Assessing the need for radiation protection measures in artisanal and small scale mining of tantalite in Oke-Ogun, Oyo State, Nigeria

A. E. Ajetunmobi\textsuperscript{a,}\textsuperscript{*}, A. O. Mustapha\textsuperscript{b}, I. C. Okeyode\textsuperscript{b}, A. M. Gbadebo\textsuperscript{c}, D. Al-Azmi\textsuperscript{d}, T. W. David\textsuperscript{a}

\textsuperscript{a} Department of Physics, Olabisi Onabanjo, Ago-Iwoye, Ogun State, P M B 2002, Ago-Iwoye, Ogun State.
\textsuperscript{b} Department of Physics, Federal University of Agriculture, Abeokuta, P M B 2240, Abeokuta, Nigeria
\textsuperscript{c} Department of Environmental Management and Toxicology, Federal University of Agriculture, Abeokuta, P M B 2240, Abeokuta, Nigeria
\textsuperscript{d} Department of Applied Sciences, College of Technological Studies, Public Authority for Applied Education and Training, Shuwaikh, P O Box 42325, Code 70654, Kuwait

Abstract

There is concern that work scenarios on the tantalite mining sites in Oke-Ogun, Oyo State, Nigeria may cause occupational radiation exposure of workers due to enhanced concentrations of naturally occurring radionuclides in some process materials. A prior radiological assessment of the mining activities was carried out to determine if and which exposure scenarios may require radiation protection measures. Samples of the materials involved, comprising tantalite (tantalum ores), soil and waste rock were collected and analyzed for activity concentrations of \(^{40}\text{K}\), \(^{226}\text{Ra}\), \(^{238}\text{U}\) and \(^{232}\text{Th}\) using a hyper pure germanium detector gamma-ray spectrometer. Radon concentrations in the mines were also measured using a continuous radon monitor – Radon-Scout Plus (Sarad, GmbH). Activity concentrations of \(^{40}\text{K}\) are below 1 Bq kg\(^{-1}\) in all the samples but all the tantalite samples contain more than 1 Bq kg\(^{-1}\) of \(^{226}\text{Ra}\) and \(^{238}\text{U}\). Hence tantalite is regarded as naturally occurring radioactive material (NORM) and the mining activity as a practice. The requirements for planned exposure situations apply to all the mining sites but, on the basis of graded approach, the optimum radiation protection measures vary from one mine to another, ranging from exemption to authorization. Exposures to radon in the underground mines pose the greatest radiological risks and portend the greatest need for regulatory control in the mining operations. The results further underscore the need to integrate radiation protection with the other health and safety measures in the mining sector.

DOI:10.46481/jnsps.2022.699

Keywords: Radiation protection, tantalite, germanium detector

Article History:
Received: 7 March 2022
Received in revised form: 27 April 2022
Accepted for publication: 29 April 2022
Published: 16 August 2022

© 2022 The Author(s). Published by the Nigerian Society of Physical Sciences under the terms of the Creative Commons Attribution 4.0 International license (https://creativecommons.org/licenses/by/4.0). Further distribution of this work must maintain attribution to the author(s) and the published article’s title, journal citation, and DOI. Communicated by: W. A. Yahya

1. Introduction

Minerals and raw materials normally contain moderate to elevated concentrations of naturally occurring radionuclides, mainly \(^{40}\text{K}\) and those in the decay series of \(^{238}\text{U}\) and \(^{232}\text{Th}\) [1-6]. Radiation safety regulations are well established in uranium
mining and processing where uranium ores contain up to several thousand $\text{Bq}^{-1}$ of radionuclides in the $^{238}\text{U}$ and $^{232}\text{Th}$ decay series [2]. There are many other industrial activities where uranium is not the main product but a contaminant or a secondary mineral [2]. The radiological hazards in such industrial activities may be significantly reduced and the need for regulatory control may not be clear. The International Atomic Energy Agency (IAEA) has provided some Guidance but the decision on which of the industrial activities to regulate is normally left for the regulatory body [2, 4, 5]. This includes establishing activity concentrations of the relevant radionuclides in the materials involved, determination of doses and appropriately designating exposure situations based on the graded approach and other requirements of the Basic Safety Standards [4].

The effective dose received by a worker exposed to naturally occurring radioactive material is reliably assessed by conducting a properly developed monitoring programme in the relevant workplaces [5, 6]. It can also be derived from model calculations using appropriate operational quantities and dose coefficients [3]. However, if the work scenarios and the activity concentration of radionuclides in relevant process materials are well known, it is possible to obtain, in advance, a broad indication of the expected dose to workers from exposure to gamma radiation and exposure due to airborne dust. The latter approach is described in the IAEA Safety Series Report No. 49 [2] as a prioritization tool for identifying the types of industrial operations and scenarios having the greatest need for radiation protection measures. This approach was adopted in the present study.

Artisanal and small-scale Mining (ASM) have been practiced over several decades in many parts of Nigeria, including Oke-Ogun, south western Nigeria. It is an informal mining sector where rudimentary techniques or machinery with small degree of mechanization are used to exploit small and shallow mineral resources. Although ASM provides livelihood alternative for some people, it contributes very little to Nigeria’s economy and it has been associated with various environmental degradation and health hazards. Formalization and reform of the ASM are now being carried to improve the mining sector as part of the national policy to diversify from the oil and gas sector in Nigeria [7]. The formalization is expected to institutionalize best practices including those that address the associated environmental and health hazards, both radiological and non-radio logical. In this regard, it is anticipated that the outcome of is study will provide some insights into the potential radiological exposures associated with ASM operations in general and tantalite mining operations in particular. The report will also serve as practical guide on how to determine the need for radiation protection measures in industrial activities that involve naturally occurring radioactive materials (NORM) based on the graded approach.

2. Materials and methods

2.1. Geographic and geological features of the study area

The mines are located in Komu, Sepeteri, Gbedu and Eluku villages in Itesiwaju, Saki East, Iwajowa and Saki Local Gov-ernment areas respectively, all in Oke-Ogun, Oyo State, southwest of Nigeria. Oke-Ogun spans over latitudes $8^\circ 00’ – 8^\circ 39’$ N and Longitudes $2^\circ 56’ – 3^\circ 46’$ E, and it is averagely at an elevation of 188m above the sea level. It has a population of about 1.4 million people according to the latest population census. The main occupations of residents in the area are artisanal mining and farming. The area is well drained by Rivers Oyan, Ofiki, Olori and many other tributaries of Ogun River, forming dendritic drainage patterns in many parts of the area (figure 1).

The major geological formations in the area include undifferentiated schist and gneiss that are widely distributed around Sepeteri in north eastern and Gbedu in south western regions. Alternate layers of granitic gneiss, biotite hornblende granite, migmatite, migmatic granite gneiss, amphibole schist and porphyroblastic gneiss run parallel from northwest to southeast, with discontinuities occasioned by minor geological formations including porphyritic granite in the southwest, quartz veins in the east, pegmatite occurrence around Sepeteri, Komu, Gbedu and Iwere-ile in the south and southwest, and syenite and pyroxene diorite in the north. Parts of Komu in the west and Eluku in the northwest are also both underlain by granitic gneiss and biotite hornblende granite, respectively. Oke-Ogun is endowed with a variety of minerals and precious stones, including tantalite and iron ore, marble, talc, beryl, etc. (figure 2).

2.2. Determination of activity concentrations of radionuclides in process materials

2.2.1. Types of operations and processes involved

The study covered eight mining sites, comprising one each in Gbedu and Sepeteri, two in Eluku and four in Komu. The four mines in Komu are underground while the rest are open pit mines. The entire mining operations were divided into two broad types, namely artisanal mining operations which involve digging manually and blasting to extract ore-bearing rocks, and processing of tantalum ore (tantalite) involving work scenarios such as crushing (dry or wet) and grinding to separate the ore-bearing rock from waste rock, physical separation of tantalite from other metallic ores using gravity, electrostatic or electromagnetic processes, and working or staying close to stock-piles of tantalite. These two types of operations are among those that have earlier been identified as being likely to require regulatory control [2].

2.2.2. Types of materials involved and sample preparation

The three involved materials considered are residual soil, waste rock and tantalite or tantalum ore. Forty samples, comprising twelve ores, seventeen soils and eleven waste rocks were collected from all the selected mining sites. The samples were air dried, pulverized and homogenized. Aliquots were sealed in 500 ml cylindrical plastic containers for a month to ensure secular equilibrium between $^{226}\text{Ra}$ and its decay products.

2.2.3. Gamma-ray spectrometry

The samples were analyzed using a hyper pure germanium (HPGe) detector gamma-ray spectrometer at the National Institute of Radiation Protection and Research, University of
Ibadan, Nigeria. A multi-radionuclides standard source was used for energy and efficiency calibrations. The radionuclides of interest are $^{40}$K, $^{226}$Ra, $^{232}$Th and $^{238}$U, and the gamma-ray energies used to estimate the activity concentrations are 609.3 keV of $^{214}$Bi for $^{226}$Ra; 911 keV of $^{228}$Ac for $^{232}$Th; 1001 keV of $^{234m}$Pa for $^{238}$U; and 1460 keV for $^{40}$K.
2.3. Measurement of radon concentration

The health effect as a result of exposure to $^{222}\text{Ra}$ is radiation induced [8], hence the need to measure radon concentration in the study. Radon concentrations were measured in the open pit and underground mines using a continuous radon monitoring device: Radon Scout Plus (Sarad, GmbH). It is portable (of dimension $175 \times 135 \times 55 \text{ mm}^3$), battery operated and capable of continuous cyclic data logging of radon concentration, air temperature, relative humidity and atmospheric pressure after every predetermined period. During the measurement, the instrument was held above the floor and away from the walls in underground mines. The sampling time varied depending mainly on the time permitted by the mine operators. The logged data were assessed by connecting the radon monitor to a personal computer through a USB connector for graphical display and further data analysis through the dedicated software - Sarad Radon vision 4.0.7 [9].
2.4. Exposure scenarios and pathways

Many work scenarios are involved in the two operations as mentioned in section 2.2.1. Each of these work scenarios may constitute an exposure scenario, but in this screening radiological assessment the exposure pathways considered for workers effective dose are:

i. External exposure due to whole body irradiation by gamma-rays emitted by materials associated with the work,

ii. Internal exposure due to intake (ingestion and inhalation) of radionuclides in dust of the materials,

iii. Inhalation of radon released into air from the surrounding materials

2.5. Assessment of effective doses to artisans

The annual effective dose (E) from exposure to $^{222}$Rn in the mine was estimated from the measured $^{222}$Rn concentrations ($C_{Rn}$) using the conventional equation [1]:

$$ E = C_{Rn} \times F \times T \times D, $$

where $F$ is an equilibrium factor between $^{222}$Rn and its decay products (0.4 for indoor/underground and 0.6 for outdoor), $T$ is hours in a year (hy$^{-1}$) spent by miners at the site and $D$ is the dose conversion factor (9.0×10$^{-6}$ mSv per Bq h m$^{-3}$), which is the effective dose received by an adult per unit $^{222}$Rn exposure.

### Table 1: Relationship between dose and activity concentration for occupational exposure to gamma radiation and to dust [2]

| Category of material | Broad estimate of annual effective dose per unit activity concentration (mSv/y per Bq/g) | Individual radionuclide activity concentration above which the expected dose may exceed 10% of the dose limit (Bq/g) |
|----------------------|---------------------------------------------------------------------------------|----------------------------------------------------------------------------------|
| Minimum               | Maximum                          |                                                                                  |
| Large quantity, e.g. ore body, large stockpile | 0.02                             | 0.4                                 | 5                                        |
| Small quantity, e.g. mineral concentrate, scale, sludge | 0.0008                           | 0.04                               | 50                                       |
| Volatilized: furnace fume and precipitator dust | 0.0006                           | 0.003                              | 500$^a$                                 |

$^a$This value refers to the activity concentration in the precipitator dust with exposure to fume having been accounted for by assuming an equivalent dust loading of 1 mg/m$^3$ at the same activity concentration (i.e. a concentration of 0.05 Bq/m$^3$ in fume) and an activity media aerodynamic diameter (AMAD) of 1 µm.

3. Results and discussion

3.1. Activity concentration of radionuclides in process materials

The ranges of activity concentrations of $^{40}$K, $^{226}$Ra, $^{232}$Th and $^{238}$U in the Tantalite, Soil and rock samples are (0.003-1.183) Bq/g, (1.231-140.982) Bq/g, (0.753-4.808) Bq/g and (2.314-173.827) Bq/g; (0.003-1.139) Bq/g, (0.008-0.369) Bq/g, (0.003-0.253) Bq/g and (0.003-0.761) Bq/g; and (0.071-1.709) Bq/g, (0.003-0.261) Bq/g, (0.003-0.318) Bq/g and (0.003-0.297) Bq/g$^1$, respectively (table 2). All the materials contain less than 10 Bq$^{-1}$ of $^{40}$K and all soil and rock samples contain less than 1 Bq$^{-1}$ of $^{226}$Ra, $^{232}$Th and $^{238}$U. Concentrations of $^{226}$Ra and $^{238}$U exceed 1 Bq/g in all the talanite samples, while those of $^{232}$Th exceed 1 Bq$^{-1}$ in about 75% of the talanite samples.

The implication of these results is that tantalite qualifies to be regarded as NORM, its mining in all the mine sites is regarded as a practice and the requirements for planned exposure situations apply as stated in the IAEA GSR-3 [4]. On the other hand, soil and rock in the mine sites do not require further radiological considerations unless there are possibilities of being used as building materials.

The results also show disequilibrium between $^{238}$U and $^{226}$Ra in all the talanite samples, but only in a few soil and rock samples. This was also reported by [3] in columbite-tantalite from the Democratic republic of Congo and by [10] in niobium ore from Brazil. The disequilibrium between $^{238}$U and $^{226}$Ra in the tantalum ores may indicate mobilization into other materials not considered in this study, such as waste water, and this may have radiological environmental impact around the mines as discussed by [3].
Table 2: Activity concentration of radionuclides in samples of materials involved in the mining operations and 222Rn concentration in the mine air

| Location and material | Range of activity concentrations (Bq/g) | Average 222Rn concentration (Bq m⁻³) |
|-----------------------|----------------------------------------|-------------------------------------|
|                       | ⁴⁰K  | ²²⁶Ra  | ²³²Th  | ²³⁸U  |                                |
| Eluku I               |      |        |        |       | 92                                 |
| Tantalite             | 0.113| 20.821  | 1.473  | 23.979|                                |
| Rock                  | 0.262| 0.047   | 0.005  | BDL   |                                |
| Soil                  | 0.071| 0.029   | 0.009  | BDL   |                                |
| Eluku II              |      |        |        |       | 55.5                               |
| Tantalite             | 0.13 | 44.674  | 1.489  | 51.01 |                                |
| Rock                  | 0.296| 0.062   | 0.007  | BDL   |                                |
| Soil                  | 0.13 | 0.032   | 0.024  | BDL   |                                |
| Gbedu                 |      |        |        |       | 56                                  |
| Tantalite             | 0.162| 24.277  | 2.786  | 27.867|                                |
| Rock                  | 1.111| 0.01    | BDL    | BDL   |                                |
| Soil                  | 0.676| 0.188   | 0.107  | 0.249 |                                |
| Komu I                |      |        |        |       | 180335.3                            |
| Tantalite             | BDL  | 127.048 | 1.583  | 155.724|                                |
| Rock                  | BDL  | 0.17    | 0.008  | 0.23  |                                |
| Soil                  | 1.709| 0.035   | 0.003  | BDL   |                                |
| Komu II               |      |        |        |       | 228.4                               |
| Tantalite             | 0.171| 95.663  | 2.917  | 118.903|                                |
| Rock                  | BDL  | 0.178   | 0.008  | 0.259 |                                |
| Soil                  | 1.638| 0.035   | 0.003  | BDL   |                                |
| Komu III              |      |        |        |       | 4387.6                              |
| Tantalite             | 0.006| 95.327  | 1.509  | 123.56|                                |
| Rock                  | 0.621| 369     | 0.253  | 0.761 |                                |
| Soil                  | 1.652| 0.035   | 0.003  | BDL   |                                |
| Komu IV               |      |        |        |       | 2565.8                              |
| Tantalite             | BDL  | 140.982 | 2.056  | 173.827|                                |
| Rock                  | 0.213| 0.239   | 0.09   | 0.417 |                                |
| Soil                  | 1.666| BDL     | BDL    | BDL   |                                |
| Sepeteri              |      |        |        |       | 55                                  |
| Tantalite             | 0.743| 18.254  | 1.966  | 55.783|                                |
| Rock                  | 0.812| 0.023   | 0.007  | 0.021 |                                |
| Soil                  | 0.656| 0.073   | 0.016  | 0.152 |                                |

BDL = Below Detection Limit; Detection limits are 3.1, 2.8, 2.9 and 2.9 (10⁻³ Bq/g) for ⁴⁰K, ²²⁶Ra, ²³²Th and ²³⁸U activity concentrations, respectively.

3.2. Radon concentration in the mines

The mean ²²²Rn concentrations vary widely from 55 to 180335 Bq m⁻³ (table 2). The values recorded in the open pit mines are higher than the global average outdoor value (10 Bq m⁻³) but within the global range (1-100 Bq m⁻³) [1]. The values recorded in the underground mines are all significantly higher than the action level for workplaces, which is 1000 Bq m⁻³ annual average [4, 5]. The operations in underground mines are further subject to the requirements for planned exposure situations on account of significantly high ²²²Rn concentrations. This decision is based on the assumption that the average radon concentration during the short-term monitoring is representative of the annual average, which could be more or less, subject to atmospheric, diurnal and seasonal variations. Unfortunately, long term or repeated measurements were not allowed by the mine owners. Presently, they seem unaware of occupation exposure to radiation or the needs for radiation protection in their industry.

3.3. Determination of optimum regulatory control options using the graded approach

A graded approach to regulation, as embodied in the IAEA Basic Safety Standards [4], states that the application of the requirements in planned exposure situations “shall be commensurate with characteristics of the practice or the source within a practice, and with the magnitude and likelihood of the exposures”. In the case of exposure to NORM, it will involve more than just establishing that the activity values of the relevant radionuclides are exceeded, but the particular types of operation, process and material will also be considered in more detail, including screening dose assessments. The outcome will determine the optimum regulatory option.
Table 3: Expected effective dose to workers, operation/scenario with the greatest need for radiation protection and recommended measure based on the graded approach

| Mine location | Mine type | Workers dose (mSv/y) | Operation/scenario with greatest need for radiation protection | Recommended radiation protection measure |
|---------------|-----------|---------------------|---------------------------------------------------------------|------------------------------------------|
|               |           | Exposure to $^{222}$Rn | to Exposure to gamma radiation and dust                       |                                          |
| Eluku I       | Open pit  | 0.73                | 2.040                                                         | Tantalite processing                     | Exemption                                |
| Eluku II      | Open pit  | 0.44                | 0.959                                                         | Tantalite processing                     | Exemption                                |
| Gbedu         | Open pit  | 0.22                | 1.115                                                         | Tantalite processing                     | Exemption                                |
| Komu I        | Underground | 5690.0             | 2.231                                                         | Underground mining                       | Authorization                            |
| Komu II       | Underground | 7.20               | 6.229                                                         | Underground mining                       | Notification                             |
| Komu III      | Underground | 80.71              | 4.756                                                         | Underground mining                       | Authorization                            |
| Komu IV       | Underground | 94.40              | 4.942                                                         | Underground mining                       | Authorization                            |
| Sepeteri      | Open pit  | 0.11                | 6.953                                                         | Tantalite processing                     | Notification                             |

The two types of operations considered in the study are mining of tantalite and processing of tantalite, and tantalite (tantalum ore) is the NORM involved in both operations, i.e. material in which concentrations of $^{226}$Ra and $^{238}$U exceed 1 Bq/g (table 2). Tantalite processing involves exposure scenarios, e.g. magnetic separation of tantalite from other metallic ores, where workers come in contact with small quantities of materials. Therefore, the expected effective doses due to workers exposed to gamma radiation and to airborne dust of tantalite were estimated using the published annual effective dose per unit activity concentration for small quantities of materials (table 1). The estimated values range from about 1 to about 7 mSv/y as shown in table 3. According to the graded approach to regulation, the operation could be exempted if the effective dose received by a worker from this pathway does not exceed 1 – 2 mSv/y. Therefore, the operations in the mines in Eluku I, Eluku II and Gbedu could be considered for exemption, meaning there is no need to impose any regulatory requirements on them.

After exemption the next level in the graded approach is notification, which is the requirement for the legal person (which in this case is the mine owner) to submit a formal notification to the regulatory body informing it of the operation. Notification alone could be sufficient provided the exposures do not exceed a small fraction of the relevant limits. Consequently, the open pit mine in Sepeteri and one of the underground mines in Komu (Komu II) could be considered for Notification.

Authorization is the next to notification when there is need to place higher obligations on the legal person. According to the graded approach, authorization may take the form of registration or licensing, depending on how stringent the regulation is required to be. Registration is the less stringent of the two and it places only limited obligations on the legal person, but “it may provide sufficient level of control in many operations involving significant, but nevertheless moderate exposures to NORM and/or radon”. Licensing is the highest level in the graded approach to regulation. It is the more appropriate authorization where optimized protection can only be achieved through the enforcement of specific exposure control measures. In this regard, the high exposures to radon in the remaining three underground mines in Komu may require authorization. While registration may provide sufficient level of control for the operations in Komu III and Komu IV, licensing may be more appropriate for the operation in Komu I where workers may experience significant exposures to radon. Specific control measures such as installation of ventilation system and other remedial actions to drastically reduce exposure to radon would be required, considering that any situation that may result in an annual dose greater than 100 mSv is unacceptable, except under emergency exposure situation [4]. There may also be need to establish suitable radiation protection programmes that entail monitoring and dose assessments.

4. Conclusion

An assessment was carried out to evaluate the need for radiation protection measures in artisanal and small scale mining of tantalite in Oke-Ogun, southwest, Nigeria. The results generally indicate there is need for radiation protection measures in the mining operations. Two types of operations were considered, namely tantalite mining in both open pit and underground mines and tantalite processing. Three of the materials involved in the operations, namely soil, rock and tantalite, were
considered but only tantalite (tantalum ore) contains more than 1 Bq/g of $^{226}$Ra and $^{238}$U and qualifies as NORM. High concentrations of $^{222}$Rn were measured in mine air, particularly in the underground mines. Doses to workers due to exposures to gamma radiation and due to dust, and exposure to $^{222}$Rn show that the mining operations are services and the requirements for planned exposure situations apply. The graded approach to regulation was applied to optimize the regulatory options for each of the mines. The optimized regulatory options range from exemption to licensing of the operations. But the greatest need for radiation protection measures in the tantalite mining operations is workers exposure to radon in underground mines. It is recommended that the regulatory body should liaise with other relevant stakeholders and seize the opportunity of the ongoing formalization and reform of the ASM sector in Nigeria to integrate radiation protection with the general OHS measures in the mining industry.

References

[1] UNSCEAR, “Dose assessment methodologies”, New York: United Nations Scientific Committee on the Effect of Atomic Radiation (2000). Report to the General Assembly UNSCEAR

[2] IAEA, “Assessing the need for Radiation Protection measures in work involving minerals and raw materials”, Safety Report Series No. 49 (2006), (Vienna: IAEA).

[3] A.O. Mustapha, P. Mbutukongira, & M. Mangala, “Occupational Radiation Exposure of Artisanal Mining of Columbite-tantalite in the Eastern Democratic Republic of Congo”, Journal Radiological Protection 27 (2007) 187.

[4] IAEA, “Radiation Protection and Safety of Radiation Sources”. (International Atomic Energy Agency) International Basic Safety Standards, General Safety Requirements Part 3, No. GSR (2004) (Vienna).

[5] IAEA, “Occupational Radiation Protection”, General Safety Guide No. GSG-7 (2018) (Vienna).

[6] IAEA, “Occupational Radiation Protection in the Uranium Mining and Processing Industry”, Safety Report Series No. 100(2020) (Vienna: IAEA)

[7] The World Bank, Nigeria, “Mineral Sector Support for Economic Diversification Project (MinDiver)”, Implementation Status and Results Report (2019).

[8] O. A. Ndubisi, B. K. M. Apaemi, S. F. Barikpe & I. T. Ari, “Analysis of Indoor Radon Level and its Health Risks Parameters in three selected towns in Port Harcourt, Rivers State, Nigeria”, J. Nig. Soc. Phys. Sci. 3 (2021) 181.

[9] Sarad, “User manual, Radon Scout, version 10/2009, SARAD GmbH”, Wiesbadener Strabe 10. (2009) D-01159 Dresden.

[10] D. O. Pires, M. A. Rio, E. C. S. Amaral, H. M. Fernandes & E. R. R. Rochedo, “Environmental radiological impact associated with non-uranium mining industries: A Proposal for screening criteria”, J. Environ. Radioact. 59 (2002) 1.