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RADIOACTIVITY IN CONSTRUCTION MATERIALS

A LITERATURE REVIEW AND BIBLIOGRAPHY

OFFICE OF RADIATION PROGRAMS

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Gregory G. Eadie

April 1975

U. S. Environmental Protection Agency
Office of Radiation Programs
Las Vegas Facility
P.O. Box 15027
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Surveys to determine the radioactive content of specific building materials used in the United States have not been reported in the literature. The external dose to the U.S. population from exposure to natural radioactive materials (exclusive of uranium mill tailings) contained in U.S. building materials has not been evaluated, and the possibly significant external exposure from the use of by-product gypsum and fly-ash materials should be evaluated. The effects of various construction materials on the attenuation of cosmic and terrestrial radiations have been evaluated in a limited number of surveys in the urban areas of Boston, New York, and Livermore, California. The measurement of radon and radon daughter product concentrations has only been reported for a few dwellings and several multi-story office buildings in Boston and in several State-owned buildings in North Carolina. This literature search has found a lack of meaningful data for use in evaluating the U.S. population exposure from building materials.

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Gregory G. Eadie

U.S. Environmental Protection Agency
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Las Vegas, Nevada 89114

PREFACE

The Office of Radiation Programs of the Environmental Protection Agency carries out a national program designed to evaluate population exposure to ionizing and non-ionizing radiation, and to promote development of controls necessary to protect the public health and safety. This literature survey was undertaken to assess the extent of the exposure of the population to naturally occurring radionuclides present in building materials and to provide a basis for further studies. Readers of this report are encouraged to inform the Office of Radiation Programs of any omissions or errors. Comments or requests for further information are also invited.

Donald W. Hendricks
Director, Office of
Radiation Programs, EPA

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INTRODUCTION

A literature search has been conducted to evaluate the exposure of the population to levels of naturally occurring radionuclides present in construction materials.

A bibliography of pertinent references has been compiled from the bibliographies by Lowder (1965) and Klement (1965 and 1970), and has been supplemented by searching the Nuclear Science Abstracts (NSA) through Volume 31, Number 4 (February 1975).

This bibliography on radioactivity in construction materials contains, to a large extent, articles from the early 1950's to the present, since few surveys were reported in the literature prior to 1950. A brief description of important topics dealt with in each article has been provided with the reference source for those articles which have been reviewed.

SUMMARY

Surveys to determine the radioactive content of specific building materials used in the United States have not been reported in the literature. The external dose to the United States population from exposure to natural radioactive materials (exclusive of uranium mill tailings) contained in United States building materials has not been evaluated, and the possibly significant external exposure from the use of by-product gypsum and fly-ash materials should be evaluated. The effects of various construction materials on the attenuation of cosmic and terrestrial radiation have been evaluated in a limited number of surveys in the urban areas of Boston, New York, and Livermore, California. The measurement of radon and radon daughter product concentrations has only been reported for a few dwellings and several multi-story office buildings in Boston and in several State-owned buildings in North Carolina. This literature search has found a lack of meaningful data for use in evaluating the U.S. population exposure from building materials.

CONCLUSIONS

1. The article by Hamilton (1971) is the only significant report of data on the radioactivity content of specific building materials.

2. Radioactivity in building materials used in the United States has received very little attention. Except for the studies to find construction materials of very low background, there are no reports of radiological surveys of any United States building materials which are used by the general population for construction purposes. Also, there are no reports of United States studies on the possible use of by-product gypsum and fly-ash products for construction materials.

3. The reports by Solon, et al., (1960); Yeates, et al., (1970 and 1972); and Lindeken, et al., (1971 and 1973) provide the only data on radiation measurements made inside United States buildings.

4. The reports by Yeates, et al., (1970 and 1972) and Aldrich and Conners (1974) are the only reported data of radon daughter product concentration measurements made inside United States buildings (exclusive of measurements made to study uranium mill tailings material usage).

5. The documentation of the evaluation of radiological hazards associated with the use of uranium mill tailings material for construction purposes in the United States has not been reported in the open literature [except for the report by Duncan and Eadie (1974)].

6. Dose assessments of radiation exposures inside dwellings, and evaluation of the internal dose due to the inhalation of radon daughter products in the air inside dwellings have been reported in a number of articles from foreign countries. These reports indicate that radon and radon daughter product concentrations in air inside dwellings of concrete or granite construction probably exceed a working level value of 0.01, which is the lower limit of the exposure

guideline currently recommended for the remedial action program in Grand Junction, Colorado (JCAE, 1971), where uranium mill tailings material has actually been used for construction purposes.

7. Table 1, taken from Oakley (1972), represents the best summary of the gonadal dose equivalent to the United States population from natural sources and concludes that the total dose equivalent is 88 mrem/yr.

RADIOACTIVITY IN CONSTRUCTION MATERIALS

There are only a few published reports which specifically discuss the concentration of radionuclides in building materials such as brick, gypsum board, concrete, etc. The article by Hamilton (1971) reports the concentrations of potassium, thorium, uranium, and radium for various building materials used in the United Kingdom. The results are expressed in "ppm" and, for comparison purposes, the concentration results were converted to a "radium equivalent" unit. This expresses the summation concentrations of potassium, thorium, and uranium in terms of the concentration of radium which gives the same gamma-ray emission per gram of material. Gypsum board, derived as a by-product of the manufacture of superphosphate fertilizers, had the highest radium equivalent of all building materials, 17.0 picocuries per gram (pCi/g). Natural gypsum products in the United Kingdom had a radium equivalent of 1.2 pCi/g. Clay bricks, the most commonly used building material, had a concentration of 1.4 pCi/g. There is some indication that concrete blocks produced from fly ash (i.e., a mixture of coal clinker, ash, and cement) have a radium equivalent greater than other brick products. As a result of Hamilton's report, the United Kingdom has considered legislation to limit the average concentration of radium in by-product gypsum materials for use in the construction industry to less than 25 pCi/g (O'Riordan, et al., 1972).

In a report to the United Kingdom's National Radiological Protection Board, O'Riordan, et al., (1972), evaluated the dose rate from the use of by-product gypsum material. Considering that the typical radium content of by-product gypsum material is 25 pCi/g, the corresponding gamma-ray dose rate to the gonads and bone marrow of the occupants of a standardized house was calculated to be 30 mrad per year. The beta-ray dose rate to the skin and lens of the eye was less than 20 mrad per year. An evaluation of the lung dose due to radon and radon daughter products produced from the use of the by-product gypsum indicated an annual exposure of 0.04 working level months (WLM). The report summarized that the radiation exposures due to the use of by-product gypsum products are about one-tenth of the annual dose limits for exposure to members of the general public as recommended by the International Commission on Radiological Protection.

Standards for natural radioactive substances in building materials have been established in the Soviet Union and are contained in the Soviet Sanitary Regulations, Numbers 437 through 463. In summary, the maximum permissible concentration of naturally radioactive substances in Soviet building materials is less than 20 pCi of the total radium-228, radium-226, thorium-228, thorium-230, uranium-234, lead-210, and polonium-210 content per gram of material.

The article by Krisyuk, et al., (1974) also discusses gamma-spectrometric analyses of various building materials used in the USSR and predicts exposure rates inside buildings. Suggested permissible concentrations of radium-226, thorium-232, and potassium-40 are 10, 7, and 126 pCi/g, respectively, based on external gamma radiation levels inside structures.

In the article by Afanas'ev and Krisyuk (1967), the exposure of occupants due to increased levels of radon and thorium and their decay products in indoor air is discussed. The article concludes that measurements of specific radon and thorium liberation rates should be evaluated on all new building materials, e.g., by-product gypsum boards. Also, different air-change factors and the influence of paint or sealants on the emission rates inside structures should be considered.

Data on the potassium, uranium, and thorium content of building materials of Taiwan are given in the article by Chang, et al., (1974). The results are expressed in parts per million per kilogram and, from these values, it was estimated that the external gamma radiation from a typical concrete building was 52 milliroentgen per year (mR/y).

Other articles dealing specifically with radioactivity in building materials are listed below:

Author	Country
Aten, et al., (1961)	Netherlands
Bergstram and Wahlberg (1967)	Sweden
Kominek (1972)	Czechoslovakia
Krisyuk, et al., (1974)	Soviet Union
Lindell and Peizenstein (1964)	Sweden
Roehnsch (1974)	Germany
Safonov (1972)	Soviet Union
Stretta and del Arenal (1963)	Mexico

Several other articles deal with the use of radioisotopes as tracers in building materials or their use in radiography of construction projects:

Author	Country
Hilaire and Le Gallic (1971)	France
Ramos Rodriquez (1970)	Spain
Toyer (1972)	France

With respect to radioactivity in building materials used in the United States, there are three articles by Wollenberg and Smith (1960, 1962, and 1966) which report the determination of the natural radioactivity content of materials used in concrete or concrete aggregate mixes. These studies were undertaken to determine the lowest background materials for use in constructing a room for a whole-body counting chamber and are, therefore, not representative of typical building materials. Schiager (1974) also discusses the selection of low-background building materials (concrete and aggregate mixes) which were used to construct a photographic film storage facility having an exposure rate less than 11 microroentgens per hour (μR/h).

Other articles which report the natural radioactivity content of various rocks and soils of the United States are listed in the bibliography. The articles by Harley and Lowder (1971) and Oakley (1972) provide good summary reports of the natural radionuclide content and dose rates from common rocks and soils, but these conditions are representative only of outdoor terrestrial exposures.

Although extensive surveys have been conducted in the United States to determine the radioactivty of water supplies and foodstuffs, very little information on the radioactivity analysis and the radiation exposure conditions of specific United States building materials, such as gypsum products, concrete, brick, etc., has been reported in the literature. No reports were found which correlate the radioactivity content of the building materials to the radiological exposure conditions measured inside the buildings.

RADIATION SURVEYS

The following references are concerned with surveys of the outdoor measurement of cosmic and terrestrial background radiation exposure for different sections of the United States:

Author	Area of Survey
Beck, et al., (1964)	Southeast, Central, & Western U.S.
Golden, Jr. (1968)	Florida
Harley and Lowder (1971)	General
Keefer and Dauer (1970)	Florida
Kinsman (1958)	General
Levin and Stoms (1969)	Michigan, Colorado, & Minnesota
Lindeken, et al., (1971, 1972, & 1973)	California
Lowder, et al., (1964)	New England states
McLaughlin (1972)	General
Moxham (1963)	Maryland
Oakley (1972)	General
Patterson, et al., (1958)	San Francisco Bay area
Pinkerton, et al., (1964)	Maryland
Segall and Reed (1963)	New England states
Solon (1958)	General
Stephens and Patterson (1961)	San Francisco Bay area
Wollenberg and Smith (1960)	San Francisco Bay area

Oakley (1972) summarized the results of studies of cosmic and terrestrial radiation exposure rates in the United States. The average dose equivalent to the United States population, considering the population distribution with respect to elevation, the influence of housing and biological shielding, and the contribution from internal emitters, are presented in table 1, from his report.

Oakley based his calculations of the terrestrial component of the United States background exposure rate on the results of the Aerial Radiological Measurement Surveys (ARMS), which were conducted from 1958 to 1963 by the U.S. Geological Survey and EG&G, Inc., under U.S. AEC sponsorship.

Table 1. GONADAL DOSE EQUIVALENT TO THE U.S. POPULATION FROM NATURAL RADIATION

SOURCE	DOSE EQUIVALENT (mrem/yr)
External	
Terrestrial	26
Housing factor = 0.80 Screening factor = 0.80	
Cosmic	44
Internal	
Potassium-40	16
Other nuclides	2
Total	88

*Taken from D. T. Oakley (1972).

The effects of different building materials on the attenuation of the natural background radiation was taken into account by introducing a "housing factor" which was defined as the average factor by which indoor living reduces man's exposure to natural external radiation. Oakley reviewed the available literature which includes both U.S. and foreign surveys, and concluded that the ratio of the indoor to outdoor dose rates for wood structures ranged from 70 to 82 percent. Similar ratios for homes of masonry construction ranged from 72 to 106 percent. Oakley therefore assumed that the ratio of inside to outside dose rates for wood dwellings was 70 percent and for masonry buildings it was 100 percent. Other conditions were also considered in the selection of parameters for the calculation of the housing factor. The type of housing (single-family versus multiple-family dwellings), type of construction (wood versus masonry structure), and occupancy time estimates for at-home versus away-from-home activities were considered in the derivation of the average housing factor of 0.8.

The following articles report the results of surveys conducted to evaluate the radiation exposure rates inside United States buildings:

Author	Area of Survey
Jones, et al., (1971)	California
Lindeken, et al., (1971 & 1973)	Livermore, California
Lowder and Condon (1965)	Vermont, New Hampshire
McLaughlin (1972)	General
Neher (1957)	General
Oakley (1972)	General
Solon, et al., (1960)	New York City
Yeates, et al., (1970 & 1972)	Boston

Neher (1957) reported the results of measurements made inside various buildings using an ionization chamber. The average exposure rate (cosmic radiation contribution subtracted) was 60 mR/yr for wood structures and 130 mR/yr for concrete buildings.

Solon, et al., (1960) also used an ionization chamber to survey dwellings in the metropolitan New York area and reported the results of the survey as cosmic plus terrestrial radiation. Measurements on the first floor of wood-frame dwellings (for three dwellings surveyed) showed a range of 76 to 97 mR/yr. Measurements on the first floor of brick and stone structures (for six dwellings surveyed) ranged from 70 to 84 mR/yr. Radiation measurements were also made in the upper floors of a brick apartment building,

but it is difficult to reach a definite conclusion from the data reported. Radiation levels outside of the surveyed structures were also reported and it appears that, in general, the radiation level inside the structure was somewhat lower than outside.

The article by Lowder and Condon (1965) discusses radiation surveys conducted in Vermont and New Hampshire, but only the results of outdoor measurement are reported. Although the authors present the conclusion that "the mean indoor levels were close to 70 percent of the corresponding outdoor levels in each area," no data were reported for the measurements made inside structures and it is, therefore, difficult to verify this conclusion.

The report by Yeates, et al., (1970) concludes that the ratio of the indoor to outdoor dose rates for wood structures is 82 percent (for five dwellings, first floor measurements), and a range of 87 to 106 percent for steel and concrete office buildings (for four structures surveyed). Yeates, et al., (1972) also discussed data from the 1970 report and has included data of gamma exposure rates inside single- and multiple-family dwellings and for several multi-story office buildings in Boston. The dose rates inside wooden single-family dwellings were from 25 to 50 percent lower than outdoor levels. For masonry multiple-family dwellings, the inside dose rate was about 10 percent lower than the outside measurement. The gamma exposure rates in multi-story office buildings were reported, but the data fail to show any significant change of the radiation level with height inside the building.

Lindeken, et al., (1971 and 1973) used TLD dosimeters to record the background radiation exposure in about 100 residences (mostly wood-frame structures) in the vicinity of Livermore, California. A median annual exposure rate of 63 mR, with a range of 52 to 120 mR/yr, was reported. Radiation levels on the second floor of the dwellings were slightly lower than the ground floor levels. Jones, et al., (1971) also reported TLD gamma surveys inside dwellings.

The article, "Radiation Levels Inside and Outside Buildings," by Goldin is included in the reference of McLaughlin (1972). Goldin summarizes the work in the United States by Yeates and presents a summary table of the radiation measurements inside dwellings in Sweden, East Germany, and Scotland. Some discussion of radon daughter concentration measurements inside buildings is reported by Hultquist (Sweden) and Yeates (United States).

The Second Workshop on the Natural Radiation Environment, W. M. Lowder, ed., (1974), contains several articles which discuss environmental gamma radiation surveys and also radon/radon daughter evaluations made indoors in Grand Junction, Colorado.

In summary, the analysis of the few reported United States surveys can only lead to general conclusions regarding the attenuation of background radiation by United States building materials. Wood structures tend to have lower radiation levels inside compared to the outside levels. Single-story concrete or brick structures may have inside radiation levels higher than the outside levels due to the presence of natural radioactivity in the construction materials. The second floor of any type of structure (i.e., wood or masonry construction) seems to have a slightly lower radiation level than does the first floor. The ground-level floors of multi-story buildings have higher radiation levels inside than outside; but, in the higher stories, the radiation levels inside the building may be less than the outside radiation levels.

The following references report the results of surveys conducted to evaluate the radiation exposure rates inside dwellings in several foreign countries:

Author	Country
Cardinale, et al., (1971)	Italy
Gustafsson (1969)	Finland
Hultquist (1956 & 1965) (in Swedish)	Sweden
Kametani, et al., (1970) (in Japanese)	Japan
Kurokawa (1971) (in Japanese)	Japan
Ohlsen (1969 & 1970) (in German)	East Germany
Pensko, et al., (1969) (in Polish)	Poland
Spiers (1960)	Scotland
Storruste and Rlinstad (1965)	Norway
Yamashita, et al., (1966)	Japan
Yeates and Ring (1973)	Western Australia

The results of most of these reports are summarized in table 2.

Table 2. GAMMA EXPOSURE RATES INSIDE BUILDINGS - mR/yr

LOCATION AND REFERENCE	TYPE OF BUILDING	AVERAGE	RANGE	REMARKS
East Germany (Ohlsen, 1970)	Frame (old)	110	30-240	
	Brick (old)	110	40-200	
	Brick (new)	97	40-150	
	Stone (old)	140	40-260	
	Industrial Const.(new)	92	40-150	
Finland (Gaustafsson, 1969)	Wood	81	78-88	(cosmic of 28 mR/yr incl.)
Italy (Cardinale, et al., 1971)	Unclassified	--	105-263	(cosmic of 37 mR/yr incl.)
Norway (Storruste and Reinstad, 1965)	Wood	87	66-118	(cosmic of 25 mR/yr incl.)
	Concrete	117	81-152	
	Brick	129	91-158	
Scotland (Spiers, 1960)	Stone (Edinburgh)	48	25-70	
	Stone (Dundee)	64	45-80	
	Granite (Aberdeen)	85	70-110	
Sweden (Hultquist, 1956)	Wood	80	70-150	
	Brick	137	70-220	
	Concrete	206	130-520	
W. Australia (Yeates and King, 1973)	Wood	52	--	
	Brick	65	--	
	Concrete	67	53-91	(office bldg.)

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*Parts of this table were taken from the article by A. S. Goldin in the report by J. E. McLaughlin (1972).

RADON AND RADON DAUGHTER PRODUCT CONCENTRATIONS INSIDE BUILDINGS

The only reported radon and radon daughter product concentrations measured inside United States buildings are contained in the articles by Yeates, et al., (1970 and 1972), and Aldrich and Conners (1974). Yeates measured the radon daughter product concentrations inside dwellings and inside multi-story buildings and compared the results to the outside measurements. The radon daughter product concentrations measured inside the dwellings (first and second floors) were comparable to the outdoor concentrations, but the concentrations in the basements were about a factor of five higher than outside. In the office buildings, the radon daughter product concentrations were low due to the effectiveness of the ventilation system for changing and filtering the air.

In the article by Aldrich and Conners (1974), data are reported on evaluations of the indoor radon daughter product concentrations for 32 State-owned office buildings in North Carolina. Only 5 out of 32 structures surveyed had working level values in excess of 0.01 WL; the highest value being 0.028 WL for the Old Senate Chamber of the State Capitol Building (granite, marble construction with no ventilation system).

Other authors [Barton, et al., (1973); Bunce (1966); Gesell (1973); Harley (1973); and Johnson, et al., (1973)] have considered the exposure of inhabitants of dwellings as a result of unvented combustion products from the use of natural gas in unvented appliances. The average concentrations of radon in natural gas were estimated to be about 20 pCi/l, whereas maximum concentrations of about 1,000 pCi/l have been measured. Other articles dealing with radon in natural gas are to be published in the Proceedings of the Noble Gases Symposium which was held in September 1973 in Las Vegas, Nevada.

Oakley (1972) also considers the internal exposure of organs, other than the lung, of an individual due to the inhalation of radon daughter products in the air inside a dwelling. Based on the UNSCEAR Report (1966), Oakley concluded that the internal exposure from the inhalation of radon daughter products would be less than 2 mrem/yr, gonadal dose equivalent.

Auxier, et al., (1974) describe a study on the effects of using plaster, asphalt, or paint on concrete surfaces to reduce the radon emission rates. Plaster and asphalt coatings did not reduce the radon emission rate. Epoxy paints did reduce the emission rate; however, this resulted in a buildup of radon daughter products at the paint to concrete interface which resulted in an increased gamma exposure rate. This study does not discuss the radioactivity content of building materials.

Kerr, et al., (1974) report on alpha-particle spectrometer measurements of indoor radon daughter products in Grand Junction, Colorado. This study was conducted to evaluate the remedial action program in nine structures where uranium mill tailings material had been used for construction purposes.

Hultquist (1969) reported the results of radon concentration measurements inside Swedish dwellings. For wooden structures (for eight buildings surveyed), a mean radon level of 0.53 pCi/l (ranging from 0.18 to 0.90 pCi/l) was measured. Brick structures (25 surveyed) had a mean radon level of 0.91 pCi/l (ranging from 0.18 to 4.1 pCi/l). Dwellings built mostly of concrete (28 surveyed) had a mean radon level of 1.86 pCi/l (ranging from 0.28 to 5.8 pCi/l).

Sievert (1965) reported results of surveys of the radon and thoron content of air inside Swedish houses of different construction types. Measurements were made before and then after the room was ventilated. The data presented by Sievert are given as the percentage of dwellings (by construction type) which were found in various radon concentration ranges (ranging from less than 0.5 pCi/l to the range of 15 to 30 pCi/l). Therefore, it appears that poor ventilation leads to high concentrations of radon inside dwellings regardless of construction type, and that concrete structures have a higher radon content than do brick dwellings, with wooden structures having the lowest radon content of any type of dwelling surveyed.

Toth (1972) reported on the determination of short-lived radon decay products in the air of unventilated living rooms in Hungary. Analyses for 841 locations completed in 14 different towns indicated mean concentrations for Ra-A, Ra-B, and Ra-C of 3.05, 2.64, and 2.49 pCi/l, respectively. On the basis of these radon daughter product concentrations, and of certain assumptions concerning occupancy time and dose conversion factors, Toth estimated that the Hungarian population receives a dose of 800 mrem/yr to the bronchial epithelium and 120 mrem/yr to the whole lungs.

Hague and Collinson (1967) also reported annual doses for different parts of the respiratory system for various radon and radon daughter concentrations in dwellings surveyed in London [Hague, et al., (1965)].

Four other articles concerned with the measurement of radon concentrations inside dwellings in foreign countries have also been reported:

Author	Country
Mikhaylov, et al., (1967)	Bulgaria
Shem'i-zade (1970)	Soviet Union
Toth (1968)	Hungary
Truelle (1971)	Czechoslovakia

Another report of possible interest, Snihs (1973), is to be published in the Proceedings of the Noble Gases Symposium held in Las Vegas, Nevada, in September 1973.

SURVEY OF BUILDINGS WHERE URANIUM MILL TAILINGS MATERIAL HAS BEEN USED FOR CONSTRUCTION PURPOSES

The extensive radiological surveys conducted by State and Federal agencies (e.g., U.S PHS, U.S. AEC, U.S. EPA, and the State of Colorado) to evaluate the hazards associated with the use of uranium mill tailings material for construction purposes have not been published in the open literature. Detailed reports have been issued to each State where tailings material has been determined to have been used for construction purposes. These State reports include the results of a mobile gamma-scanning survey (used to denote locations with suspected tailings use) and the results of portable instrument radiation surveys completed inside and outside of selected dwellings. For some dwellings, where the use of tailings material could not be completely evaluated from the gamma radiation surveys only, measurements of the indoor radon and radon daughter product concentrations (reported in units of "Working Level") were made and the results reported in the state reports. The extent of the problem resulting from the use of uranium mill tailings material for construction purposes is recorded in the Hearings by the Joint Committee on Atomic Energy (October 1971).

Auxier (1973) also considered the problems associated with the use of uranium mill tailings for construction purposes. Duncan and Eadie (1974) present data on the evaluation of the indoor working level values and also some indoor gamma radiation survey results for structures built immediately adjacent to a uranium mill tailings pile located in Salt Lake City, Utah.

SPECIAL STUDIES IN AREAS OF HIGH RADIATION BACKGROUND

Several articles are included in the bibliography which discuss radiological surveys conducted in areas (in India and Brazil) which have high natural radiation background. Little information concerning the radiological exposure conditions, which result from the use of sands and rocks of high radioactivity content for construction purposes, has been reported in these articles.

Bharatwal and Vaza give the results of gamma radiation surveys conducted inside houses in a monazite sand area, and conclude that, irrespective of the construction material used in the dwelling, the gamma exposure rate inside was almost the same as that outside. No studies have been reported which evaluate the radon levels inside dwellings in these areas of high radium and thorium bearing soils.

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