CHAPTER 11

HIGH NATURAL RADIOACTIVITY IN SOME TROPICAL LANDS – BOON OR BANE?

Natural radiation has been in existence from the time planet earth started to form and there are about 60 radionuclides present in nature. These are found naturally in air, water, soil, rocks, minerals and food. About 82% of this environmental radiation is from natural sources, the most important of which is radon.

The United Nations Scientific Committee on Effects of Atomic Radiation Report (UNSCEAR, 2000) states that while the natural background radiation is the largest contributor to human exposure, the worldwide average annual effective dose per capita is 2.4 mSv. Some radionuclides such as ²³⁸U, ²³²Th and ⁴⁰K among others are important as naturally occurring radionuclides and geological and geochemical processes play the major role in their distribution in nature. These are found in some minerals such as monazites and zircons. Some areas of the world have anomalously high background radiation and these are called high background areas (HBRAs) (Table 11.1). In these areas, the geology and geochemistry of the source rocks and minerals have the greatest influence on the localization high natural radiation.

In radioactive material measurement, the International System of Units (SI) is generally used. However, there are several other units that are used in the literature and this causes a certain amount of confusion among the readers. The common reference measurements are radioactivity and radiation dose. These units are often referred to in both their SI units (Becquerel and sievert) and the traditional units (curie and rem). The University of Ottawa (2007) has created the conversion between the two systems as given in http://www.uottawa.ca/services/ehss/ionizconversion.htm.

Table 11.2 shows the mineral sources of the naturally occurring radioactive materials. Extreme HBRAs are found in Guarapari (Brazil), Southwest France, Ramsar (Iran), Kerala coast (India) and Yangjiang (China) (Table

| 11.1). Of these | the | majority | of | HBRAs | are | found | in | tropical, | , arid | and |
|-----------------|-----|----------|----|-------|-----|-------|----|-----------|--------|-----|
| semiarid areas. | | | | | | | | | | |

| Area | Mean (mGy/year) | Maximum (mGy/year) |
|-------------------|--------------------|-----------------------|
| Ramsar, Iran | 10.21 | (260) |
| Guarapari, Brazil | 5.52 | (35) |
| Kerala, India | 3.82 | (35) |
| Yangjiang, China | 3.51 | (5.4) |
| Hong Kong, China | 0.67 | (1.00) |
| Norway | 0.63 | (10.5) |
| France | 0.60 | (2.20) |
| China | 0.54 | (3.01) |
| Italy | 0.50 | (4.38) |
| World average | 0.50 | |
| India | 0.48 | (9.6) |
| Germany | 0.48 | (3.8) |
| Japan | 0.43 | (1.26) |
| USA | 0.40 | (0.88) |
| Austria | 0.37 | (1.34) |
| Ireland | 0.36 | (1.58) |
| Denmark | 0.33 | (0.45) |

Table 11.1. Natural Radioactivity in some selected areas (based on the UNSCEAR Report, 1993)

> ¹High Levels of Natural Radiation 1996, M. Sohrabi, p 57-68 Elsevier Science B.V. 1997) ²1982 UNSCEAR report

Terrestrial Radiation in Beach Sands in Brazil

In certain beaches in Brazil, monazite sand deposits are abundant. The external radiation levels on these black beach sands range up to 5 mrad/hr (50µ Gy/hr) and is nearly 400 times the normal background in USA. The Brazilian coastal sands have several minerals among which are monazite, rutile, ilmenite, zircon, cassiterite, thorianite, pyrochlore and niobate- tantalite. Ilmenite (FeTiO₃)-96%, rutile (TiO₂) - 0.5%, zircon (ZrSiO₄)-2.5% and monazite- $REE_3(PO_4)$ rare earth phosphate- 3% comprise the main minerals. The monazite from the beach sands of Brazil contain up to 39% cerium oxide (CeO₂), 16% of lanthanum oxide (La₂O₃), 14% of neodymium oxides (Nd₂O₃), 5% yttrium, 6% thorium dioxide (ThO₂) and 0.31% uranium oxide and phosphates.

Fujinami et al. (1999) studied the exposure rates from terrestrial radiation at Guarapari and Meaipe in Brazil and made measurements of absorbed dose rate in air one meter above the ground and on the surface with a portable NaI (Tl) scintillation survey meter. The highest dose rate was 0.6 μ Gy/h at one meter height in the streets and the highest rate at one meter height on the beach was 6.2 μ Gy/h and on the surface of the beach it was 15 μ Gy/h. Interestingly the beach at "Areia Preta"-meaning black sand, is highly sought after by Brazilian people for their alleged health benefits (Eisenbud, 1973). In an area on the beach at "Areia Preta" where black sand was not found, the absorbed dose rate was 0.04 μ Gy/h and in the Meio Beach where white sands are found and separated from the Areita Preta beach, the dose rate was 0.09 μ Gy/h.

| Ore | Main Constituent | Typical Active Concentrations (KBq/kg) | | | |
|------------------------|---------------------|---|-------------|--|--|
| | | Thorium-232 | Uranium-238 | | |
| Phosphate rock | Calcium phosphate | 0.1 | 1.5 | | |
| Ilmenite | Iron titanium oxide | 1 | 2 | | |
| Rutile | Titanium dioxide | 0.2 | 0.2 | | |
| Rare earth concentrate | Cerium oxide | 5 | 0.1 | | |
| Baddeleyite | Zirconium oxide | 1 | 10 | | |
| Zircon | Zirconium silicate | 0.6 | 3 | | |
| Pyrochlore | Niobium oxide | 80 | 10 | | |
| Monazite | Cerium phosphate | 300 | 40 | | |

 Table 11.2. Ores containing naturally occurring radioactive materials (Hipkin and Shaw, 1999)

At Meaipe beach, the highest dose rate in the street area was 5.2 μ Gy/h while on the surface of the beach near the breakwater the highest value reached a level of 32 μ Gy/h.

Lauria and Godoy (2002), reported anomalously high ²²⁸Ra concentrations up to 2 Bq/L in waters of a coastal lagoon close to a monazite sand separation plant in Buena lagoon area in the Rio de Janeiro state in Brazil. Even though it had earlier been suggested that this was caused by mineral processing, it was later concluded that the abnormal radium concentrations had a natural origin from springs at the lagoon head with high ²²⁸Ra and ²²⁶Ra concentrations.

Strong relationship among radium and light rare earth elements (LREEs), ²²⁸Ra/²²⁶Ra activity ratio and the REE pattern were suggestive of monazites as the main source of nuclides in the water. It was suggested that low pH

(3.7) and high salinity (14‰) caused a disturbance of the chemical stability of monazites. There was a relatively low mobility of thorium but a high mobility of radium and LREEs.

In other parts of the world, the radium concentration in surface waters normally range from 0.01 to 0.1 Bq/L (Iyengar, 1990). In the Buena lagoon area in Brazil, the ²²⁸Ra concentration in the spring waters varied between, 1.7 and 2.5 Bq/L and for ²²⁶Ra it was 0.5 Bq/L. The LREE concentrations in the water were also very high with La = 50 μ g/L, Ce = 110 μ g/L and Nd = 60 μ g/L.

The recent study by Dias da Cunha et al. (2004) on the airborne particles in the Buena village using lichen samples showed that during the last 15 years, the inhabitants of the village had been exposed to monazite particles. The result form aerosols and lichens also suggested that the swampy area is a source of ²²⁶Ra and ²¹⁰Pb-bearing particles in addition to the monazite dust.

In Morro do Ferro in the Pocos de Caldas Plateau of Brazil is an uninhabited hill 0.35 km^2 in an area consisting of large rare earth ore deposits that rise to above 250 meters from the surrounding area. The radiation levels here are 1 to 2 mrad/hr (0.01 to 0.02 mGy/hr) over the 0.35 km² area and are between 100 to 200 times the non- elevated natural gamma radiation background (Paschoa and Godoy, 2002).

Monazite Rich Beach Sands of India

As shown in Table 11.3 some of the beach deposits of India, notably the Kerala coast Tamil Nadu coast, Bhimilipatanam (Andhra Pradesh) coast and Chhatrapur (Orissa) coast are prominent HBRAs. As in the case of Brazil, the high levels of natural radiation is due to the abundance of monazite along with other heavy minerals such as ilmenite, rutile, zircon and garnet among others. The coastal regions of peninsular India have therefore received great international attention on account of the high natural radioactivity (Paul et al., 1998).

Along the 570 km long coastline of Kerala in the Southwest of India, there are major deposits of monazite-rich heavy minerals with very high natural radiation. The monazite deposits are larger than those in Brazil and the dose from external radiation, is on the average similar to doses reported in

Brazil, 5-6 mGy/yr. Individual doses up to 32.6 mGy/yr, however, have also been reported.

Table 11.3. Comparison of radiation dose rates of Chhatrapur beach placer deposit area and other high background radiation areas (HBRAs) (after Mohanty et al., 2004a)

| Area (Country) | Characteristics of area | Absorbed dose rate in air (nGy/h) |
|------------------------------------|----------------------------------|--------------------------------------|
| Guarapari (Brazil) | Monazite sands | 90-90,000 |
| Morro Do Forro (Brazil) | Th-rich alkaline intru- sives | 2800 |
| Yangjiang (China) | Monazite sands | 370 |
| Nile Delta (Egypt) | Monazite sands | 20-400 |
| Southwest (France) | Uranium minerals | 10-10,000 |
| Ramsar (Iran) | 226Ra in spring water | 70-17,000 |
| Cox's Bazar (Bangladesh) India | Monazite sands | 260-400 |
| Kerala coast | Monazite sands | 200-4000 |
| Ullal (Karnataka) | Monazite sands | 2100 |
| Tamil Nadu coast | Monazite sands | 200-4000 |
| Kudiraimozhi (Tamil Nadu) | Monazite sands | 200-900 |
| Bhimilipatanam (Andhra Pradesh) | Monazite sands | 200-3000 |
| Kalpakkam | Monazite sands | 3500 |
| (Tamil Nadu) | | |
| Ayirmamthengu (Kerala) | Monazite sands | 200-1400 |
| Neendakara (Kerala) | Monazite sands | 200-3000 |
| Chhatrapur (Orissa) | Monazite sands | 375-5000 |

Paul et al. (1998) studied 4 different locations (Fig 11.1) in coastal India for their natural radiation and the results are shown in Table 11.4. The radiation and fields ranged from 200 to 3000 nGy/h at the different sites, with large radiations in the fields, in some cases up to an order of magnitude. The Neendakara and Bhimilipatnam deposits of the Kerala coast had the highest levels and were associated with the beach placer deposits. It was also observed that with the onset of tropical monsoon, higher deposition of heavy minerals takes place and hence the enhanced natural radiation at these beaches.

Mohanty et al. (2004a, 2004b) carried out some studies on the natural radioactivity of high background radiation areas on the eastern coast of Orissa, India. They found that the average activity concentrations of radioactive elements such as 232 Th, 238 U, and 40 K were 2825±50; 350±20 and 180±25 Bq/kg, respectively for the bulk sand samples. The absorbed gamma dose rates in air due to the naturally occurring radionuclides varied from 650 to 3150 nGy/h with a mean value of 1925±718 nGy/h This is very much higher than the world average value of 55 nGy/h (UNSCEAR, 1988). The presence of 232 Th contributed a maximum of 91% (1750 nGy/h) to total absorbed dose rate in air followed by 238 U of 8.5% (165 nGy/h) and by 40 K of 0.5% (8 nGy/h). The annual external effective dose rates for the region varied from 0.78 to 3.86 mSv/yr with a mean value of 2.36±0.88 mSv/yr.

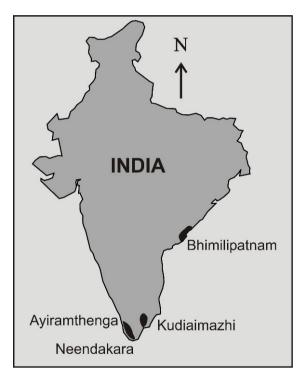


Fig. 11.1. Locations of some areas of high natural radioactivity in India (Paul et al., 1998)

This area is therefore a high background radiation area similar to other radiation areas of southern and southwestern coastal regions of India (Mohanty et al., 2004b). Similar results were obtained by Mohanty et al. (2004a) for the Chhatrapur beach placer deposit, also of Orissa, India. Here too, the enhanced levels of radiation were due to monazite and zircon sands.

| Site | No. of Locations Monitored | Radiation Field | (nGyh ⁻¹) |
|---------------|-------------------------------|-----------------|-----------------------|
| | | Range | Mean±SD |
| Ayiramthengu | 64 | 200-1400 | 300±270 |
| Neendakara | 64 | 200-3000 | 500±330 |
| Kudiraimozhi | 43 | 200-900 | 370±140 |
| Bhimilipatnam | 40 | 200-3000 | 430±330 |

Table 11.4. External gamma radiation fields (Mohanty et al., 2004a)

High Natural Radioactivity of the Minjingu Phosphate Mine, Tanzania

The Minjingu phosphate mine in northern Tanzania located to the east of the saline lake Manyura at Arusha is a HBRA. Phosphate deposits are known to contain significant traces of the long-lived radioelements ²³⁸U. ²³²Th and ⁴⁰K. These contribute to the internal and external radiations to man (Halbert et al., 1990; Bettencourt et al., 1990). There are about 2000 inhabitants in the village of Minjingu and these people may be exposed to naturally occurring radionuclides from the emissions caused by open dry phosphate mining (Banzi et al., 2000). Bianconi (1987) had earlier reported that the deposit has uranium activity with a maximum concentration of 800 mg/kg U_3O_8 . It had been observed that the average dose rate in air of 1415 nGy/h is 28 times the average terrestrial dose rate of 50 nGy/h in air worldwide (UNSCEAR, 1982). Further, the estimated average effective dose over 5 years at Minjingu is about 12 mSv/y which is 12 times the allowed average dose limit of 1 mSv/y for members of the public (IAEA, 1996). Based on these observations the Minjingu phosphate mine is rated as an area ranked with the highest natural radiation background recorded in the world (UNSCEAR, 1982).

Very High Natural Radiation in Ramsar, Iran

UNSCEAR (2000) has classified Ramsar, a city located in northern Iran, as an area with one of the highest natural radiation levels in the world. The city lies between the Elburz Mountains and the Caspian Sea at an average elevation near sea level, and is a popular tourist resort. The natural radiation in some locations at Ramsar is 55-200 times higher than the normal background areas. The annual exposure levels in areas with elevated levels of natural radiation in Ramsar reach values as high as 260 mGy/y and

average exposure rates are about 10 mGy/y for a population of about 2000 residents (Mortazavi et al., 2001).

The origin of the high levels of natural radioactivity is shown in Figure 11.2. The radioactivity in the high level natural radiation areas of Ramsar is due to ²²⁶Ra and its decay products brought up to the earth's surface by the water of hot springs, of which there are at least 9 with different concentrations of radium.

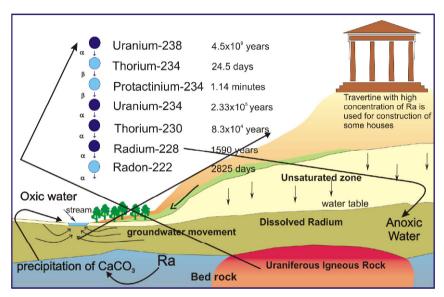


Fig. 11.2. Origin of high natural radioactivity in Ramsar, Iran (Mortazavi et al., 2001)

When the groundwater reaches the surface at hot spring locations, travertine, a calcium carbonate (CaCO₃) mineral precipitates out of solution with dissolved radium substituting for calcium in the mineral as RaCO₃. High concentrations of radium carbonate are found in the residual deposits (up to 30 m thick) of the hot springs. These materials are often used as building materials by the residents of the area. The gamma radiation at waist level in these houses reaches up to about 20 μ Sv/h. The walls of the bed rooms (made of sedimentary rocks from the hot springs) of these dwellings also show dose rates as high as 143 μ Gy/h. In spite of the fact that the levels of natural radiation in these areas is up to 200 times higher than normal background levels, nearly all inhabitants still live in their old ancestral dwellings, without any ill effects (Mortazavi, 2003).

High Natural Background Radiation in Yangjiang, China

The Yangjiang area in China is also classified as a HBRA and has dose rates about 300-400 mrad/yr (3-4 mGy/yr). As in the case of Kerala and Orissa in India and Guarapari in Brazil, the main source of the radiation is monazite containing Th, U, and Ra.

The Oklo Natural Reactor

A most interesting geological phenomenon occurred during the Proterozoic period (around 1800 my) when a natural nuclear reactor was formed at Oklo in Gabon, West Africa. This was discovered quite by accident, when in 1972 some French scientists working on a shipment of uranium from Oklo observed that it had a much smaller proportion (as low as 0.440%) of ²³⁵U than normal (Cowan, 1976). Natural uranium always has the identical isotopic composition ²³⁸U (99.27%), ²³⁵U (0.72%) and traces of ²³⁴U. Of these, only ²³⁵U can take part in chain reactions and it is therefore very closely studied.

Since the ²³⁵U isotope is present only in low abundance (0.72%), enrichment up to 3% or greater is required for use in commercial nuclear reactors. During the Proterozoic period around 2000 my ago however, this critical level of 3% was present in nature and this was sufficient for a sustained nuclear reaction. The abundant groundwater present at the Oklo site served as a moderator, reflector and cooler for the fission reaction. The presence of neodymium provided strong evidence that the natural reactor had indeed operated and that it had consumed six tons of uranium over several hundred thousand years releasing 15,000 MW years of energy.

Lovelock (1988), described the geological process that acted as a natural reactor (Figure 11.3). Uranium is known to be readily soluble in the presence of oxygen and during the Proterozoic times, when oxygen was present, the groundwater was oxidizing. Under these conditions, uranium dissolved yielding the uranyl ion which was present in trace quantities. At the Oklo site, a stream had flowed into an algal mat which had the microorganisms having the capability of concentrating uranium specifically. The efficiency of this process of uranium concentration by the microorganisms was so high that uranium oxide was deposited in the pure state enabling a nuclear reaction to start. The water present enables the slowing and reflecting of neutrons so that it is controlled and self-regulating (Lovelock, 1988).

The unique feature about the Oklo natural reactor is that it was functional as far back as 2000 my ago and that it was a complete natural geochemical and microbiological phenomenon that was entirely self regulating. It was after a period of nearly 2 billion years that, modern scientists were able to duplicate nature's natural reactor at Oklo and produce the first man-made nuclear reactor.

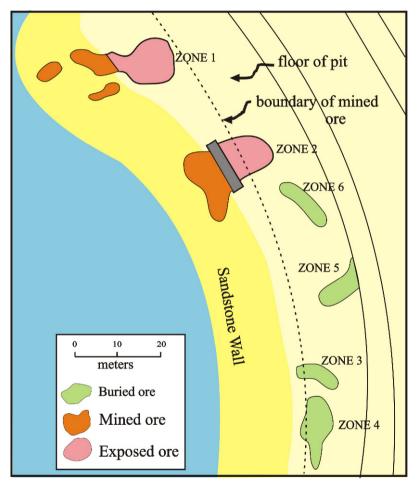


Fig. 11.3. Disposition of six of the Oklo, 2 billion year old natural reactors (from Anomalies in Geology, Science Frontiers Jan-Feb 1999 (Corliss, 1999)

RADIATION AND HEALTH

Radiation is a form of energy and which comes from man-made sources such as X-ray machines or from natural sources such as the sun, outer space and radioactive minerals in the environment. Of the naturally occurring elements, some, known as radionuclides emit energy in the form of alpha rays, beta rays and gamma rays. These rays interact with surrounding matter which results in ionization. When this type of ionization radiation enters the human body, it is referred to as internal exposure, as opposed to external exposure from radiation outside the body, such as from the sun or from radioactive materials. When the radiation is absorbed in the human body, the resulting chemical reactions can cause adverse health effects which may be observed in some cases after many years. These adverse health effects take the form of skin diseases, cancer and even death. About 80% of human exposure is from natural sources while the balance is from man-made sources (Table 11.5).

It is therefore the natural background which has evoked a great deal of interest in environmental health. Figure 11.4 illustrates the importance of the natural background radiation as against anthropogenic causes. The radiation encountered from natural radiation is several orders of magnitude compared to those emitted by nuclear fall outs, and medical diagnostics. The intriguing question therefore is "what is the effect of natural background radiation on human health?"

It is interesting to note that in all the case studies such as Guarapari (Brazil), Ramsar (Iran), Yangjiang (China), Orissa and Kerala (India), the adverse health effects appear to be very low. On the contrary, in some cases the population exposed to these HBRAs appears even healthier with higher life spans as compared to the control areas.

An epidemiological survey carried out in the Yangjiang HBRA in China has revealed that there are no observed adverse health effects based on cancer mortality data from 1,008,769 person-years in HBRA and 995,070 person-years in control areas (Zha et al., 1996). The study also included hereditary diseases and congenital malformations, human chromosome aberrations and immune functions.

Between 1970 and 1986 the Yangjiang HBRA population (background radiation 5.5 mSv/y) was compared with people living in two adjacent lowbackground counties Enping and Taishan (background radiation 2.1 mSv/y), the cohorts being 74,000 (Yangjiang) and 77,000 (Enping and Taishan). For the age group of 10 to 79, the results showed that the general cancer mortality was 14.6% less in Yangjiang compared with Enping and Taishan. The leukemia mortality was 16% lower in men and 60% lower in women from Yangjiang.

 Table 11.5. Man's exposure to ionizing radiation (source: Australian Radiation Protection and Nuclear Safety Agency, 2004)

| Source of Exposure | Exposure | | | |
|---|-------------------|--|--|--|
| Natural Radiation (Terrestrial and Airborne) | 1.2 mSv per year | | | |
| Natural Radiation (Cosmic radiation at sea level) | 0.3 mSv per year | | | |
| Total Natural Radiation | 1.5 mSv per year | | | |
| Seven Hour Aeroplane Flight | 0.05 mSv | | | |
| Chest X-Ray | 0.04 mSv | | | |
| Nuclear Fallout (From atmospheric tests in 50's & 60's) | 0.02 mSv per Year | | | |
| Chernobyl (People living in Control Zones near Chernobyl) 10 mSv per year | | | | |
| Cosmic Radiation Exposure of Domestic Airline Pilot | 2 mSv per year | | | |

In Ramsar, Iran where the natural radiation, as explained earlier was very high, some epidemiological studies (Mortazavi, 2003), have shown no significant adverse health effects. Preliminary results on chromosome aberrations showed no significant difference among those who lived in houses with extraordinarily elevated levels of natural radiation. A cytogenetical study was carried out on 21 healthy inhabitants of the high level maturation radiation areas and 14 residents of a nearby control area. There was no observed positive correlation between the frequency of chromosome aberrations and cumulative dose of the inhabitants. In the case of hematological parameters too, there was no statistically significant alteration in the very high level natural radiation areas (VHLNRAs) of Ramsar as compared to those living in a neighbouring control area.

High doses of ionizing radiation are known to suppress the activity of the immune system, whereas low-level whole body irradiation (WBI) can enhance the immunological response. This was tested in Ramsar (Mortazavi, 2003) and the preliminary results indicated that relatively high doses are not immunosuppressive. The results however, were not conclusive.

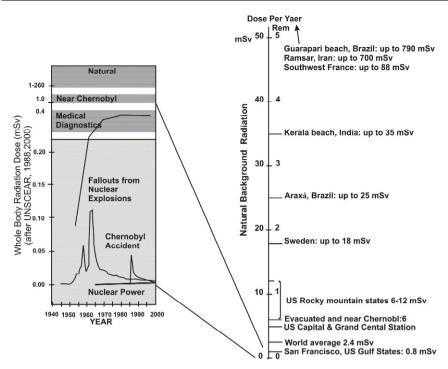


Fig. 11.4. Radiation dose received from natural and human sources (Jaworowski, 2000)

Adaptation to high levels of natural radiation has been the subject of many studies (Olivieri et al., 1984; Sanderson and Morely, 1986; Kelsey et al., 1991; Rigand et al., 1993; Joiner 1994; Azzam et al., 1994). In order to assess the possible induction of adaptive response in the inhabitants of HLNRAS of Ramsar, blood samples of the residents and a nearby control area were exposed to a challenge dose, of 1.5 Gy (natural radiation was used as the adapting dose). The results showed 56% fewer MCAPC (mean number of chromosomal aberrations per cell) caused by the challenge dose among HLNRA s as compared to lymphocytes from residents of low background areas (Ghiassi-nejad et al., 2002). It had been reported that the Ramsar results suggest that chronic low dose radiation may not only reduce mortality form all causes and cancer mortality, but may also be protective against accidental high dose radiation (Pollycove and Feinendegen, 2001).

In Kerala India, another area of high background natural radiation, it has been reported that over 140,000 inhabitants receive an annual average dose of 15-25 mbq (Kesavan, 1996). The average life span of the inhabitants of

Kerala is 72 years while for all India it is only 54 years (Goraczko, 2000). It has been shown by Nair et al. (1999), following a detailed study, that the residents of HLNRA s of Kerala showed no evidence of enhanced rates of cancer. Further, there was no correlation with the natural radiation levels in the densely populated areas of the monazite-rich Kerala sands with the stratification of newborns with malformations, still births or twinning (Jaikrishan et al., 1999).

The observation that there are no noteworthy adverse health effects in some of the worlds very high level natural radiation areas brings into question the 'safety levels of radiation'. Jaworowski (2000) in his presentation on "Ionization radiation and radioactivity in the 20th century" summed up his views as follows.

"Man's contribution to the contents and flows of radionuclides and of radiation energy in the particular compartments of the environment consists of a tiny fraction of the natural contribution. In some areas in the world, the natural radiation doses to man and to other biota are many hundreds of times higher than the currently accepted dose limit for the general population. No adverse health effects were found in humans, animals and plants in these areas. In the future reconstruction of the edifice of radiation protection that now stands on the abstract LNT foundations, down-to-earth approach will be necessary taking into account apparently safe chronic doses in the high natural radiation areas, rather than the statistical variation around an average global value. It seems, therefore, that studies of these areas deserve special attention and support in the coming years".

> Man still has to learn a lot from nature – Geology provides the clues!