text{Th}$ in sediment samples of different rivers of France. The reported range and mean values are 23 - 52 Bq kg$^{-1}$ and 44 Bq kg$^{-1}$ respectively. Bikit et al., (2006) reported the $^{232}\text{Th}$ activity (30.9 Bq kg$^{-1}$) in sediment samples of Danube river. Kalyani et al., (1990) have measured the concentration of $^{232}\text{Th}$ in sediment samples of off Krishna delta collected from different depths and reported an increasing trend in the concentration with depth. Rekhakutty et al., (1993) have reported the equilibrium studies in thorium series in soil and sand samples collected from different parts of India. They have observed equilibrium between different daughter products of thorium. The reported population - weighted world average $^{232}\text{Th}$ concentration in soil (UNSCEAR, 2000) is 45 Bq kg$^{-1}$. As per the UNSCEAR (2000) survey, the activity in Indian soil ranged from 14 - 160 Bq kg$^{-1}$
with a mean value of 64 Bq kg\(^{-1}\). However the concentrations of \(^{232}\)Th in high background radiation areas are significantly higher.

1.6 Radioactivity in aquatic environs

The measurement of radioactivity in potable water allows the determination of the exposure of the population to radiation of the habitual consumption of water. Levels of natural radionuclides in potable water may be increased by a number of human activities. Radionuclides from the nuclear fuel cycle, medical and other uses of radioactive materials may enter potable water supplies (Duenas et al. 1997). The occurrence of these radionuclides gives rise to human internal exposure, directly taken into the body through ingestion and inhalation and indirectly when they are incorporated as part of the human food chain.

\(^{238}\)U is the most abundant (99.28\%) isotopes of natural uranium and is found in all types of rocks and soil in varied concentration. It leaches out from rocks and minerals, mixes with water due to radiochemical reactions and it will be found in trace quantities. Consequently uranium is found in small quantities in dissolved and suspended particulate forms in surface water. Mining and milling of uranium rocks, granite, lignite, monazite sands and minerals such as uranite, comatite and pitchblende may also contribute to the presence of uranium in surface water. In addition, uses of phosphate fertilizers in agriculture also contribute to the presence of uranium in surface waters. Uranium, when present in large amount in drinking water, may lead to harmful biological effects in human beings. It is toxic to kidney and an exposure of about 0.1 mg kg\(^{-1}\) of body weight
of soluble uranium leads to chemical damage of the kidneys (Lussenhop et al., 1958).

$^{226}\text{Ra}$ (T$_{1/2}=1620$ years), a member of $^{238}\text{U}$ decay series, has a high degree of toxicity is a bone seeker, and as an alpha emitter has a high potential biological damage. When $^{226}\text{Ra}$ is taken into the body, due to its similar metabolic behavior to that of calcium an appreciable fraction is deposited in bone. More than 70% of the radium is contained in bone, the remaining fraction being distributed rather uniformly in soft tissues. It has been reported that the annual effective dose equivalent resulted from $^{226}\text{Ra}$ intake in normal areas was about 7 $\mu$Sv. The contribution of drinking water to the total intake is generally small when the drinking water is drawn from surface water. However, in ground water supplies (Which in many countries serve a large portion of the population) $^{226}\text{Ra}$ concentrations vary widely (UNSCEAR, 1988).

$^{222}\text{Rn}$ a noble radioactive gas produced by decay of $^{226}\text{Ra}$, is a member of $^{238}\text{U}$ series. Uranium, a source of radon is present in almost all types of rocks, soils, plants and ground waters. Natural water contains dissolved radon from the uranium series present in soil and rocks. The occurrence of radon in ground water can be reasonably related to the uranium contents of the bedrocks and it can easily enter into the interacting ground water by the effect of lithostatic pressure (Tosconi et al., 2001). $^{222}\text{Rn}$ in water can get released to indoor air when used in showers, humidifiers, cloths washers, dish washers, cooking and so on. Exposure to water borne $^{222}\text{Rn}$ may occur by ingestion (drinking water containing $^{222}\text{Rn}$) and by inhalation (breathing $^{222}\text{Rn}$ gas which has been released from household water). Both mechanisms pose potential health hazards (Yu et al., 1994; Barnett
et al., 1995; Tayyeb et al., 1998). Exposure to indoor radon and its progeny is believed to be associated with a potential health risk of lung cancer (Galan Lopez et al., 2004).

$^{210}\text{Pb}$ and $^{210}\text{Po}$, also the members of $^{238}\text{U}$ decay series are found to be present in almost all environmental matrices. The main source of these radionuclides in the environment is the exhalation of $^{222}\text{Rn}$ from the earth’s surface released into the atmosphere. In the atmosphere, $^{222}\text{Rn}$ decays to $^{210}\text{Pb}$ and $^{210}\text{Bi}$ via three short lived radionuclides, which readily attach themselves to atmospheric particulate and are transported back to the earth surface by natural wet and dry precipitation. The concentration of $^{210}\text{Pb}$ and $^{210}\text{Po}$ in surface water originate from various sources such as rain, leaching of surrounding material, suspended solids. Concentrations of these radionuclides in groundwater are mainly due to decay of dissolved $^{222}\text{Rn}$. Therefore, it is important to study the concentration and distribution of these radionuclides in aquatic environment especially in water samples of different origins. In view of this, systematic studies on measurement of radionuclides are important because the ground water and river water are main source of potable water in the study region due to inadequate supply of treated water.

1.6.1 Natural radionuclide concentrations in surface water

Several investigators have reported the concentration of natural radionuclides in potable water of different origin such as bore well; open well, tap water and stream water. Edington (1965) and Cothern and Lapenbusch (1983)
have reported the uranium concentration in potable water of U.S. They reported that the activity of uranium in potable water vary in the range 0.01 to 3.04 μg l⁻¹. The concentration of uranium in river water has been reported by Frissel et al., 1989; Martinez Aguirre and Garcia Leon (1992) and Vukovic et al., (1996) Rodriguez-Alvarez and Sanchez (1999), Pujol and Sanchez Cabeza (2000). The reported values are 0.08-2.60 μg l⁻¹ for river water samples. Cuculic et al., (2006) reported for Karka river estuary (2.1 μg l⁻¹). In Indian environs, Nagaiah et al., (1996) and Singh et al., (1996) have reported the uranium concentrations in the range 1.1-4.8 μg l⁻¹ for Indian rivers. U can be present in river water over a wide range of concentration between less than 0.02 μg l⁻¹ (which is the practical detection limit) and more than 30 μg l⁻¹, average concentration being taken as 0.6 μg l⁻¹ (Durrance, 1986, USEPA, 2000).

The studies on ²²⁶Ra concentrations in rivers have been carried out and reported by Miyake et al. (1964); Strain and Watson (1979); Cothem et al.(1986); Martinez Aguirre and Garcia Leon (1992); Othman and Yassine (1996); Sanchez and Rodriguez-Alvarez (1999), Pujol and Sanchez Cabeza (2000) for river water of different parts of the world. The literature available on the study of ²¹⁰Pb in surface water is limited. Holtzman (1964) reported for the distribution of ²¹⁰Pb concentrations in surface water of Illinois. Duenas et al., (1999) have reported the activity of ²²⁶Ra in potable water (bottled water) of European countries and the activity vary in the range 3 mBq l⁻¹ to 2185 mBq l⁻¹. Iyengar et al. (1989) have studied the ²²⁶Ra concentrations in surface water of east coast of India.

Carvalho (1995) has reported the activity of ²¹⁰Pb and ²¹⁰Po in drinking water for Portugal environment. Shahul Hameed et al, (1997) have carried out
Introduction

studies on $^{210}$Pb concentration in the ecosystem of Kaveri river. Iyengar et al., (1979, 1982) have carried out extensive measurements on the concentrations of $^{210}$Po for coastal sea water of Bay of Bengal. Sastry et al., (1996) have reported the activity of $^{226}$Ra, $^{210}$Pb and $^{210}$Po in potable water. The overall reported world average values for $^{226}$Ra, $^{210}$Pb and $^{210}$Po in potable water are found to be 0.5 mBq l$^{-1}$, 10 mBq l$^{-1}$ and 5 mBq l$^{-1}$ respectively (UNSCEAR, 1999).

1.7 Influence of Geology on radioactivity

The presence of radioactive elements in soils and sediments is strongly conditioned by those existing in the parent material. Although the percentage of an element can vary in a given rock as a function of the process to which it has been subject. It is commonly accepted that granites contain high concentration of uranium, thorium and potassium. The first two were incorporated into the rocks in the crystallization of the last magmas and residual solutions, since their large ionic radii stop them from crystallizing out in the early silicates (Ivanovich and Harmon, 1982). Potassium-40 is due to the high potassium content in the feldspars, which form part of the granites. In metamorphic rocks, the uranium, thorium and potassium abundances depend mainly on migration of the nuclides during metamorphism. Since the high temperatures and pressures of this phenomenon provoke the loss of fluids and increase the mobility of the elements. Lastly, soils on top of sedimentary materials present diverse concentrations of these elements depending on those present in the eroded substrate and on the phenomena
Chapter 1

associated with sedimentation.

Baeza et al. (1997) and Ramli et al. (2001) have studied the geological influence on terrestrial gamma dose rate for the province of Caceres and Malaysian State of Johore respectively and reported that there is a strong link between terrestrial gamma radiation dose-rates and underlying geology. Baeza et al., (1995) have extensively studied the activities of $^{226}$Ra, $^{232}$Th and $^{40}$K in soils of different origin and reported that the mean value of the activities of these radionuclides in granitic soil is significantly higher than for the metamorphic soils. The activity of these radionuclides in sedimentary soils lies between the other two types.

1.8 Influence of Physico-chemical parameters

The measurement of radionuclide transfer and accumulation in various ecosystems is important for the assessment and prediction of risk to biota and for the formation of effective emergency plans (Krouglov et al. 1998). Distribution of radionuclides in various environmental matrices depends on the nature of radionuclides itself and the site specific characteristics such as soil type and its physico-chemical properties and variation in plant cover. In order to understand the dispersion, transport and biological impact of radionuclides in the environment, knowledge about their specification, i.e, distribution between different physico-chemical forms is also essential.

Influence of physico-chemical properties of soil and sediment like pH and organic matter on the deposition of fall-out radionuclides and their soil-to-plant
transfer coefficients have reported by various investigators (Bergeijk et al., 1992; Shenber and Eriksson, 1993; Schuch et al. 1994; Baeza et al., 1995a; Lee et al., 1997; Kruglov et al. 1998; Bygaart and Protz, 1999; Baeza et al. 1999). The content of organic matter in the surface soil plays a major role in fixing and retention of fallout radionuclides. Due to its large cation exchange capacity, the organic matter content is a characteristic parameter that has a great influence on the dynamics of fallout-originated radionuclides in soil (Livens and Baxter, 1988; Alberts et al., 1989; Pavlotskaya et al., 1991; Baeza et al., 1995). Lee et al. (1997) and Baeza et al., 1995 have observed significant correlation between organic matter content with $^{137}$Cs activity in surface soil.

1.9 Geological background of the study area

Since, local geology and geological parameters play a major role in the distribution of natural radionuclides in the environment, brief idea of geological background of the study region is given in the following sections.

1.9.1 Geology of the study area

*Kali River*

The Kali River originates in the Western ghats, flows westward for 184 km and discharges into the Arabian sea along coastal Karnataka. The lithology of the study area consists largely of greywackes in the upstream catchment area, whereas tonalitic gneisses occur in the downstream segment. Patches of
limestone and ultramofics are the minor rock types. The principal rock types found in the drainage basin are pre-Cambrian gneisses, granites, granite gneisses, schists and karnockites. Laterities are prevalent in the coast which formed mainly by the chemical weathering of tonalitic gneisses, (Balakrishna et al., 2001, Kidwai et al., 1981).

Sharavathi River

The Sharavathi river originates in Western ghats, flow due east or northeast initially, then take a sudden turn to the west, flow down the steep western slopes of the ghats and after meandering through the short plateaus join the sea passing through the coastal plain. The length of the Sharavathi river is 128 km. The Sharavathi river basin consists of greywacke, dolamite, quartzite, meta volcanics, laterities, tonalitic gneisses and local ferruginous chert are noticed. The Western ghats provide a principal geographical barrier in the path of the Arabian sea branch. The ghats are prominent with steep western scrap face and gently inclined surface in the eastern side. Western ghats consisting of Dharwarian metamorphosed rocks of gneisses, schists, phyllites, quartzite and chlorite schists (Krishnan, 1968).

Netravathi River

The Netravathi river originates in Charmadi ghats, which is part of the Western Ghats flowing westward for 94 km and discharges into the Arabian Sea along coastal Karnataka. The Netravathi river drainage basin mainly a
metamorphic terrain consists of pre-Cambrian granite gneisses, dolerites, ferruginous quartzites, felsic and mafic dykes, meta volcanics, metasediments and chamockite. In the lower reaches the river drains plateaus covered with laterite and tertiaries (Kidwai et al., 1981, Balasubrahmanyan, 1978).

These rivers in the Western ghats region generally originate at an elevation ranging from 400 meters to 1,600 meters above the mean sea level, close to the Western ghats ridge. The rivers are very steep in the upper reaches and fairly steep in the middle reaches. It is only near the sea that the rivers have relatively flat gradients and some sort of flood plain. The catchment areas of these rivers are rich in mineral resources, such as manganese in Supa taluk, low-grade iron ore around Karwar, Kumta and Honnavar, limestone in the central part around Yellapur and granite, lime-shells, sand and clay throughout the region.

1.10 Aim and Scope

The main objective of the present investigation is to study the transport of radionuclides in the major rivers, Kali, Sharavathi and Netravathi which are originating from Western Ghats and join Arabian sea. Attempts were made to trace the source and processes that are responsible for the newly discovered monazite patches near Ullal beach area. Though the presence of monazite deposit in these areas has been well established, systematic studies on the sources and geochemical processes leading to the formation of monazite deposits are sparse. Samples were collected in different seasons of a year and analysed to study the seasonal variations in radionuclide concentrations.
The study was carried out systematically on the concentration of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in the rocks of catchment areas of the rivers. Any enhanced concentration of the above natural radionuclides in sediments and rocks in catchment areas of the rivers would throw light on release of these radionuclides by weathering and transport by rivers and the subsequent deposition in the adjacent beach areas. A more extensive and systematic studies were carried out on water, soil and sediment samples of the three major rivers of coastal Karnataka to throw light on transport and enrichment of radionuclides in the region. Radiation hazards associated with natural radionuclides in soil, sediment and rocks were evaluated using standard relations.

The external gamma absorbed dose rates in air prevailing in the riverbanks were measured using a plastic scintillometer. Natural samples of riverbank soil, sediment and river basin rocks were collected and processed following standard procedure (EML procedure manual, 1983; IAEA Technical reports, 1990). The concentration of $^{226}\text{Ra}$, $^{232}\text{Th}$ and $^{40}\text{K}$ in riverbank soil, sediment and rock samples were precisely measured by gamma spectrometry method. A 5 inch X 5 inch Telydyne flat type NaI(Tl) detector along with associated electronics was employed for measuring the gamma activity. The detector efficiencies were measured using radioactive standard sources procured from the International Atomic Energy Agency (IAEA), Vienna.

The riverbank soil, sediment and water samples were analysed for the concentration of $^{210}\text{Po}$ and $^{210}\text{Pb}$. The concentration of alpha active radionuclide $^{210}\text{Po}$ was estimated by chemical deposition of this radionuclide onto silver disc and subsequent counting ZnS(Ag) alpha counter. The solution, after plating, was
preserved and re-plated after one year to estimate the activity of $^{210}\text{Pb}$. The concentration of Uranium in water was measured employing Laser Fluorimeter. Water radon concentrations were measured using an Alpha Guard PQ2000Pro (Genitron GmbH) equipped with an appropriate unit (Aquakit), following a protocol proposed by the manufacturer. Dose due to ingestion of these river water are also estimated.

The annual intake of radionuclides by the population of the region has been evaluated. The internal dose resulting from intake of radionuclides through ingestion of uranium and radon has been estimated and the results are compared with the data reported for other environs.

The data collected on various aspects cited above are carefully analysed and the results are discussed with the literature values reported for other environs and possible conclusions are drawn. The results of these systematic investigations, which constitute the first ever report on the radiation level, radionuclide distribution and transport of radionuclides in the riverine environs of major rivers of coastal Karnataka are presented in this thesis.

1.11 The Thesis

The thesis is divided into five chapters including, this introduction, which constitutes first chapter. Chapter 2 deals with the details of experimental techniques followed and the instruments used in the present investigation. Details of the instruments like Portable plastic scintillometer, alpha counting system, NaI (TI) spectrometer, Laser Fluorimeter and Alpha guard used to carryout the present
work and their salient features are discussed in this chapter.

Chapter 3, provides a detailed discussion on selection of the sampling stations, collection of samples, sample processing and radiochemical methods followed to separate various radionuclides. The methods employed to calculate the activities of radionuclides in the environmental samples from measured counts and estimation of radiation hazard to the population of the region are discussed in this chapter.

The results obtained in the present work are discussed in Chapter 4. They include gamma absorbed dose rates in air and the distribution and transport of natural radionuclide in riverine environments of coastal Karnataka. Radiation hazard parameters associated with the natural radionuclides were calculated and compared with the international recommended values. The seasonal variations in these radionuclide concentrations are also discussed in this chapter. The effects of physico-chemical properties of sediment on these radionuclides are also studied. Natural radionuclide concentrations in surface water were measured and the ingestion doses through drinking water to the population were discussed in this chapter.

Important conclusions drawn from the present investigations are summarised in Chapter 5. Suggestions for future studies in this important field are also highlighted.