

LETTERS

Pu Breeders in the Sky Not a Burning Issue after All

In a recent letter (May, page 88) Alex Gabbard posed the question of whether "plutonium is being bred in the biosphere by natural, but unidentified, means." He then went on to suggest that "tiny quantities of plutonium bred through entirely natural mechanisms could significantly alter the radiotoxicity of the biosphere."

The answer to the question of whether there is a natural source of plutonium was provided in 1942 by Glenn Seaborg and Morris Perlman,¹ who discovered Pu-239 in a sample of Canadian pitchblende that contained 13.5 percent uranium. The plutonium is thought to be produced from uranium via the reactions $U-238(n,\gamma)$ $U-239 \xrightarrow{\beta} Np-239 \xrightarrow{\beta} Pu-239$.

Subsequent work by Seaborg and others² demonstrated conclusively that plutonium does occur in mineral deposits at levels up to approximately $[Pu-239]/[U-238] = 10^{-11}$. Given the 2.7-barn thermal-neutron capture cross section of U-238, one infers that a thermal-neutron fluence of 3.7×10^{12} per square centimeter was required to produce the level of Pu-239 observed in these ore samples. The plutonium now present in such ores must have been produced recently (that is, over a period comparable to the 24 000-year halflife of Pu-239). Since the flux of neutrons used in breeding the plutonium is intimately tied to the alpha-decay rates of U-238 and thorium-232, it is reasonable to believe that this plutonium was produced at a constant rate over this time. This implies that the thermal-neutron flux in these ore samples was approximately 4 per square centimeter per second. The flux of neutrons that can be produced by the spontaneous fission of U-238 is much less than that amount. Thus it was inferred that the most likely source of the neutrons was (α,n) reactions on light elements contained in the ore sample. The alpha particles themselves are produced by the decays of uranium, thorium and their daughters contained in the ore.

Free neutrons are present at and near Earth's surface as a result of interactions between cosmic rays and

the constituent nuclei present in the atmosphere (nitrogen-14, oxygen-16 and so on). The neutron flux at Earth's surface is approximately $1 \times 10^{-2} \text{ cm}^{-2} \text{ s}^{-1}$ and increases by less than a factor of 200 in going from the surface to an altitude of 40 000 feet.³ Most of these neutrons become thermalized through collisions with nuclei and eventually undergo an (n,p) reaction on N-14 to produce carbon-14. From measurements of the C-14 levels in the biosphere,⁴ one can thus infer that the average neutron flux in the atmosphere is approximately $3 \times 10^{-2} \text{ cm}^{-2} \text{ s}^{-1}$. If one then assumes that uranium was dispersed into the biosphere at a constant rate over the last 100 years through the burning of coal and during that time was exposed to this average atmospheric neutron flux, then this "natural mechanism" would have produced $[Pu-239]/[U-238] = 1.4 \times 10^{-16}$. Taking Gabbard's estimate of 5.1×10^9 tons of coal burned in 1991 and a uranium concentration of 2 parts per million in the coal as being representative, the amount of Pu-239 produced from neutron capture on U-238 on or near Earth's surface would be at most 0.2 milligrams. This quantity of naturally produced plutonium should be compared with the roughly 5×10^3 kilograms of Pu-239 released into the biosphere by atmospheric nuclear weapons tests!⁵ Thus the truly negligible amounts of plutonium that are naturally produced from neutron-capture reactions on the ashes from coal burning play virtually no role in the radiotoxicity of the biosphere.

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LETTERS (continued from page 15)

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The letter by Alex Gabbard presents a stunning number—"The radiotoxicity of plutonium is more than 11 orders of magnitude greater than those of natural uranium and thorium"—and then draws inferences both for the environment ("could significantly alter the radiotoxicity of the biosphere") and for the allocation of research funds ("appears to be another grand challenge for science").

I assume that "order of magnitude" is just the usual pompous way of saying "factor of 10." Gabbard is thus saying that Pu-239 is 10^{11} times more radioactive than U-238; that the radiation is 10^{11} times more deadly than that of uranium; or that the product of the two effects is 10^{11} . The correct figures are 180 000 for the ratio of the activities of equal numbers of atoms and about 1.2 for the ratio of the biological hazards,¹ making Pu-239 2.2×10^5 times as hazardous as U-238 on a per-atom basis. Therefore Gabbard is in error by a factor of 4.5×10^5 . In other words, he overestimates the hazard by a factor of about a half-million. A similar overestimate of the hazard of driving would lead one to believe that the entire population of Earth would be killed off in three months of driving on US highways!

By way of comparison, radium-226 is about 40 times as hazardous as Pu-239 on a per-atom basis and vastly more prevalent in the environment, even if all of the U-238 released in coal burning were converted to Pu-239.

It is well understood that occasional environmental neutrons may transmute U-238 to Pu-239, and it is well documented that there is uranium in coal. But there is nothing unique about the uranium in coal with respect to concentration, with respect to dispersion or with respect to absorption of neutrons. To gain some appreciation of natural sources, one merely needs to reflect on the enormous amount of uranium-containing dust that has been blown into the atmosphere by many natural processes, including dust storms, volcanic eruptions and forest fires, during the last halflife of Pu-239, a time that goes back to when woolly mammoths

and saber-toothed cats roamed Earth. If plutonium were to build up to appreciable levels because of airborne uranium, it would surely have done so.

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I agree with Alex Gabbard that breeding plutonium-239 by cosmic-ray neutrons captured in uranium-238 adsorbed to particles of coal smoke is theoretically possible. However, the amount of plutonium produced by this mechanism is so small that it does not deserve any practical attention.

Let us make a rough estimate. Suppose that 10 billion metric tons of coal containing 1 part per million of uranium are burned per year and that 10 percent of the uranium escapes with the smoke into the atmosphere, where it remains suspended for one year. The total amount of U-238 released per year is thus 1000 tons, or 2.5×10^{30} atoms. Assuming that the volume of the atmosphere is roughly 10^{25} cubic centimeters, we get 2.5×10^{5} cm⁻³ as the average equilibrium concentration of U-238 atoms in the air.

It is much more difficult to obtain a reasonably accurate estimate of the flux and energy of neutrons, particularly in the upper layers of the atmosphere where most of the neutron-capture reactions in U-238 seem to take place. Drawing on data normally used in dosimetry,¹ we may conservatively assume that the average total neutron flux in the atmosphere is on the order of 1 cm⁻² per second and that the average neutron energy is in the interval between 1000 and 1 million electron volts. Assuming 1 barn to be a conservative approximation of the U-238 average neutron-capture cross section in this interval and using the average equilibrium concentration of U-238 atoms, we get 2.5×10^{-19} cm⁻¹ as an estimate of the macroscopic cross section for the Pu-239 breeding reaction. We get the corresponding reaction rate by multiplying this estimate by the total neutron flux. The result is 2.5×10^{-19} reactions per cubic centimeter per second. This total corresponds to approximately 8×10^{13} reactions in the entire atmosphere per year. The weight of Pu-239 produced in these reactions is approximately 3×10^{-8} grams. The radioactivity of this amount of plutonium is about 100 becquerels, which is approximately equivalent to the natural

radioactivity of 1 cubic meter of air in a concrete building owing to radon decay. According to the standards of the International Commission on Radiological Protection, this amount of Pu-239 would contaminate approximately 10 kilograms of drinking water.² The contribution of other Pu isotopes is comparatively negligible.

Owing to uncertainties in the calculation (particularly in determining the effective neutron flux and energy), the results can be one or two orders of magnitude wrong. However, even if we assume that 100 times more plutonium is produced by coal burning, the total amount is still only on the order of milligrams.

Compare such an amount with "artificial" plutonium releases to the environment resulting from the reprocessing of reactor fuel. Typically, 150 kilograms of Pu-239, 70 kilograms of Pu-240, 30 kilograms of Pu-241 and 15 kilograms of Pu-242 are discharged as spent fuel from a 1000-megawatt electric pressurized water reactor per year. As there are approximately 430 nuclear power plants operating in the world³ with total net electric power of around 350 000 MW_e, the total world production of plutonium is at least 90 000 kg per year, not counting military production. This amount is 16 orders of magnitude more than the "natural" plutonium bred in the sky. Of course, only a tiny fraction of this artificial plutonium leaks into the environment. However, even if we assume that the artificial releases are on the order of grams per year, they significantly exceed those from natural sources.

Overestimating the importance of natural plutonium could lead environmental scientists down the wrong path. On the other hand it could also redirect the attention (and funds) of the nuclear community away from important problems concerning the reprocessing and disposal of plutonium.

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The letter by Alex Gabbard misses what is by far the most important source of radiotoxicity due to coal burning: The uranium impurity in the coal and its thorium-230 and radium-226 daughters end up in the

ground and eventually become radon, an important radiotoxic that the Environmental Protection Agency estimates is killing 14 000 Americans per year.

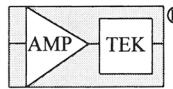
Let's calculate this effect.¹ The 3.8-day half-life of radon allows the radon to diffuse up through about 1 meter of soil before decay. From standard measures of US land area, average soil density and uranium concentration, one can calculate that the amount of uranium in the top 1 m of US soil totals 5.4×10^7 metric tons. From the rate at which rivers carry dissolved and suspended material into the oceans, one can calculate that the surface of the US is eroding at a rate of 4.5×10^{-5} m per year, which means that the average residence time of an atom in the top meter of soil is about 22 000 years. Over that period, therefore, the uranium in the soil will cause a total of 3.1×10^8 deaths, or 5.7 deaths per ton of uranium.

In addition, the 1.3 parts per million of uranium in the 8×10^8 tons of coal burned annually in the US will end up in the ground and thus eventually kill 5700 Americans. If we assume roughly that this uranium is distributed through the top 5 m of US soil, these deaths will occur over the next 110 000 years.

One might argue that if the coal were not mined, erosion would cause this uranium to eventually reach the surface anyhow after some millions of years, resulting in the same number of deaths from the same uranium. However, burning coal makes the carbon unavailable as a soil material (it is converted into carbon dioxide), and when erosion would have brought this unmined carbon to the surface, its volume near the surface will be taken by other rock, which we treat as "average rock." The 2.7 ppm of uranium in this average rock, replacing the carbon in the coal that by definition is not uranium (the uranium in the coal has already been accounted for), causes additional radioactivity. One year of coal burning in the US will thus eventually cause something like 8600 deaths.

In summary, each year of burning coal at current levels of consumption will result in the deaths of 5700 Americans over the next 110 000 years and of 2900 more Americans over the following few millions of years—solely as a consequence of radiotoxicity.

It is interesting to contrast the effects of burning coal for energy with those using nuclear power, which removes uranium from the ground and thus saves people from radon's radio-



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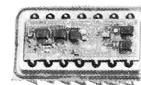
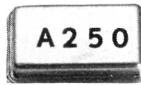
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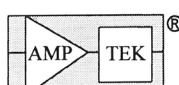
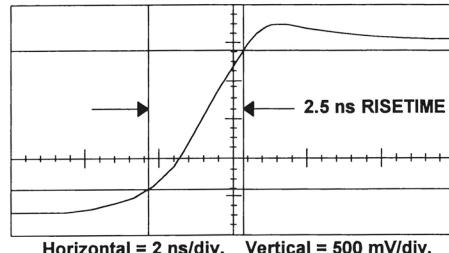
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toxicity. In principle, the 800×10^8 tons of coal burned annually in the US could be replaced by mining 40 000 tons of uranium, eventually saving 230 000 lives by removing the uranium from the ground, in addition to the 8600 lives that would be saved by not burning coal.

The conclusion is very clear: If one considers the very-long-term effects of radiotoxicity, coal burning is a major killer, and nuclear power is a major lifesaver.

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GABBARD REPLIES: My letter, under the headline "Can Coal Combustion Breed Pu in the Sky?" (May, page 88), has generated considerable discussion, which has prompted a re-evaluation of the data I utilized from the 1990 reference document,¹ a standard at this facility. Calculations of specific data relevant to my letter have shown the reference data to be in error to such an extent that the points I raised are no longer at issue. When the 1960 source document² was consulted, the erroneous reference data could not be duplicated and now stand as an inexplicable error that apparently was repeated in the 1990 document used in my study. Consequently, the recalculated radiotoxicity data are in line with international standards, and discussions of natural-source plutonium are of no significance.

I trust that this resolves the issue.

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Universities Use Their Faculties Well

In his broadside (August, page 13) against the participants, such as the undersigned, in PHYSICS TODAY's roundtable on research universities (March, page 42), Martin E. Ross poses a series of questions, such as "How many of the introductory lectures and laboratory courses at their

respective universities are taught by graduate students?" As for Cornell University's physics department, I can report that all such lectures are given by faculty, and largely by senior faculty, as are a substantial portion of discussion and laboratory sections.

At any time, several of these courses are undergoing major renovations led by senior faculty and requiring sizable commitments in university and departmental resources. Cornell is not a singular case; many other research universities have departments with very strong research and graduate programs and are led by faculties deeply committed to undergraduate education.

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sions of cosmology include several such examples—the problem does become a serious concern. Errors in scientific logic, exaggerated claims and confusion between what is known and what is assumed have become alarmingly common in cosmology.

The second case in point involves the respected cosmologist P. J. E. Peebles, who stated in a recent paper: "A semiempirical cosmogony uses a general framework within which one *adjusts parameters to fit observations* [emphasis added]. If this cannot be done it shows the framework is wrong. If it can, the parameter adjustment may force us to a useful approximation to reality; the evidence would be that the result accounts for more observations than there are free parameters."³

But hold! I hear shades from Ptolemy's era explaining the rationale for using epicycles to model retrograde motions of planets. This is not the scientific method—a system that has worked beautifully for centuries. The standard version of the scientific method is as follows. (1) Observe nature objectively (not with mathematical or philosophical prejudices in mind). (2) Hopefully an idea, theory or model, based on a principle or pattern, will emerge. (3) If the hypothesis has real merit, it will lead to definitive predictions.⁴ (4) Empirically test the predictions and humbly accept the results. If a prediction fails, do not tweak the model or fudge the data; instead go back to (1).

Perhaps our concerns have been inflated, but a growing number of us see cosmology as a loose cannon. The scientific method is a priceless guide to knowledge; we should not accept dubious substitutes.

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JAKOBSEN REPLIES: While we agree with Oldershaw that research in cosmology is often somewhat wanting in terms of scientific rigor and that, as he implies, one should choose one's words with great care when publishing in this field, my coauthors and I beg to differ in the case at hand and plead not guilty to the accusations made.

In keeping with Oldershaw's expla-