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ELEVATED INDOOR RADON ASSOCIATED WITH SOILS DEVELOPED ON PLEISTOCENE LIMESTONES IN TROPICAL AND SUBTROPICAL LATITUDES

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ABSTRACT

Indoor radon data and geologic, geochemical, and radiochemical studies suggest that substantial quantities of radium have accumulated in soils formed on karstic limestones and related carbonate-rich sediments of various ages in Florida, Guam, and various islands in the Carribbean. These limestones are typically nearshore marine in origin and are elevated relative to sea level. In Florida, indoor radon levels exceeding 40 pCi/L and, in Guam, indoor radon levels exceeding 200 pCi/L occur in homes built on these soils in spite of climatic, housing, and lifestyle factors that mitigate against high levels. In southeastern Florida, radium concentrations in soils of as much as 11.3 pCi/g have been measured in soils. The source of the radium in the Florida soils is uncertain; however, clayey, Fe-rich soils with high radium concentrations ratios have been documented in many islands of the Carribbean and the Florida Keys. A primary component of these latter soils is Saharan airborne dust. We infer that the Florida soils have a similar origin. In Guam, aerosol dust from central Asia and other sources may be the parent material for the clayey, Fe-rich soils and inferred high radium concentrations. Elevated indoor radon levels may be expected in housing sited on these soil types on limestone terranes elsewhere at subtropical and tropical latitudes throughout the world where aerosol dust is a significant contributor to soil development.

INTRODUCTION

Elevated indoor radon levels are associated with a number of geologic settings throughout the United States in which uranium-rich rocks or soils occur in near-surface environments. Surprisingly, elevated soil radioactivity and elevated indoor radon concentrations also occur over uranium-poor limestone and dolomite rocks in the Piedmont and Valley and Ridge Provinces of the eastern U.S. (Sachs et al 1982). In the Piedmont and Valley and Ridge, the accumulation of uranium and radium is believed to occur in deep residual soils formed on the limestones and dolomites over very long periods of time (Greeman et al 1990). Very thick intervals of carbonate rock are dissolved over long periods of time and the uranium and radium are retained by residual clays and iron oxyhydroxides.

More recently, elevated indoor radon levels (to about 40 pCi/L) have been found in housing sited on late Pleistocene limestone in southwestern Dade County, Florida (Florida Dept. of Health and Rehabilitative Services, unpublished data, 1992; Moore and Gussow, 1991). However, in this area the limestone is very young, deep, residual soils are not present, and thin, clayey, limonitic soil horizons with thin, overlying sandy horizons are present. In addition, significant indoor radon levels (to 220 pCi/L) have been found on the island of Guam in the Mariannas Islands chain in the western Pacific. Here, the source for the high indoor radon levels also seems to be thin, clayey, limonitic soils developed on elevated marine limestone plateaus.

Herein, we propose a common origin for these radium-enriched soils in Florida and Guam and similar radium-enriched soils in the Carribbean. We further propose that the radium-enriched soils in the Carribbean and similar soils elsewhere in tropical latitudes are also likely to produce elevated indoor radon levels.

SOUTHEASTERN FLORIDA

Geographic and Geologic Setting

In Dade County and nearby areas of southeastern Florida, Pleistocene limestones, primarily the Miami Limestone, form the principal bedrock. The topography of the county is flat and only about 1.6 m above sea level except for the Atlantic Coastal Ridge, underlain by a bedrock high, which forms a low linear feature 1.6 to 5 m above sea level (Fig. 1). Throughout most of the area, the limestone bedrock is overlain by fine to medium quartz sand with lesser marl, peat, and muck (Fig. 2). The sand on and just west of the Atlantic Coastal Ridge was emplaced by storm surge events during hurricanes. In the northeastern part of the area, overlying sands are thick enough that a separate geologic unit, the Pamlico Sand, has been identified (Fish and Stewart 1991). In most areas, however, the depth to limestone bedrock ranges from 0 to 3m. In areas of central to southwestern Dade County where the limestone bedrock is at or just below the surface a distinctive soil type, the Rockdale soil, is present (area designated Rocklands in Fig. 2). This soil is most often reddish in color and the name Redlands has been used to describe the area of the county where these soils are present. Urbanization of the Miami metropolitan area (dotted line, Fig. 2) has proceeded from the north to the south and southwest along the higher ground comprising sandy soils of the Atlantic coastal ridge and along sandy flatlands and rocklands just to the west of the ridge.

Sections exposed at 10 excavations at construction and pipeline trench sites in Dade County show the presence of clayey, limonite-stained sands preserved in depressions and pockets in the moderately to deeply karstified surface of the Miami Limestone. Overlying these are sands, peaty sands, and peat distributed much like that portrayed in Fig. 2. The overlying sands seem to thicken from south to north along the Atlantic Coastal Ridge. These reddish clayey sands appear to be largely responsible for the reddish color characteristic of the Rockdale soils; however, they also appear to be widely distributed in the subsurface above the Miami Limestone and beneath other sandy and marl-bearing soil types mapped across the county.

Radiochemical and indoor radon data

During the National Uranium Resource Evaluation program conducted by the U.S. Department of Energy, aeroradiometric data was gathered for the entire U.S., including much of Dade County (see flightline locations in Fig. 3). These data showed that a broad area in the central part of the county exceeds 2 ppm eU (equivalent uranium, calculated from the ^{214}Bi gamma-ray intensity, assuming radioactive equilibrium) and locally reaches 5 ppm eU (Fig. 3). This area of >2ppm eU corresponds to the broad area of thin sandy soils developed on the Atlantic Coastal Ridge south of Miami and the adjacent Rockland soils to the west. These are areas where the clayey iron-rich horizon is at or near the surface. Note in Fig. 3 that the areas of intense urbanization do not yet extend far into areas of highest aeroradioactivity.

Indoor radon data for 126 houses in Dade County were gathered by the State of Florida during a statewide study of indoor radon conducted in 1986 (Nagda and others, 1987). These data showed that several zipcodes in the Homestead area in south-central Dade County had elevated levels of indoor radon.

Moore and Gussow (1991) noted the elevated radioactivity and indoor radon in the county. They sampled rock and surface soils for radium (n=88) across Dade County. Six samples of Miami Limestone ranged from 0.17 to 0.73 pCi/g. Surface soil samples ranged from 0.13 to 8.69 pCi/g with a mean value of 1.54 pCi/g. The samples highest in radium were from mapped Rockdale soils in areas of elevated, but not the highest, aeroradioactivity and in zipcodes with elevated indoor radon near Homestead. Soils composed of marl were lower in radium and soils composed of fine sand were lowest (0.13 to 0.77 pCi/g).

We collected and analyzed samples of the limestone, weathered limestone, and clayey, iron-rich sands, peaty sands, and clean, well-sorted fine sands that occur above the limestones at 10 construction sites from the northern edge of the area of high aeroradioactivity in Dade County. Fresh-appearing Miami Limestone contained 1.1 pCi/g radium but had a 0.05 emanating power (ep). Weathered Miami Limestone (9 samples) ranged from 0.8 to 3.4 pCi/g radium and ranged 0.05 to 0.33 ep. Clayey iron-stained sand, sandy clay, and brown loam (7 samples) filling solution pockets in the Miami Limestone ranged from 1.9 to 11.3 pCi/g radium and ranged in ep from 0.49 to 0.86. Clean sand and peaty sand (3 samples) overlying the Miami Limestone were below detection limits in radium (<0.4 pCi/g) and ep could not be measured.

The Florida Department of Health and Rehabilitative Services has also been gathering indoor radon data from this area (1548 measurements; N.M. Gilley, written commun., 1992). These data show that the areas of Rockdale soils and thin, fine sandy soils on the Atlantic Coastal ridge in central Dade County are areas where values above 4 pCi/L cluster in the dataset (Fig. 4). Values above 4 pCi/L in the northeastern part of the county (from the central part of Miami northward) all occur in multiple-story buildings with deep foundations (apartment buildings and schools). These data suggest that the clayey limonitic horizon is the principal source of radon in Dade County and that it can cause elevated indoor radon where it is near the surface or where deep foundations encounter the suspest horizon at depth.

GUAM

Geologic Setting

The island of Guam may be divided physiographically and geologically into two separate provinces; a northern province characterized by rolling uplands underlain by Pliocene and Pleistocene limestone which rest on a core of older, little- exposed volcanic rocks, and a southerly province composed of steep mountainous topography underlain by Eocene to Miocene basaltic to andesitic volcanic rocks, clastic sedimentary rocks derived from them, older limestones, and fringing areas of younger limestone uplands (Fig. 5, Tracey and others, 1964). The limestones of the northern province, formed largely from coral reefs, are extremely pure (0.1 percent acid-insoluble residue) and contain few components derived from the volcanic rocks. Limestones fringing the volcanic terrane are clayey and contain abundant clastic detritus derived from the volcanic rocks.

Soils developed on the northern limestone uplands are 10 to 25 cm thick except where deep karst pockets have formed (Guam soils; Young 1988). They are composed almost exclusively of alumina clays and iron oxyhydroxides, have very low silica (SiO_2) content (1 to 2 percent; Carroll et al 1963), and a modest phosphate content (0.68 to 2.5 percent P_2O_5). In contrast, the soils formed on the argillaceous limestones are 25 to 50 cm thick (Pulantat and Kagman soils; Young 1988) and have SiO_2 contents of 20 to 35 percent and iron (Fe_2O_3) contents of about 20 percent (Carroll et al 1963). Deeper residual soils (50 to 100 cm) have formed on the volcanic rocks (Akina, Atate, and Agfayan soils; Young 1988). These have even higher SiO_2 content of 25 to 50 percent and somewhat lower iron contents (10 to 20 percent).

Radiochemical and indoor radon data

Aeroradiometric data and radiochemical data for rocks and soils on Guam are not available. Gamma-ray logs of boreholes at waste disposal sites on the Air Force base on the northern part of Guam show counts in the soil horizons several tens of times count in the limestone (S. Terraciano, oral commun., 1993). The extremely low potassium content of these soils (<0.2 percent; Carroll et al 1963) suggests that most of this gamma count comes from uranium, thorium, or their decay products.

A screening survey of indoor radon levels in housing and schools in Guam has been completed by the Guam Environmental Protection Agency (Guam EPA) for areas throughout the island (Kladder et al 1991) and by the U.S. Air Force for residences and other buildings on Andersen Air Force base at the north end of the island. The Guam EPA school survey showed that 13 percent of classrooms overall tested above 4 pCi/L and that 57 percent of all schools had at least one classroom with radon levels above 4 pCi/L. In private residences (n=208) 27 percent exceeded 4 pCi/L and the highest reading was 143 pCi/L. Residences in villages on the limestones and argillaceous limestones had the highest readings. The Guam EPA data show that schools in villages sited on the limestone and argillaceous limestone yield the highest indoor radon levels for individual classrooms (maximum 117 pCi/L) and highest school average readings. Schools sited on the volcanic rocks yield no classroom readings over 1 pCi/L. The Air Force base is sited mostly on the pure carbonate reef rocks on which Guam soils have formed. The indoor radon levels on the Air Force Base (2 month alpha-track wintertime readings) average 7.9 pCi/L with 56.7 percent of 1406 readings exceeding 4 pCi/L.

These data and observations suggest that soils formed on the limestones and argillaceous limestones have accumulated significant amounts of radium in the soil profiles. Neither the very pure limestones which have had little time for deep weathering nor the volcanic rocks which are basaltic and andesitic in composition would seem to be good sources for the radium or its parent uranium.

POSSIBLE ORIGINS OF THE SOILS

Studies of the origins of the radium-enriched soils in Florida and the radon-producing soils in Guam have not been done. We suggest that, for southeastern Florida, studies of the origin of similar soils in the nearby Florida Keys, Barbados, Bahamas, and Jamaica offer a reasonable hypothesis. In these areas, red clay-rich soils ranging in age from about 125,000 to 870,000 yr have developed on Pleistocene coral limestone terraces and carbonate eolianites. Ratios of relatively immobile elements in these soils (Al_2O_3/TiO_2, Ti/Y, Ti/Zr, and Ti/Th- Muhs et al 1990b) lie slightly above ratios for these elements in Sahara dusts and significantly lower than the ratios for these elements in silicic volcanic ashes from sources in the Lesser Antilles volcanic arc. These data suggest that the soils are formed primarily from the Saharan dust source with a lesser contribution of volcanic ash.

Significant enrichment of radium occurs in these same soil profiles. Radium concentrations in soil profiles on Barbados range from 0.8 to 3.7 pCi/g (n=54); 1.2 to 6.2 pCi/g on Jamaica (n=29); 1.0 to 2.1 pCi/g on New Providence Island in the Bahamas (n=4); and 1.0 to 12.6 pCi/g in the Florida Keys east of Key West (n=4). Solution pipe-fill sediment on Bermuda ranges from 0.3 to 12.5 pCi/g (n=31) (D.R. Muhs and C.A. Bush, oral commun., 1993). These radium concentrations fall in the range of those observed by us and Moore and Gussow (1991) for iron-stained weathered limestone and clayey iron-rich soils sampled in Dade County.

The Saharan dusts contain significant amounts of uranium. Rydell and Prospero (1972) measured an average of 3.6 ppm U (about 1.2 pCi/g radium, assuming equilibrium) in modern airborne dust samples of Saharan origin (n=15) collected at Barbados. In these samples, uranium is in approximate radioactive equilibrium with its daughter products. However, a soil profile from Barbados displays disequilibrium excesses of uranium daughters such as ^{230}Th and ^{226}Ra (Muhs et al 1990a).

These dusts are not sufficiently enriched to explain the many higher radium readings (>1.2 pCi/g). The higher readings are probably due to the concentration of the decay products of uranium during intense weathering of the aerosol dusts. Uranium (and many other major soil constituents) is likely to be highly mobile in the soil environments present on these islands, but ^{230}Th is readily sorbed by clays and iron oxyhydroxides under these conditions and would tend to be retained with the residual material (Muhs et al 1990a). ^{226}Ra either follows ^{230}Th geochemically or grows into radioactive equilibrium with it in about 7000 years.

The thin, reddish, radium-rich, clayey soils of the Redlands area (the Rockdale soil) and reddish, radium-rich, clayey horizons found above the Miami limestone beneath other sand and marl soil layers in Dade County are herein thought to be similarly formed from Saharan dust parent material although further study will be needed to evaluate other potential sources such as uraniferous phosphatic material from the Hawthorn Group.

Geochemical studies of the origins of deep-sea pelagic muds in the western Pacific (see Olivarez et al 1991 for a review) suggest that these muds are derived, in part, from aerosol dusts from central Asian sources. We suggest that soils developed on the very pure limestones on the northern part of Guam are derived, in part, from aerosol dusts from central Asia with a significant component of locally derived aerosol dust. Soils formed on the argillaceous limestones in the southern part of the island are likely derived, in part, from this source and from the clayey material in the underlying limestones and alluvial material and windblown dust from the volcanic uplands. Although to our knowledge no radium measurements have been made of soils developed on limestones from Guam, because of the unusually high indoor radon levels, we expect the values to be similar to and perhaps exceed those seen in soils in the Carribbean and southern Florida.

CONCLUSIONS

Elevated indoor radon levels observed in southeastern Florida and Guam may have a common geologic origin. Radium-enriched soils formed by weathering of aerosol dust from distant and local sources deposited on limestone terranes rather than from residual soils formed from the limestones themselves. These dusts contained both uranium and radium but soil-forming processes probably caused separation of uranium and radium and preferential enrichment in radium. High levels of indoor radon may be associated with similarly formed soil types developed over limestone substrates in Jamaica, Barbados, the Florida Keys and other tropical coastal areas.

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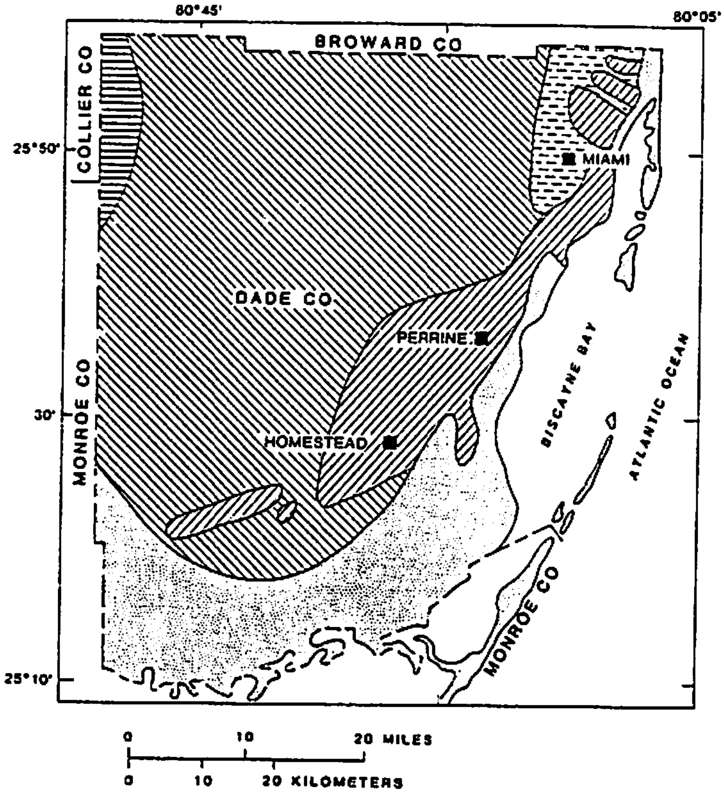


Figure 1- Physiographic features of Dade County prior to development. From Fish and Stewart, 1991.

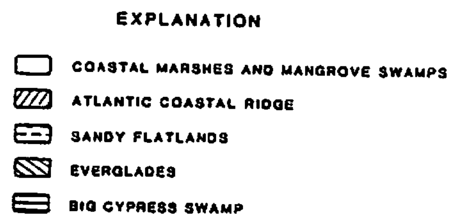
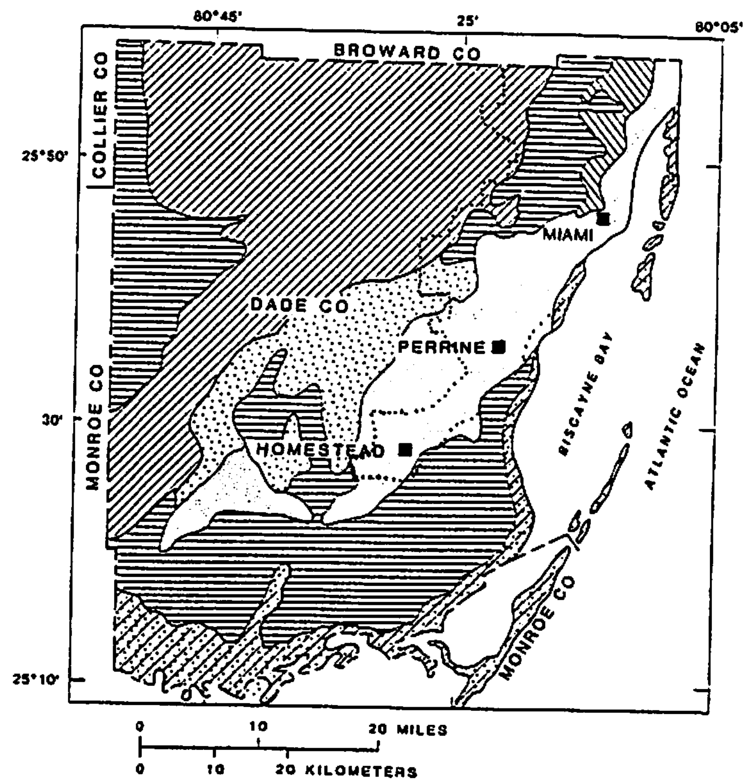
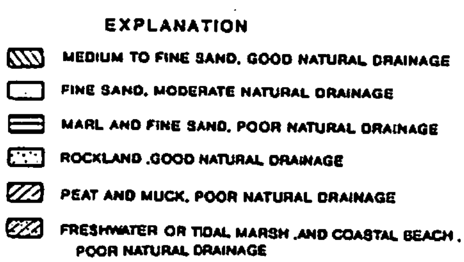


Figure 2- Distribution of soil types in Dade County. From Fish and Stewart, 1991.





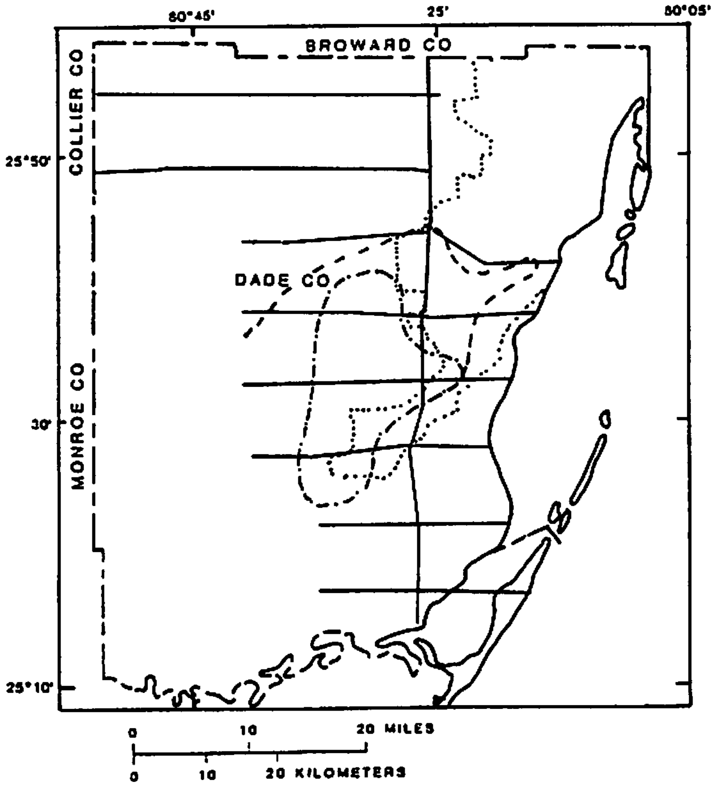


Figure 3- Map showing NURE aerorad data for Dade County from U.S. Department of Energy, 1981 Solid lines show flightline locations in the county. Dashed line encloses area of values of eU ranging from 2.0-2.5 ppm. Dash-dot line marks boundary of area ranging from 2.5-5.0 ppm eU. Dotted line marks extent of intensive development in the county.

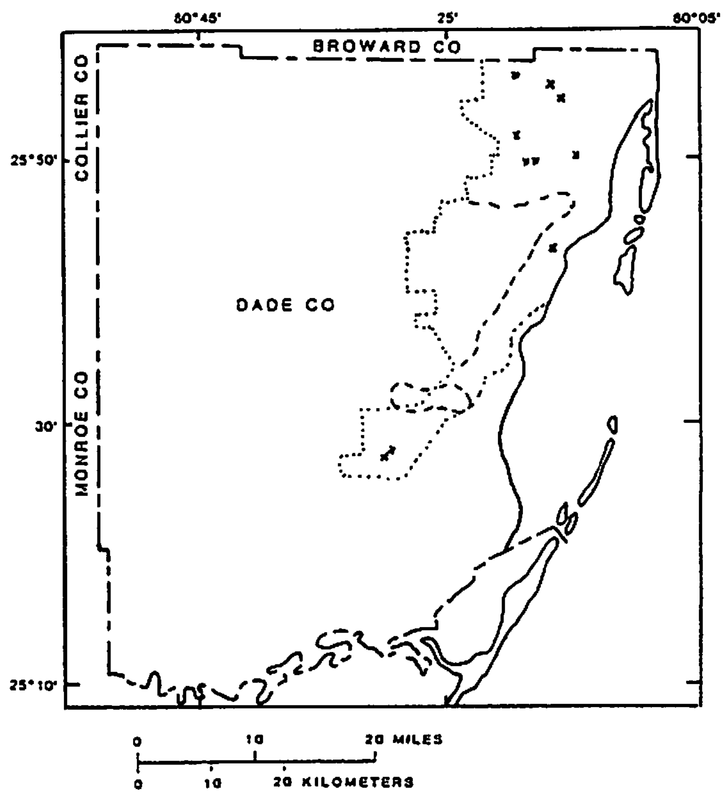


Figure 4- Location of indoor radon values over 4 pCi/L in the Florida Department of Health and Rehabilitative Services dataset. Dashed line marks areas that contain a large number of houses of >4 pCi/L (n=55). X-isolated occurrences. Dotted line marks extent of intensive development.

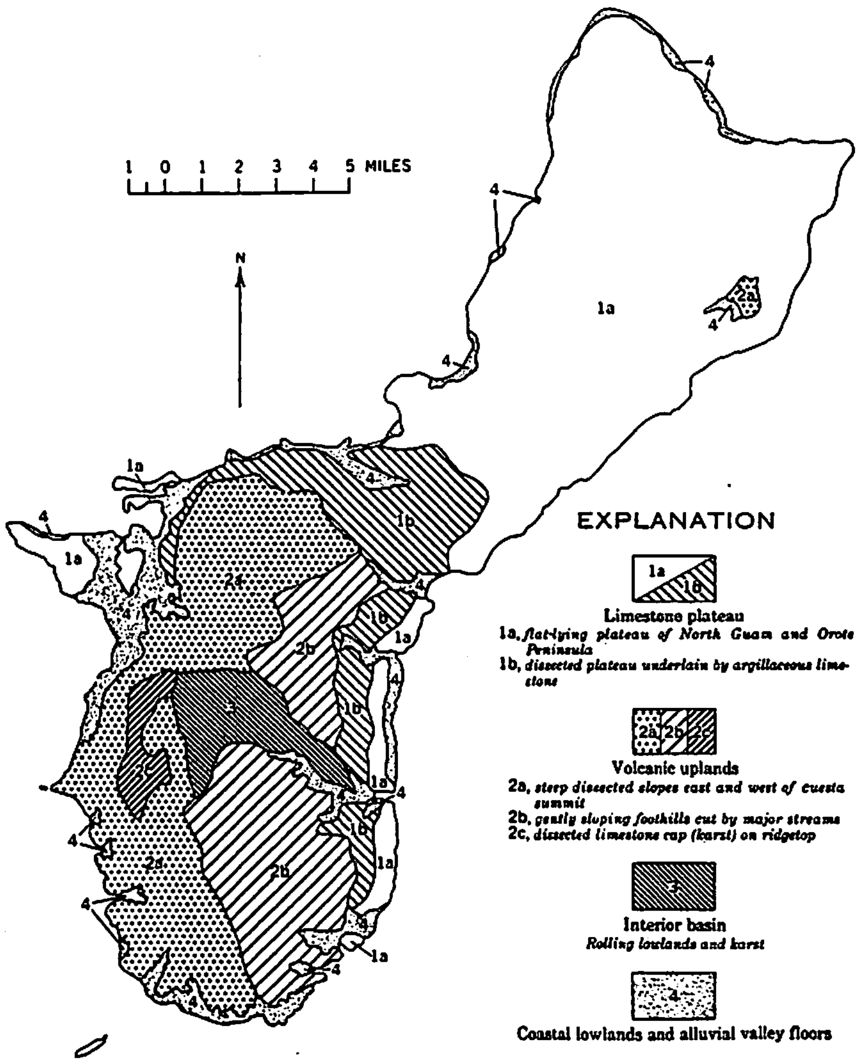


Figure 5- Map showing physiographic provinces of Guam. From Tracey and others, 1964.