Paper

CALCULATIONS OF BACKGROUND BETA-GAMMA RADIATION DOSE THROUGH GEOLOGIC TIME

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Abstract—**Life on earth is exposed to a background level of ionizing radiation from a number of sources, including beta and gamma radiation from geologic and biologic materials. Radiation dose from geologic emitters has changed because of the chemical evolution of the continental crust, changes in the relative abundances of ^{235}U and ^{238}U, and the radioactive decay of uranium, thorium, and ^{40}K with time. The radiation dose from internal ^{40}K has decreased by a factor of about eight because of changes in the activity concentration of ^{40}K in potassium over the past 4 billion years. Radiation exposure from geologic materials has decreased from about 1.6 mGy y^{-1} to 0.66 mGy y^{-1} over the past 4 billion years, and radiation exposure to an organism with a potassium concentration of 250 mmol L^{-1} has decreased from about 5.5 to about 0.70 mGy y^{-1}. Accordingly, background radiation exposure from these two sources has dropped from about 7.0 to 1.35 mGy y^{-1} during the time life has existed on Earth. The conservative nature of mutation repair mechanisms in modern organisms suggest that these mechanisms may have evolved in the distant past and that organisms may retain some of the capability of efficiently repairing damage from higher radiation levels than exist at present.**

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Key words: radiation, background; linear hypothesis; naturally occurring radionuclides; radioactivity, environmental

INTRODUCTION

IONIZING RADIATION interacts with organisms at the cellular level by modification of DNA, leading to mutations that generally range in severity from harmless to lethal (with occasional beneficial mutations). Cellular biochemistry has evolved mechanisms to repair genetic damage caused by ionizing radiation and other mutagens. These mechanisms are common to a wide variety of organisms (Prasad 1995), suggesting their evolution early in the history of life. At the time life evolved in the oceans about 3.5–4.0 billion years ago (Schopf 1992), Earth's background radiation field was significantly higher than at present. It is possible that the mutation repair mechanisms that exist today reflect the response of early life to the higher background radiation field under which they evolved. This could provide a geological context to the threshold in radiation dose-response in modern organisms noted by some researchers (Kondo 1994; Luckey 1991).

The three primary components to beta-gamma radiation dose in the distant past include geologic emitters (including nuclides dissolved in seawater), internal emitters (primarily ^{40}K), and cosmic and cosmogenic sources. Radiation dose from cosmic and cosmogenic sources, while potentially significant (Terry and Tucker 1968; Karam 1998), is the subject of continuing research by us and is not considered here.

Radiation dose due to radon and radon progeny is not considered because most of the damage caused by radon and radon progeny is to the lungs and gills of organisms. Lungs and gills are relatively recent developments in the history of life (evolving about 350 to 400 million years ago), and radiation dose due to radon and radon progeny is virtually unchanged in that time. This paper does not address alpha-emitting nuclides because of the likelihood that life evolved in the oceans, the low range of alpha particles in water, and the relatively low dose from dissolved alpha-emitting nuclides to marine and freshwater organisms (UNSCEAR 1996). While radon is a significant source of radiation exposure to many people and to animals dwelling underground (NCRP 1987), it is not considered in this work for the reasons mentioned above, although a more thorough treatment is warranted at some time in the future.

DOSE DUE TO GEOLOGIC RADIONUCLIDES

The continental crust is enriched in uranium, thorium, and potassium compared to the bulk Earth (Condie 1993). Continental crust covers not just the continents, but part of the seafloor as well. The continental shelves are composed of continental crust, and sediments derived from the continents cover the continental shelves and beyond. The rate at which the continental crust has evolved chemically over geologic time has directly impacted the radiation dose received from geologic materials over time.

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Two general rock types are considered here, felsic (rich in feldspar, silica, and enriched in uranium, thorium, and potassium) and mafic (rich in Mg, Fe, and depleted in uranium, thorium, and potassium). Felsic rocks include granite, dacite, rhyolite, and others that comprise the greater part of the continents. Mafic rocks include basalt, gabbro, andesite, and similar rocks that are typical of oceanic or ocean island settings. Over time, mafic rocks have become increasingly depleted in large ions (including uranium, thorium, and potassium) while felsic rocks have become increasingly enriched in these elements.

The chemical evolution of the continental crust is subject to great debate. Three general models exist: early formation, late formation, and constant formation. The rock record seems to indicate that the majority of the continental crust had formed by about 2.5 billion years ago (Ga) (Condie 1989) and that the rate of crust formation has since slowed (Condie 1993). It appears that the chemical concentrations of uranium, thorium, and potassium in felsic and mafic rocks have not changed dramatically over geologic time, although the radioactivity concentrations have decreased due to radioactive decay (Condie 1993; Karam 1998). We use the continental crust formation model developed by Karam (1998) and shown in Fig. 1 to describe the fraction of continental crust that was felsic in character during the past 4 billion years. This model describes activity concentrations of uranium, thorium, and potassium in mafic and felsic rocks of various ages using dose conversion factors noted in Eisenbud and Gessell (1997) to derive a radiation dose rate for organisms living on contact with them. The derivation of this model is described in the following section.

The chemical evolution of the continental crust

Large ions such as uranium, thorium, and potassium tend to concentrate in the liquid phase of solidifying magma. As a result, magma expelled at or near Earth's surface tends to contain higher concentrations of these elements than does the source rock for the magma. This is why granites and other felsic rocks generally have higher levels of uranium, thorium, and potassium than do basalts, which are derived from source rocks in the mantle that have been depleted in large ions through time.

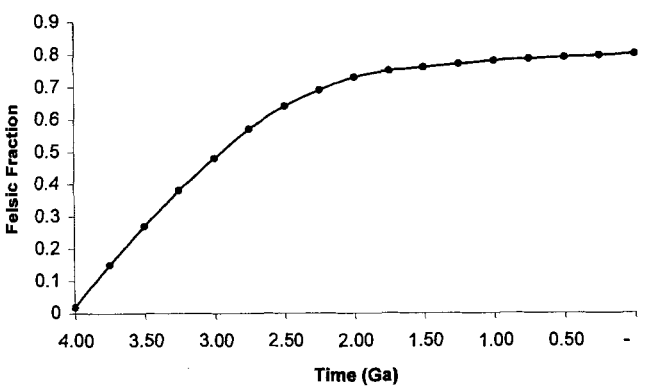

Fig. 1. Model for continental crust growth (Karam 1998). This model assumes that continental crust formation took place relatively early in the history of Earth, and has slowed considerably since about 2.5 billion years ago (Ga). Like most continental crust growth models, this model suggests that over half of the continental crust was formed by 2.5 Ga, in agreement with the rock record.

A database was compiled containing the results of more than 1,100 rock samples reporting concentrations of uranium, thorium, and/or potassium in rocks of various ages in order to determine chemical concentrations of these elements with respect to time for felsic and mafic rocks. Data including rock age, rock type (mafic or felsic), and the uranium, thorium, and/or potassium concentration were collected, corrected for radioactive decay over the age of the rock,[‡] and plotted as elemental concentration vs. time for both mafic and felsic rocks. This resulted in six plots: uranium, thorium, and potassium concentrations vs. time for mafic rocks and felsic rocks. Curves were fit to these data using statistical regression tools in Microsoft Excel for Office 97.[§]

This analysis yielded formulae for determining changes in the chemical concentration of each element with time for mafic and felsic rocks. From these curves we determined the radioactivity concentrations in rock using standard radioactive decay calculations and, from the radioactivity concentrations we used the dose conversion factors discussed elsewhere in this paper to derive radiation dose on contact with felsic and mafic rocks in the past.

The final phase in constructing a model that describes the chemical evolution of the continental crust involved determining the portion of continental crust composed of felsic and mafic rock in the past. We used a model showing early, rapid formation of the continental crust in which the majority of the crust was formed by 2.5 Ga, and in which the rate of continental crust formation slows with time (Condie 1989; Taylor and McLennan 1985). This model was used to determine the approximate fraction of the continental crust composed of felsic rocks through time, and this fraction and the resulting fraction of mafic rocks in the continental crust were used as weighting factors for the calculated radiation dose at times in the past.

Of particular interest is the dose from uranium over time because the abundance of ^{235}U in natural uranium has changed greatly, from nearly 17% 4 billion years ago to 0.72% today. The ^{235}U decay series emits fewer gamma rays than that of ^{238}U, with a lower cumulative gamma energy. ^{235}U should have a correspondingly lower dose conversion factor that is proportional of the sum of gamma energies and emission probability through the entire decay series. The cumulative series gamma decay energies for ^{235}U and ^{238}U were determined by multiplying the gamma ray energy by the probability of emission and the branch ratio for a particular nuclide as shown in eqn (1):

[‡] It is not standard practice for geochemical papers to perform this correction. Such data are almost invariably reported in terms of chemical concentration measured at the present time and not at the time the rock formed.

[§] Microsoft, One Microsoft Way, Redmond, WA 98052-6399; 1997.

$$E_{tot} = \sum E_\gamma I_\gamma f_n, \quad (1)$$

where E_γ is the gamma energy, I_γ is the intensity (or probability) of emission, and f_n is the branching ratio to the nuclide emitting the gamma ray. These decay series energies were used to determine a dose conversion factor for ^{235}U using the value of 0.376 mGy y^{-1} for each Bq g^{-1} of ^{238}U activity in a rock (Eisenbud and Gessell 1997). The dose conversion factor for ^{235}U was calculated to be 0.111 mGy y^{-1} for each Bq g^{-1} of ^{235}U activity present. The dose rate from uranium in rocks was corrected for the changing ^{235}U abundance using eqn (2):

$$D_U = C_u[0.376f_{238} + 0.111(1 - f_{238})], \quad (2)$$

where C_U is the concentration of uranium in the rock, f_{238} is the fraction of uranium that is ^{238}U, 0.376 is the dose conversion factor for ^{238}U and progeny nuclides, and 0.111 is the dose conversion factor for ^{235}U and progeny nuclides. Table 1 summarizes the dose conversion factors, ^{235}U abundance, and relative abundance of uranium at 500 million-year intervals from 4 billion years ago until the present. The relative abundance of uranium in this table refers to the fact that, due to radioactive decay, a rock containing one gram of uranium today would have contained 2.2 g of uranium 4 billion years ago, the remaining uranium having decayed in the intervening years.

In Fig. 2, changes in uranium, thorium, and potassium radioactivity concentrations with time are shown. Fig. 3 shows similar changes in radiation dose in the model continental crust from these elements (and series nuclides) with time, and Fig. 4 shows changes in mafic rocks, felsic rocks, and the model continental crust over time. The dose from geologic materials at any time in the past depends on the concentration of radionuclides in the continental crust, the specific activity of the element, and (in the case of uranium) the radiation dose per unit activity concentration

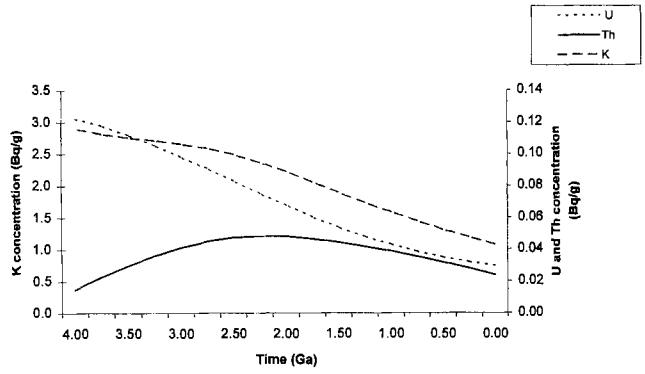

Fig. 2. Age-corrected uranium, thorium, and potassium radioactivity concentrations in the model continental crust through time. Note that at all times ^{40}K is present in greater radioactivity concentrations in the bulk continental crust. Changes in thorium concentrations are due primarily to the formation of felsic continental crust because felsic rocks have a much greater thorium concentration than do mafic rocks.

at that time in the past. In these figures, progeny nuclides are assumed to be in secular equilibrium at all times because these rocks are assumed to be closed systems with respect to potassium, uranium, and thorium series nuclides. While this is a simplifying assumption, its use is justified by noting that many rocks with ages of hundreds of millions to billions of years are routinely dated using radiometric techniques based on similar assumptions.

Radiation dose rates from potassium and thorium in rocks were determined using the dose conversion factors of 0.378 and 0.584 mGy y^{-1}, respectively, for each Bq g^{-1} of activity in the rock (Eisenbud and Gessell 1997). These dose conversion factors were applied to the concentrations of potassium and thorium present in the continental crust as described above.

Table 1. Some changing properties of uranium with time. All columns show the changes in an arbitrary volume of rock containing 1 g of elemental uranium at present. The numbers of atoms of ^{235}U and ^{238}U at present were calculated using the atomic weight of uranium, and the atomic weight and the current abundances of these two isotopes. The numbers of atoms of ^{235}U and ^{238}U in the past were calculated using the law of radioactive decay. The total uranium decay energy was calculated using the values from Wetherill (1966) for ^{235}U, ^{238}U, and progeny nuclides. Specific activities for ^{235}U and ^{238}U are from Schleien (1992). Dose conversion factors were calculated as described in the text. Total uranium decay energy, dose conversion factors, and specific activity calculations in the past were determined by weighting them for the relative abundance of ^{235}U and ^{238}U.

Age (Ga)	#^{238}U atoms per gram uranium ($\times 10^{21}$)	#^{235}U atoms per gram uranium ($\times 10^{19}$)	Age-corrected weight of uranium (g)	Atom % ^{235}U	Total uranium decay energy (MeV)	Specific activity of elemental uranium ($\times 10^4$ Bq g^{-1})	Dose conversion factor (mGy y^{-1} per Bq g^{-1} uranium in rock)
0.0	2.51	1.82	1.00	0.72	47.38	1.29	0.374
0.5	2.71	2.98	1.08	1.09	51.34	1.31	0.373
1.0	2.93	4.87	1.18	1.64	55.68	1.35	0.372
1.5	3.17	7.97	1.28	2.45	60.47	1.41	0.369
2.0	3.42	13.0	1.40	3.67	65.80	1.49	0.366
2.5	3.70	21.3	1.55	5.45	71.76	1.61	0.362
3.0	4.00	34.9	1.72	8.03	78.48	1.78	0.355
3.5	4.32	57.1	1.93	11.7	86.12	2.03	0.345
4.0	4.67	93.4	2.21	16.7	94.85	2.37	0.332

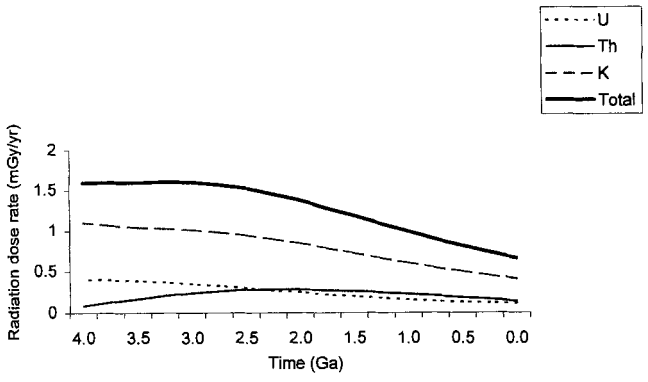

Fig. 3. Radiation dose rate from geologic emitters through time. As in Fig. 2, most of the radiation dose at any time in the past is due to ^{40}K because of the high concentration of potassium in felsic rocks. Note that for nearly two billion years the rate of continental crust formation was approximately the same as the integrated rate of radioactive decay, resulting in nearly constant radiation dose rates from geologic materials during this time.

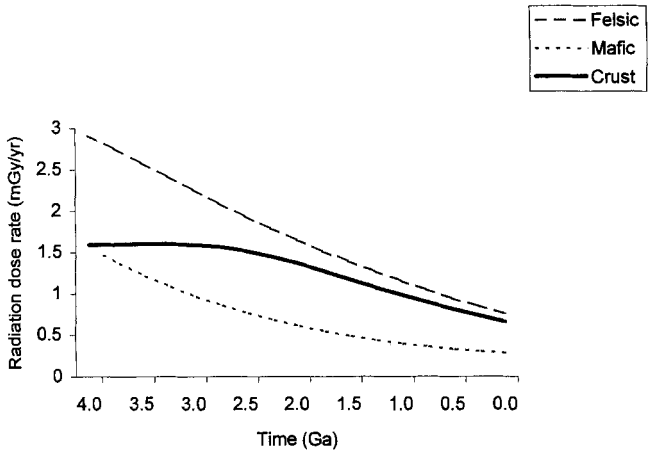

Fig. 4. Radiation dose rate from uranium, thorium, and potassium in mafic rocks, felsic rocks, and the model continental crust through time. Mafic rocks are those rocks (such as basalt) with high Mg and Fe concentrations and felsic rocks are rich in feldspar and silica (such as granites). The model continental crust has the proportions of mafic and felsic rocks shown in Fig. 1.

The calculated radiation dose from all geologic materials was about twice that of current levels when the first continental crust formed about 4 billion years ago (Fig. 3). This dose was virtually identical to that from mafic rocks at that time because very little felsic crust existed. Dose increased gradually until about 2.5 Ga, reflecting the partition of radionuclides into the continental crust (i.e., continental crust formation) at a faster rate than they decayed. As the continental crust formation rate slowed, radiation levels began to fall, reflecting the decay of uranium, thorium, and, most importantly, potassium. The modeled radiation level at present (0.66 mGy y^{-1}) is higher than the 0.28 mGy y^{-1} typically attributed to dose from geologic materials (NCRP 1987) because soils (included in the 0.28 mGy y^{-1} figure) are usually deficient in uranium, thorium, and potassium compared to rocks, and our calculated doses reflect the chemical composition of rocks alone. It is likely that soils resembling those at present did not begin to form until the colonization of land by large plants about 380 million years ago. Before this time soils would be expected to closely resemble, both chemically and radiologically, the underlying rock. Our calculated dose does compare favorably with doses derived from radionuclide composition information reported in Eisenbud and Gesell (1997) and UNSCEAR (1958) (0.71 mGy y^{-1} and 0.77 mGy y^{-1}, respectively). Our calculated dose is less than that noted in UNSCEAR (1996) to benthic molluscs from gamma-emitting nuclides in sediments (1.40 mGy y^{-1}). According to this report, this high dose is because "The fine particles making up the food scavenge radionuclides efficiently from the water, thus leading to higher body burdens of the radionuclides."

The curves calculated for radiation dose from mafic and felsic rocks (Fig. 4) should be considered the lower and upper constraints for radiation dose from geologic materials at any time in the past. The intermediate curve (Fig. 4) represents our best estimate of what the radiation dose might have been, based on our model of continental crust formation. Even assuming the continental crust always consisted entirely of mafic rocks (radiologically the most conservative assumption possible, and undoubtedly untrue) suggests that early life living on contact with continental crust was exposed to radiation doses that were significantly higher than those at present.

DOSE DUE TO INTERNAL RADIONUCLIDES

Internal emitters include ^{40}K, ^{14}C, and ^{3}H, among others. ^{40}K is a primordial radionuclide with a half-life of 1.28 billion years. Tritium and ^{14}C are produced by cosmic ray interactions with atmospheric gases, and their concentrations should reflect solar activity and galactic cosmic ray flux. Radiation dose from internal ^{40}K is between 0.28 and 0.40 mGy y^{-1} (UNSCEAR 1993 and NCRP 1987, respectively) for organisms with potassium concentrations of 150 mmol L^{-1}, the concentration found in mammalian muscle. Radiation dose from homogeneously-distributed radionuclides should remain constant regardless of organism size because each unit mass receives approximately the same deposition of energy from radioactive decay. Mammalian muscle tissue has a low concentration of potassium compared to other organisms, especially compared to prokaryotes and archaea. Table 2 lists the radiation dose from a variety of internal ^{40}K concentrations at various times in the geologic past. We have modeled radiation dose to organisms with a potassium concentration of 250 mmol L^{-1} because that is the potassium concentration of the average bacterium (Dodson and Dodson 1985) and single-celled organisms have dominated the history of life on Earth (Margulis and Sagan 1986).

While the half-life of ^{40}K is long (1.28 billion y), life is thought to have evolved about three ^{40}K half-lives ago, resulting in substantially higher dose at that time. Fig. 5 shows the calculated dose from ^{40}K over geologic time to organisms with a potassium concentration of 250 mmol

Table 2. Radiation dose rate (mGy y^{-1}) vs. geologic time for a variety of biological potassium concentrations. The time in the past is in billions of years (Ga) in the left-most column, while the potassium concentrations for which calculations were performed are in bold numbers in the top row. The "average" bacterium has a potassium concentration of about 250 mmol L^{-1} (Kirschner 1991) and mammalian muscle has a potassium concentration of about 150 mmol L^{-1}.

Time (Ga)	Potassium concentration (mmol L^{-1})								
	100	150	200	250	300	350	400	450	500
Present	0.37	0.56	0.74	0.93	1.12	1.30	1.46	1.67	1.86
0.5	0.49	0.73	0.98	1.2	1.46	1.71	1.95	2.19	2.44
1.0	0.64	0.96	1.28	1.6	1.92	2.24	2.56	2.88	3.20
1.5	0.84	1.26	1.68	2.10	2.51	2.93	3.35	3.77	4.19
2.0	1.10	1.65	2.20	2.75	3.30	3.85	4.40	4.94	5.49
2.5	1.44	2.16	2.88	3.60	4.32	5.04	5.76	6.48	7.20
3.0	1.89	2.83	3.78	4.72	5.67	6.61	7.55	8.50	9.44
3.5	2.48	3.71	4.95	6.19	7.43	8.66	9.90	11.14	12.38
4.0	3.25	4.87	6.49	9.11	9.74	11.36	12.98	14.60	16.23

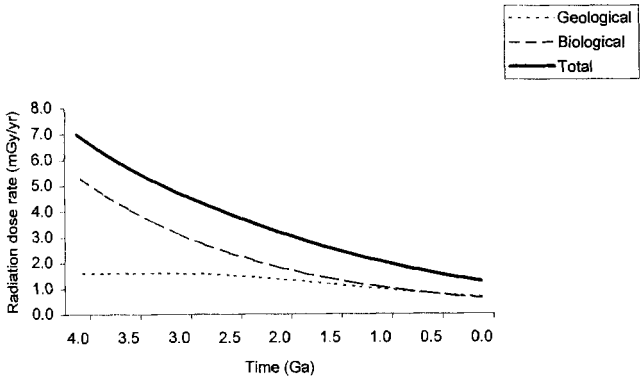

Fig. 5. Radiation dose rate from geological and biological radionuclides through time. The curve for radiation dose rate from biological sources assumes a potassium concentration of 250 mmol L^{-1} and changes according to the actual potassium concentration in an organism. The dose from biological potassium assumes that early organisms were sufficiently thick to stop all ^{40}K beta radiation (about 85% of the total decay energy) but allowed the gamma rays to escape. This is reasonable because the maximum range of the ^{40}K beta particle is about 0.66 cm in tissue and many early organisms are thought to have existed as algal or bacterial mats at least that thick.

L^{-1}. ^{40}K decays by emission of a 1.33 MeV beta particle 89% of the time and by emission of a 1.46 MeV gamma 11% of the time. Fossil remains of early organisms suggest that much of early life consisted of mat-forming colonies at least 1 cm thick. We assumed that all beta energy was deposited within the organism and that all gamma energy (about 15% of total ^{40}K decay energy) was lost, contributing no dose. Using these assumptions, the first organisms were exposed to ^{40}K radiation dose rates of at least 6.2 mGy y^{-1} and the first eukaryotic life, which evolved about 2 billion years ago (Runnegar 1994), was exposed to radiation dose rates of about 3.6 mGy y^{-1}; dose rates that have steadily dropped to current levels.

DISCUSSION

Early life was exposed to much higher background radiation fields than is the case today. The radiation doses due to internal and geologic emitters over the past 4 billion years are shown in Fig. 5. We have summed these curves to arrive at a calculated background radiation dose rate from geologic and biologic sources since life evolved on Earth. At the time life first arose (about 3.5 to 4.0 billion years ago) it was likely exposed to a background radiation field of at least 6.2 mGy y^{-1}, and probably more, depending on the exact potassium concentration of early organisms. This is significantly higher than the average background beta-gamma radiation dose rate to organisms today.

Background radiation presently accounts for 1–6% of background mutations (Prasad 1995). The radiation-induced mutation rate is thought to be directly proportional to radiation dose, so radiation may have accounted for up to 33% of mutations to the first life forms.[‖] The actual fraction of radiation-induced mutations was probably different because the impact of other mutagenic agents in the environment has changed with time as well, as have oxygen levels and other relevant factors. It is likely that mutation repair mechanisms in modern organisms are conservative (Mackinodan and James 1990); i.e., they are very similar in widely disparate kingdoms such as monera (bacteria), archaea (archaea bacteria, including thermophilic bacteria and sulfur-reducing bacteria), and animalia (the animal kingdom). Based on this observation, it is most parsimonious to assume that they evolved only once, in the common ancestor to all modern life forms, before life diverged to form the modern kingdoms. If this is the case, then the mutation repair mechanisms in humans likely evolved in an environment that had up to eight times the background radiation levels we experience today. This possibility gives a geological and historical context to the idea of a threshold level, below which life's cellular repair mechanisms can adequately repair radiation damage with little or no expected harmful effects to the organism.

The earliest life consisted of prokaryotes (bacteria) and archaea, small cells with few organelles and no nucleus.

[‖] Consider an organism undergoing 100 mutations of which 6 are radiation-induced. If mutation rate is directly proportional to radiation dose, then an eight-fold increase in radiation dose would result in a total of 142 mutations, of which 48 are radiation-induced. This results in an overall increase in mutation rate of 42% of which nearly one third are radiogenic.

Prokaryotes, especially archaea, can live in extreme environments not inhabited by other kingdoms of life, although they are common in virtually all environments on Earth. Eukaryotes are thought to have evolved about 2 billion years ago (Runnegar 1994) as the result of symbiosis between prokaryotes (Margulis and Sagan 1986). Eukaryotes are larger and more complex than prokaryotes, containing numerous organelles and a nucleus containing genetic information. Yeast, many micro-organisms, and all "higher" plants and animals are composed of eukaryotic cells.

Eukaryotic cells are generally more sensitive to the effects of radiation than are prokaryotes and archaea (UNSCEAR 1996). This sensitivity could reflect the greater complexity of eukaryotic cells, making their functioning easier to disrupt; the relatively greater chromosome volume, giving more "targets" for mutating events; their evolution in a background radiation field lower than that in which prokaryotes evolved; some combination of these; or another cause entirely. In any event, it is interesting to note that many mutation repair mechanisms are shared by both prokaryotic and eukaryotic cells, while others are unique to each kingdom of life (Prasad 1995).

The United Nations Science Committee on the Effects of Atomic Radiation (1996) published a table showing lethal radiation dosage ranges for a number of phyla. In general, more recently-evolved organisms show higher sensitivity to the effects of radiation as evidenced by lower lethal dose ranges. It is tempting to claim that this demonstrates the evolution of these organisms in successively lower radiation fields over time. However, with the possible exception of eukaryotes noted above, we feel this is an oversimplification because multicellular life forms evolved quickly with respect to the half-life of ^{40}K (the primary source of radiation exposure), so dose rates did not change significantly between the evolution of molluscs (about 550 million years ago) and the first mammals (about 200 million years ago).

CONCLUSION

The background radiation field at the surface of the Earth due to potassium, uranium, thorium, and decay series radionuclides in the continental crust was about 1.6 mGy y^{-1} at the time that life evolved and remained relatively constant for almost 2 billion years. The radiation dose rates from internal ^{40}K have decreased steadily since life evolved from about 5.5 mGy y^{-1} to about 0.70 mGy y^{-1} at present.

The presence of significantly elevated levels of background radiation over most of the history of life on Earth suggests that modern organisms may be able to repair radiation damage resulting from exposure to radiation doses higher than those currently found in nature because mutation repair mechanisms are thought to have evolved early in the history of life. This may, in turn, provide a historical context for the current debate regarding the existence of a threshold level below which radiation exposure may do no harm.

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