data from all observations clearly show that the effect is universal. The data from the two missions need not agree because the ground tracks did not overlap in detail at the mare edge crossings and because there may have been additional radon emanation during the 9 months between observations.

There are two results which any model for the origin of the edge effect must explain:

1) The distribution of ^{210}Po activity is sharply peaked at the edges of the maria.

2) The ^{210}Po activity exceeds that of its progenitor ^{222}Rn at the edges of the maria, implying that the process which leads to the emission of ^{222}Rn is varying in time. The time scale of the variation is less than a few times the effective half-life of ^{210}Po (21 years).

In any hypothesis based on an external process, which frees radon from lunar material, for example, meteorite bombardment or mass slumping, it would be necessary to explain why the process is so pronounced at the edges of the maria. Also, an external process would involve an amount of lunar material that is too large. For example, if one assumes that all the existing radon is released instantaneously from all the lunar material disturbed by a meteorite impact, then the total mass of lunar material required is several tenths of a gram per square centimeter per year. In view of the persistence of lunar surface features on a time scale of 10^9 years, the large mass requirement appears to rule out external processes as a source of radon.

Let us hypothesize that radon is a very minor component of the gases released at the lunar surface. The peak activity rate of ^{210}Po at the edge of Mare Fecunditatis is 1.6×10^{-2} disintegration $\sec^{-1}$ cm^{-2}. If we assume radioactive equilibrium for the purpose of this estimate, the implied peak diffusion rate of ^{222}Rn is approximately twice the decay rate of ^{210}Po. Averaged over the lunar surface, the diffusion rate would be of the order of 10^{-2} atom $\sec^{-1}$ cm^{-2}. Measurements of other gases in the lunar atmosphere indicate that the total concentration of the sum of O_2, CO_2, N_2, CO, and H_2O, which are mostly of solar origin, is less than a few times 10^4 molecule cm^{-3} on the nighttime side (*3*). We assume that these gases have a scale height of 10^7 cm and a loss time in the lunar atmosphere of 10^7 seconds (*4*). Therefore, if radon atoms constituted only one part in 10^5, on the average, of gases emitted from the surface, present measurements of the density of the lunar atmosphere would not be exceeded. Middlehurst (*5*) and Cameron (*6*) have studied historical records of transient optical events on the lunar surface. The phenomena have been recorded over several centuries of observation and include sudden brightenings, color changes, and obscurations. Middlehurst has noted that the sites of these events tend to group at the mare edges and near prominent young craters. The general correlation of both transient optical events and time-varying radon emanation with the edges of the maria is particularly significant in light of the fact that the crater Aristarchus, from which we detected ^{222}Rn emission during the Apollo 15 mission (*1*), is the most frequently reported site of transient events. Salisbury *et al.* have mapped the spatial distribution of dark-haloed craters on the visible lunar surface (*7*) and have found that they tend to occur at the edges of the maria. These craters are generally small, are surrounded by material of low albedo, and may be of volcanic origin (*8*). A factor that could be common to the polonium effect, the transient optical events, and the occurrence of dark-haloed lunar craters is transient venting of volatile materials at the edges of the maria. The polonium enhancement could be due either to variable diffuse venting over a large portion of the mare perimeters or to a few well-localized centers. Perhaps optical events are detectable when the gas venting is most pronounced and localized. If it is true that dark-haloed craters are of volcanic origin, then in the past the edges of the maria were areas at which gas venting occurred. The results presented here establish that at least one gas, radon, is being emitted from the edges of the maria at the present time.

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Radon-222 from the Island of Hawaii: Deep Soils Are More Important than Lava Fields or Volcanoes

Abstract. *The mean flux of radon-222 atoms from the island of Hawaii is 0.45 atom per square centimeter per second. Lava fields occupy 50 percent of the land area, but their radon flux is only 1 percent of that from deep volcanic soils. The island yields approximately 10 curies of radon-222 per hour to the air surrounding it. The radon-222 contribution of volcanoes is negligible.*

The island of Hawaii, situated 3800 km or more from the nearest upwind landmass, is ideally located for the use of naturally occurring ^{222}Rn (half-life $T_{1/2} = 3.82$ days) and its long-lived daughter ^{210}Pb ($T_{1/2} = 22$ years) in both local and large-scale circulation studies (*1*). The purpose of the work reported here was to determine the ^{222}Rn exhalation from the lava and soils in order to provide a better understanding of the distribution of ^{222}Rn and its daughter nuclides in the air over the island.

On the basis of earlier studies of radon exhalation (*2*), it can be expected that the land surface of Hawaii will contribute ^{222}Rn to the island atmosphere at an average rate dependent upon three factors: (i) the concentration of ^{226}Ra in the rocks and soils of the island; (ii) the fraction of the total number of ^{226}Ra atoms that are located on or near the surface of mineral grains and soil particles in a position that favors the escape of the newly formed daughter atoms into the soil gas; and (iii) the physical character-

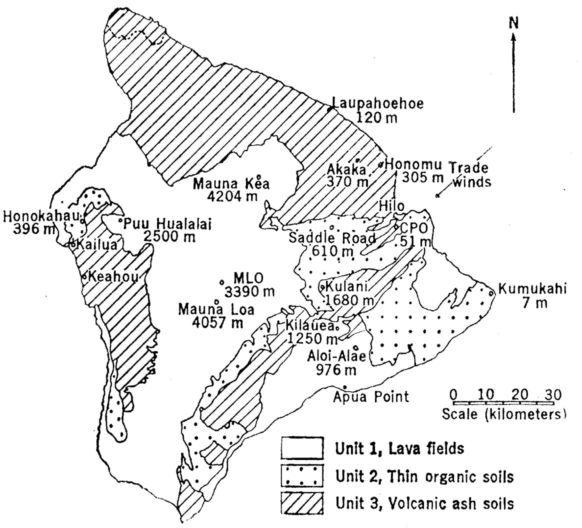

Fig. 1. The island of Hawaii showing soil units used in the ^{222}Rn exhalation study; *MLO*, Mauna Loa Observatory, National Oceanic and Atmospheric Administration; *CPO*, Cloud Physics Observatory, University of Hawaii.

istics of the surface and near-surface lavas and soils, which determine the rate of diffusion of radon within the soil gas and the rate of transport of radon across the earth-air interface. The data presented in this study clearly reflect the relative importance of these factors in the widely differing island soil types. The ^{222}Rn contributed to the atmosphere by volcanic activity relative to the amount of ^{222}Rn diffusing from the soil is of special interest on the island of Hawaii.

The land area of Hawaii may be divided into three major soil classification types for determining the total radon exhalation. This division (shown in Fig. 1) is based on information from the Hawaii County soil map (*3*). Unit 1 consists of barren lava fields; unit 2 (thin organic soils) is made up predominantly of areas where a very thin layer of organic matter covers older lava flows; and unit 3 is composed of soil associations which together make up the finer-grained, relatively deep, agricultural soils of the island. The soils of unit 3 originated primarily as volcanic ash from Mauna Kea except for a small area in the extreme northwestern tip of the island where the soils are from weathered basalt. Since these latter soils comprise only 4 percent of the total, they are omitted as a separate group in this study.

The areas of the three units are given in Table 1. Barren lava fields account for one-half of the island surface. A large fraction of this unit lies above the trade wind inversion beginning at about 2100 m above sea level.

The radium analyses listed in Table 1 are mean values of two or more determinations for soils from the locations given. A total of 30 samples were analyzed from among the three units. The errors given are standard deviations calculated from the total samples in each unit. I used the radon method for the ^{226}Ra determinations, following standard procedures (*4*).

The ^{226}Ra content in lavas from Mauna Loa and Cape Kumukahi, 0.24 ± 0.09 pg of ^{226}Ra per gram of lava, is in general agreement with the mean value of Evans and Goodman (0.31 ± 0.03 pg/g) for 35 basic igneous rocks (*5*). The earlier work by Piggot (*6*) on Hawaiian igneous rocks gave radium concentrations considered by Evans and Goodman to be too high, but it is significant that Piggot found little change in radium concentration in samples from Mauna Loa, Hualalai, and Mauna Kea lavas, nor did he find much difference in radium concentrations in lavas taken from the same area but from different eruptive ages.

Radium concentrations for the thin organic soils were nearly the same as for the lavas, thus reflecting their direct association with the lavas in most cases. The deep soils exhibited quite variable radium concentrations, and the average is appreciably higher than that of the lavas. The ^{226}Ra concentration in soils is known to vary, depending upon source rock, weathering, and absorption processes.

The "effective radium" (^{226}Ra-eff) in Table 1 is the radium which, by virtue of its location on or very near the surfaces of soil particles or mineral grains, supplies radon atoms to the soil gas. When this quantity is divided by the total ^{226}Ra concentration, one obtains the "emanating power" (^{226}Ra-eff/^{226}Ra) as defined by early workers in the field (*7*). It is the fraction of the total ^{226}Ra present that is capable of supplying radon atoms to the soil gas and, hence, to the atmosphere. The emanating power is very low, 2 percent, for the lava fields, 55 percent for the organic soils, and 70 percent for deep soils as shown in Table 1. The samples of Mauna Loa lava and cane field soil were analyzed independently by Barretto (*8*). His "escape-to-production" results are analogous to the ^{226}Ra-eff/^{226}Ra ratios presented here. He found values of 2 percent for the lava fields and 65 percent for the cane field soils, in good agreement with the corresponding figures from my analyses.

The low value of emanating power found for the lavas indicates that, although ^{226}Ra is present in the volcanic materials, the ^{222}Rn atoms are trapped

Table 1. Radium concentrations and radon flux on the island of Hawaii.

Sampling areas	Area (km^2)	^{226}Ra concentration (pg/g)	^{226}Ra-eff concentration (pg/g)	Emanating power	^{222}Rn flux (atom cm^{-2} sec^{-1})	Total ^{222}Rn exhalation (× 10^{15} atom/hour)
			Lava fields			
Unit 1 (Mauna Loa, Kumukahi)	5210	0.24 ± 0.09	0.005 ± 0.003	0.02	0.012 ± 0.003	2
			Thin organic soils			
Unit 2 (Hilo, Kulani, Saddle Road)	2190	0.20 ± 0.08	0.11 ± 0.07	0.55	0.08 ± 0.02	6
			Deep agricultural soils			
Unit 3 (Akaka, Honomu, Laupahoehoe)	3020	0.7 ± 0.2	0.50 ± 0.21	0.7	1.5 ± 0.4	160
Total flux					1.7×10^{17} atom/hour $\simeq 10$ c/hour	

in the crystalline and glassy substances formed as the magma cools. Escape by recoil at the instant of decay of the ^{226}Ra is limited to a thin layer a few hundredths of a micrometer thick on the surface of solid particles, and diffusion from greater distances within the lava is severely limited by the 3.82-day half-life of ^{222}Rn. Weathering and other soil-formation processes which would increase the escape probability of the radon atoms have little significance in the geologically recent lavas found on the island surface. On the other hand, unusually high emanating power is associated with the fine-grained deep soils of the island which originated from volcanic ash resulting from the explosive eruptions of Mauna Kea. This activity occurred at a much earlier age than the Mauna Loa–Kilauea flow type of eruptions which are largely responsible for the major lava fields of the island. The details of the escape mechanisms of radon from rock, minerals, and soils is a subject of continuing research at several laboratories.

The ^{222}Rn flux measurements listed in Table 1 were made by the accumulator method (*9*) and represent a total of 13 separate determinations distributed among the three soil units. The errors given include those due to random uncertainties in a single determination totaling about 30 percent. The low flux values for the Mauna Loa and Cape Kumukahi sites reflect the low emanating power of lavas, and the high values found in unit 3 are consistent both with the relatively high ^{226}Ra concentration and the high emanating characteristics of these deep soils. The weighted mean flux for the island based upon values in Table 1 is 0.45 atom cm^{-2} sec^{-1}. This can be compared with 1.1 ± 0.3 atom cm^{-2} sec^{-1} for New Mexico soils and a worldwide estimate of 0.75 atom cm^{-2} sec^{-1} based on 994 published radon flux measurements from a wide variety of soil types (*9*).

The final column of Table 1 gives the total exhalation of radon from each of the three soil units calculated from their areas and mean fluxes. Within the limitations imposed by the relatively small sample obtained, the total exhalation from the island is 10 c of ^{222}Rn per hour, of which almost 95 percent originates from the deep soils of the island. The rate of ^{222}Rn production can be expected to fluctuate on both diurnal and annual scales, to be sensitive to atmospheric pressure changes, and, to a lesser extent, to change with rainfall and prevailing and local winds.

No consideration of the island of Hawaii as a source of atmospheric radon would be complete without an analysis of the contribution made by volcanic activity. Crustal rocks contain small concentrations of ^{226}Ra; hence, it is reasonable to expect that ^{222}Rn should be present in gases which escape at the time of volcanic eruptions. One measure of the radon contribution can be obtained from the total mass of lava expelled, on the assumption that all of the ^{222}Rn contained in the magma is released at the time of eruption. During 1969 an estimated 58×10^6 m^3 of lava were expelled from the active upper east rift zone of Kilauea. This was the most lava produced in a 1-year period since 1955 when an estimated 115×10^6 m^3 were ejected in the Puna district. A density of 2.8 g/cm^3 is used from Piggot's determinations, and a radium concentration of 0.24 pg per gram of lava is taken from Table 1. If *all* the equilibrium quantity of ^{222}Rn is removed at the time of eruption, then for the 10-month period of the Aloi-Alae eruption series 48 c would have been contributed to the atmosphere of Hawaii. This quantity, equivalent to 6.6×10^{-3} c/hour, is negligible compared with the 10 c/hour determined from flux measurements. The work of Björnsson on Surtsey samples indicates that only about 2/3 to 9/10 of the radon from hot lavas actually escape during the eruptive phases (*10*).

An ideal method of obtaining the ^{222}Rn contribution from volcanoes would be to sample active vents directly. A fumarole sampled on 27 June 1969, 34 hours after the end of phase IV of the Aloi-Alae eruption, was found to have a radon concentration of 1.4 pc/liter above that of air. It would take 7×10^7 fumaroles of this type, expelling gases continuously, to equal the contribution of the natural radon flux from the island surface. Björnsson (*10*) and Iwasaki *et al.* (*11*) have measured the ^{222}Rn concentrations in gases from volcanoes in Iceland and Japan, respectively. Although their results vary widely, none are more than an order of magnitude greater than the value found for the Aloi-Alae fumarole given above.

An unusual opportunity for taking air samples above, within, and below the trade wind inversion over the island occurred during phase III of the Aloi-Alae eruption on the upper east rift zone of Kilauea during 12–13 June 1969. Volcanic ash and gases were effectively trapped under conditions which permitted vertical profiles of radon concentration to be taken aboard an aircraft within a few hours after the end of the eruption. They showed only a slight excess of ^{222}Rn concentration at altitudes where the haze and sulfur fumes were at maximum levels. The net increase was only 0.004 pc/liter, which, by estimating the volume involved, could have been supplied by 0.2 c/hour for the 22-hour duration of phase III of the Aloi-Alae eruption. Even when averaged over the extraordinary year of 1969, which had 12 eruptions, the number of curies per hour is negligible in comparison with the 10 c/hour from surface exhalation.

On the basis of these considerations, exhalation from deep soils is found to far exceed lava fields or volcanic activity as the chief contributor to ^{222}Rn in the air over the island of Hawaii.

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