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Tallahassee, FL 32306
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Dr. Dade Moeller
Advisory Committee on Nuclear Waste
P-315
Nuclear regulatory Commission
Washington, DC 20555

Dear Dade,

Attached is a brief summary of the ideas that occurred to me as a consequence of the October 26, 1990, ACNW subcommittee meeting on the carbon-14 problem in HLW repositories. Marty Steindler asked for a "pithy" report, and so here it is. I hope it is of some use in providing a geologic perspective on the problem.

Sincerely yours

Joseph F Donoghue

Joseph F. Donoghue
Associate Professor

cc: M. Steindler

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Atmospheric Carbon-14 Variation over Geologic Time and Its Implications for High Level Waste Disposal

Evaluation of the health effects of carbon-14 in high level waste is unlike that for nearly all of the other radionuclide components of the waste. C-14 is extremely mobile, unlike most of the actinides, and occurs predominantly in the form of carbon dioxide gas.

In the case of uranium- or thorium-series nuclides it is a relatively simple geochemical problem to compare the repository inventory with that of a typical natural ore body, both in terms of concentration and migration rates. The natural background level of radiation from such nuclides can also be determined with a reasonable level of accuracy.

For C-14, however, the natural background level in the atmosphere has varied considerably over geologic time, perhaps by as much as an order of magnitude. The reason for the wide variation is the fact that radiocarbon is affected by a variety of major geologic cycles.

These cycles include: (1) Changes in the flux of cosmic radiation, caused by variation in the solar constant. (2) Changes in the outer core affecting the earth's external magnetic field, which in turn affects cosmic ray flux to the upper atmosphere and the rate at which the (n,p) reaction on N-14 generates new radiocarbon. (3) Variation in the global climate cycle, which affects the rate of production of organic carbon. Organic carbon removes carbon dioxide from the atmosphere or ocean and incoporates it into sedimentary rocks for intermediate to long periods of time. (4) Changes in the global tectonic cycle, which lead to overproduction of marine limestone ($CaCO_3$) during times of high sea level and underproduction of limestone during times of low sea level. Limestone and other carbonate rocks sequester carbon dioxide, removing it from solution in seawater, thereby lowering the partial pressure of carbon dioxide in the atmosphere.

The earth's magnetic field fluctuations can substantially affect the production of radiocarbon. The earth's magnetic field is usually modeled as a dipole. There is evidence in igneous rocks and marine sediments that the earth's internal field has reversed polarity frequently over geologic time. There has been a major reversal on the average every 200,000 yr over the past 5,000,000 yr, the last one (Brunhes-Matuyama) having occurred approximately 730,000 yr before present. Evidence of reversals extends back at least through mid-Mesozoic time (approximately 200,000,000 yr ago). During a reversal there is some relatively long period during which the field strength is low. During such a time cosmic ray flux to the earth's atmosphere and surface must be proportionately higher.

As an example of the extent to which changes in the earth's magnetic field might affect atmospheric radiocarbon concentrations, a recent study of beryllium-10 in marine sediments is instructive (Railsbeck, et al., 1985). Beryllium-10, like carbon-14, is produced in the upper atmosphere by the action of cosmic radiation. It can be expected to vary inversely with changes in cosmic ray flux. Direct measurement of the variation of Be-10 in marine sediments across the Bruhnes-Matuyama boundary reveals an increase of Be-10 production by a factor of 2 to 3 during the polarity reversal event. The same is probably true of C-14. This is thought to be a conservative estimate of overproduction of cosmogenic radionuclides during a reversal. The theoretical increase is approximately a factor of eight.

The concentration of CO_2, and therefore C-14, has also varied due to the global climatic variations of the Ice Ages. Examination of CO_2 trapped in ice cores from Antarctica has provided a record of atmospheric CO_2 content and paleotemperatures over the past 160,000 yr. During the last interglacial period, about 140,000 yr ago, CO_2 concentrations reached a maximum of approximately 300 ppm. During the subsequent glacial period, the concentration dropped to approximately 170 ppm (Baranola, et al., 1987; Genthon, et al, 1987). The current level is about 350 ppm (Moore and Bolin, 1987). These fluctuations have commensurately augmented and diluted the concentration of C-14 in the atmosphere.

The variation of carbon dioxide, and radiocarbon, in the atmosphere as a result of tectonic cycles is even more pronounced. Over Phanerozoic time, i.e., the past approximately 570 million years, examination of the rock record indicates that the amount of carbon dioxide in the atmosphere has varied by more than an order of magnitude, from a high during the early Paleozoic (c. 500 ma. BP) to a low during Pennsylvanian time (c. 300 ma. BP), and a relative low at present (Berner, 1990).

Over the past approximately 8,000 yr, atmospheric C-14 levels have varied considerably, at rates as high as 1% per 20 years. This variation appears to be related to the sunspot cycle, and exhibits a periodicity of about 200 yr (Neftel, et al., 1981).

Additionally, man's activities over the past century have measurably affected C-14 concentrations in the atmosphere. The increased combustion of fossil fuels that has accompanied the Industrial Revolution has increased C-12 concentration in the atmosphere and correspondingly decreased C-14 concentration by about 2%, a phenomenon known as the "Suess effect" (Faure, 1977). An opposite effect has resulted from the production of C-14 in the atmosphere by atmospheric testing of nuclear weapons since the early 1950's. Atmospheric concentrations of C-14 increased by nearly an order of magnitude during the peak of the weapons testing in the early 1960's (Telgadas, 1971).

I think these examples make it clear that the inventories and potential concentrations of C-14 discussed at the ACNW workshop are very minor relative to the natural levels. In light of the wide range of variation in atmospheric C-14 over geologic time, and even over historic time, releases of C-14 from an HLW repository have to be considered to be below regulatory concern. Even if the worst-case, nearly impossible, scenario of an instantaneous release of the entire inventory of C-14 were to occur, the result would be a less than 0.1% increase in the present global inventory of C-14.

Additional Comments Resulting from the October 26 Meeting:

(1) Most of the C-14 inventory of both PWRs and BWRs is in the core hardware, rather than the fuel elements: about 50% for PWRs and about 70% for BWRs. This core hardware is, for the most part, materials other than zircaloy cladding (NCRP, 1985, pp. 14-15). All of the speakers at the ACNW workshop focused on the C-14 in the fuel and cladding, which is in a gaseous or oxide state, and therefore relatively mobile. But it would appear that most of the reactor C-14 inventory is not in fuel or cladding, it is relatively immobile, and also may not end up in the HLW repository anyway.

(2) If most of the C-14 that is expected to be sent to the repository is yet to be produced, why not focus on decreasing production, rather than contemplating the expenditure of several billion dollars to bring the waste package design into compliance with the EPA and NRC disposal rules? Much of the reactor-produced C-14 is already being released to the atmosphere through normal reactor operations. More of it could be released by handling spent fuel differently. Ageing the spent fuel longer at the surface, removing oxide layers, and perhaps allowing gases to vent from the interior of the fuel rods would all diminish the inventory of C-14 that would have to be buried in a repository.

(3) It was stated at the ACNW workshop that one of the factors limiting the release of C-14 from the waste package is its conversion rate to carbon dioxide. Therefore, the presence of oxygen in the repository atmosphere enhances C-14 mobility. Why not flood the repository with an inert gas just prior to closure, in order to slow the conversion process?

Joseph F. Donoghue
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