Appendix B

Radioactivity in Coal and Fly Ash

by Kjell Johansen, Ph.D.*

*Dr. Johansen is a Sr. Chemist at the Point Beach Nuclear Plant in Two Rivers, WI, where he is responsible for air and water related effluent compliance issues. He received an MS in Radiological Health Physics from North Dakota State University, an MS in Environmental Health Sciences from the University of Michigan, and a PhD in Oceanography from the University of Michigan. He spent 12 years at the UM Great Lakes Research Division as a radiolimnologist measuring NORM and fallout radionuclides in Great Lakes' sediments to determine the fate and historical inputs of pollutants to the Great Lakes. While a Radiological Engineer at We Energies from 1983 - 2000, he was responsible for Point Beach's radiological effluent and environmental monitoring programs. During that time, he also served as the Radiation Safety Officer for the use of radioactive level gauges at We Energies' fossil plants.

Radioactivity in Coal and Fly Ash

A. We live in a radioactive world. The naturally occurring radioactive atoms, or radionuclides, in the earth, the air, the vegetation, and our bodies constantly irradiate us. Each second naturally occurring radioactive atoms in the earth bombard us with 15,000 photons. Photons are a form of electromagnetic radiation given off by the radioactive atoms as they transform into stable atoms. When the nuclear transformations occur in the form of emitted particles, the original atom is transformed into a different element, which also may be radioactive. These radioactive transformations or decays continue until a stable element is formed. The earth contains two main classes of natural radioactive elements: primordial and cosmogenic.

B. Primordial radionuclides have been present since the formation of the earth. Uranium and thorium, the most well-know primordial radionuclides, have no stable isotopes. (Isotopes are atoms of the same element that have the same chemical property but differ slightly in atomic weight due to the number of neutrons in the nucleus.) In contrast, normal, non-radioactive potassium has one radioactive, primordial isotope, potassium-40 or K-40. Out of every one million potassium atoms, 119 will be primordial K-40 atoms. Whereas K-40 decays directly to a stable element, uranium and thorium decay to stable lead isotopes via a series of decays that produce numerous other radioactive elements, such as radium and radon, in the process.

C. Cosmogenic radionuclides are continually being made by the cosmic ray bombardment of the earth's atmosphere. There are 22 different cosmogenic radionuclides that become incorporated into plants and other living material to varying degrees based upon their chemical properties. The most important cosmogenic radionuclides are carbon-14 (C-14), hydrogen-3 (H-3), and beryllium-7 (Be-7).

D. The common unit for the decay rate, or transformations per unit time, is the curie or Ci (named for the Polish scientist, Marie Curie). One curie equals 2.22 trillion decays (2,220,000,000,000) per minute. Not all radionuclides decay at the same rate. The more unstable the nucleus, the faster the decay rate. Two properties directly follow from the variation in decay rates. One, it takes more atoms of a low decay rate radionuclide to produce one curie than it does for a high decay rate radionuclide to produce one curie. Two, atoms with a high decay rate will disappear faster than atoms with a low decay rate. Therefore, just because there are equal curie amounts of radionuclides present does not mean that there are an equal number of atoms present

E. Inversely related to the decay rate is the atoms half-life. One half-life is the time it takes the initial number of atoms to decay to half that number. The C-14 half-life is 5760 years where as that of Be-7 is 53.3 days. The half-life of H-3 is in between these two, 12.28 years. By comparison, the half-lives of the primordial radionuclides uranium, thorium, and K-40 are the order of a billion years. One of the radionuclides formed by the decay of uranium has a half-life on the order of microseconds.

F. Based on their known cosmic ray production rates, atoms per unit area per unit time (National Council on Radiation Protection and Measurements, Report #94, p. 39. 1987) and their known decay rates, we calculate the annual number of curies of each of the major cosmogenic radionuclides produced in the air over Wisconsin (56,154 square miles) to be as follows: 11.9 Ci of C-14, 552 Ci of H-3, and 15,100 Ci of Be-7.

G. While you may remember NORM as a character from the TV sitcom "Cheers," in the field of environmental radioactivity NORM is an acronym for **N**aturally **O**ccurring **R**adioactive **M**aterial. The air, soil, water, vegetation, and even our bodies are NORM because they contain varying amounts of naturally occurring radioactive atoms. The most common NORM radionuclides are uranium, thorium, radium, potassium-40, and carbon-14. Because of the low radionuclide concentrations in NORM, the unit used to express these values is the picoCurie or pCi. A pCi is a very small number, one-trillionth of a curie. As mentioned above, a curie is 2.22 trillion disintegrations per minute. Hence, one pCi equals 2.22 disintegrations per minute.

H. The standard 70 kilogram (154 pound) adult contains the following amounts of the aforementioned radionuclides: 30 pCi of uranium, 3 pCi of thorium, 30 pCi of radium, 110,000 pCi of K-40, and 400,000 pCi of C-14 (International Commission of Radiation Protection – Publication 39 and National Council on Radiation Protection and Measurements –Report No. 94).

I. Radioactive elements enter our bodies through the food we eat and the air we breathe. C-14 and K-40 react chemically in the same manner as the stable or non-radioactive isotopes of these elements and are continually being incorporated into the plants and animals in the food chain. Because the chemical composition of our bodies is internally regulated with respect to the amount of stable carbon and potassium present, the concentrations of C-14 and K-40 are regulated as well. Uranium, thorium, and radium also enter our bodies through the food chain, but to a lesser extent as evidenced by the pCi quantities of NORM in our bodies mentioned in the preceding paragraph. Because radium is chemically similar to calcium, long-lived radium-226 (half-life = 1600 years) will build up in the skeleton. Uranium and thorium exhibit a lesser degree of build up. Because of the relative chemical inactivity of Ra, Th, and U compared to the C and K, it takes a longer time to remove the Ra, Th, and U once they are incorporated in our bodies.

J. The amount of NORM you consume each day depends upon the foods you eat. Norm has been measured in many food items. Foods high in potassium have a correspondingly higher amount of K-40. For example, a serving of dried apricots has 409 pCi of K-40; a fresh banana, 368 pCi; a glass of orange juice, 409 pCi; bran flakes, 155 pCi; a glass of skim milk, 285 pCi; a medium potato, 690 pCi; spinach, 97 pCi; substituting lite salt (potassium chloride) for 1.2 grams of common table salt, 499 pCi; and 3 oz. of chicken breast, 180 pCi. (If you know the grams of potassium in your food, multiply by 818 to get the number of pCi of K-40). Because the body's K-40 is chemically regulated along with non-radioactive potassium, K-40 will not build up in the body but vary as stable potassium varies as a function of muscle mass and age.

K. The most common mode of radium ingestion is via drinking water. As recently noted in the Journal-Sentinel, 53 Wisconsin communities will have to reduce the radium content of their drinking water because it contains more than the EPA allowable concentration of 5 pCi/liter, (about 19 pCi per gallon).

A person drinking the recommended 8 glasses of water a day would consume about 10 pCi of radium per day, of which about 30% would be absorbed into the body (International Commission on Radiation Protection, Report of Committee 2, 1963). The food highest in radium is the Brazil nut. Brazil nuts selectively concentrate calcium family elements such as barium and radium (R. L. Kathren, 1984, Radioactivity in the Environment, Harwood Academic Publishers, p. 67). This concentration process gives Brazil nuts a radium concentration of 1-7 pCi per gram or, in a comparison to water on a weight basis, 1000 – 7000 pCi per liter. All other foods contain, on average, $1/1000^{th}$ of the radium found in Brazil nuts. The US Nuclear Regulatory Commission sets the annual ingestion limit for Ra-226 at 2,000,000 pCi/yr (Title 10, Code of Federal Regulations, Part 20, Appendix B).

L. Radon, a chemically inert, radioactive gas produced by the decay of radium, is a normal constituent of air and enters the body by breathing. Radon generated by the decay of radium diffuses into the soil pore water where it can reach concentrations of 100 – 1000 pCi/liter. The pore water radon then diffuses out of the ground into the air to yield concentrations on the order 0.1 – 0.2 pCi/liter in the northern hemisphere (NCRP Report No. 94). The amount and rate of radon entering the air from the ground depends not only upon the amount of radium in the soil but also on the physical condition of the soil containing the radium. Frozen soil and snow cover slow down the transfer of radon to the air. Radon diffuses out of porous soils more quickly than out of rock or compacted soil. Meteorological conditions like wind speed and the air pressure also affect the transfer of radon from the soil to the air. Unlike the other elements, radon does not react chemically with the body and so is readily exhaled as well as inhaled. The concentration of radon in our lungs is normally in equilibrium with the concentration in the air that we breathe.

M. The energy released by radioactive elements can be measured. The amount of energy deposited in the human body from radioactive decay is called dose. As mentioned above, radionuclides enter the body through air and foods we eat. Energy deposited in our bodies from the radioactive isotopes in our bodies is called internal dose. External doses result from gamma rays emitted by terrestrial NORM sources such as the ground and building materials and from cosmic rays. Roughly 1,000,000 photons per minute are responsible for the terrestrial component of the total NORM dose. About 500,000 decays per minute in our bodies contribute to our internal NORM dose (M. Eisenbud, Environment Vol.26 (10): 6-33, 1984). This internal NORM acts as an external radiation source to people around us. Based on the amount of K-40, the standard 154-pound adult emits about 24,400 photons per minute, which contributes dose to nearby individuals.

N. The standard dose unit in the United States is the rem. Because doses from NORM are small, these doses are reported in millirem (mrem), or $1/1000^{th}$ of a rem. In the US, the average annual NORM dose is 300 mrem. The largest part of this dose, 200 mrem, comes from the radon in the air. When we say the dose is from the radon in air, this actually is shorthand for radon and the radionuclides to which the radon decays. It is the decay products that produce most of the dose because these decay products, as opposed to a noble gas, are particulates that remain in the lungs for a longer period of time. Two of these decay products, lead-210 (22.3 yr half-life) and polonium-210 (138 day half-life) contribute most of the dose. The remaining 100 mrem is divided among cosmic (30 mrem), internal (40 mrem), and terrestrial sources (30 mrem). In the case of human-to-human irradiation mentioned above, the K-40 dose from spending 8 hours a day at 1 foot from an adult emitting 24,400 photons per minute is about 0.4 mrem/yr.

O. Cosmic ray doses increase with elevation above sea level. Typical doses in Wisconsin are around 27 mrem/yr. In Denver, the mile-high city, the cosmic ray dose is 50 mrem/yr. The highest cosmic ray dose in the US, 125 mrem/year, occurs in Leadville, CO. La Paz, Bolivia has a cosmic ray dose of 202 mrem/yr. A passenger in a New York to Los Angles flight at an altitude of 39,000 feet would get 2.5 mrem for the 5-hour flight.

P. The major contributor to the annual internal dose is K-40 (18 mrem). Lesser contributions result from two radon decay products, Pb –210 and Po-210 (14 mrem), from Ra-226 (1 mrem), and from C-14 (0.1 mrem). Note that even though the human body contains 400,000 pCi of C-14, roughly four times the pCi content of K-40, the resulting dose is very much less than that from K-40. This happens because the energy emitted per decay of C-14 is much less than that per disintegration of K-40. [United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR), 1993; NCRP Report #94; Medical Effects of Ionizing Radiation, F.A. Mettler and R.D. Mosely, 1985; D.C. Kocher, Radioactive Decay Data Tables, Dept. of Energy TIC-11026, 1981]

Q. Scientists have determined the NORM terrestrial doses in many parts of the world. These doses vary depending upon the geology of the area. Regions with high amounts of uranium and thorium in the soil and bedrock also have higher radium and radon concentrations. The US average is 30 mrem. The highest US terrestrial dose is 88 mrem. The highest measured terrestrial dose, 26,000 mrem/yr, occurs in Ramsar, Iran. Other high annual terrestrial doses occur in areas of Brazil and India (3,500 mrem), China (1,000 mrem), Norway (1,050 mrem), and Italy (438 mrem). The areas in Iran, India, and Brazil are associated with high concentrations of uranium and thorium in the soil. Epidemiological studies of the people in these areas have been made to determine, what, if any, affect these high radiation dose levels have on health. To date, no radiation related health effects have been found. [UNSCEAR 1993; NCRP Report #94]

R. Consumer products also generate NORM radiation exposures. The most common and highest consumer product exposure results from cigarettes. Smoking 30 cigarettes a day for a year delivers a lung dose of 16,000 mrem/yr, which is equivalent to a whole body dose of 1,300 mrem. By comparison to cigarettes, a chest X-ray delivers 20-30 mrem to the same tissues. Masonry buildings typically contribute 13 mrem/yr to its occupants from the uranium, thorium, and K-40 in the building material. Some electrodes used for arc welding contain thorium in order to produce greater arc stability and less weld metal contamination. Using these rods on an occasional basis results in less the 1 mrem/yr, most of which is in the form of external radiation (NCRP Reports #94 & 95).

S. Carbon based fuels also are NORM. Natural gas contains 10 – 20 pCi of radon per liter. [A liter is slightly larger than a quart with 1 gallon = 3.785 liters.] As a result, cooking with natural gas produces a dose of 0.4 mrem/yr (NCRP Reports #94 & 95). Coal contains numerous radionuclides. The US

Geological Survey maintains a large database of uranium and thorium data on coal from various US coal fields. Based on more than 5000 coal samples from all the major coal regions in the US, the average U content of 1.3 parts per million (ppm) equals 0.44 pCi/g. The average thorium (3.32 ppm) concentration is 0.37 pCi/g. These concentrations are not that much different from soil: 1.0 pCi/g for uranium (range 0.12 – 3.8 pCi/g) and 0.98 pCi/g for thorium (range 0.1 – 3.4 pCi/g). Both uranium and thorium decay to stable Pb and along the way produce radioactive isotopes of uranium, thorium, radium, radon, bismuth, lead, and polonium. Ra-226 analyses of coal indicate concentrations in the range of 0.2 – 3 pCi/g [J. Tadmore, J. of Environmental Radioactivity 4(1986) 177-204]. Lignite, a low-grade coal, has slightly higher concentrations: U-238, 8.26 pCi/g; Ra-226, 9.34 pCi/g; Th-232, 0.51 pCi/g; K-40, 4.67 pCi/g [Rouni *et. al*., Sci. Total Environment 272(2001) 261-272]. In coal-fired power plants, some of the NORM is released via the stack whereas most is trapped in the resulting ash. Studies in Great Britain (K. R. Smith et. al., Radiological Impact of the UK Population of Industries Which Use or Produce Materials Containing Enhanced Levels of Naturally Occurring Radionuclides, Part I: Coal-fired Electricity Generation, National Radiation Protection Board report, NRPB-R327, 2001) and the United States (EPA, Study of Hazardous Air Pollutant Emissions from Electric Utility Steam Generating Units – Final Report to Congress, EPA-453/R-98-004a, Feb. 1998) conclude that NORM emissions from coal-fired plants do not pose a health problem. A United Nations group of experts reached a similar conclusion (UNSCEAR, 1993).

T. The NORM concentration in coal ash is higher than in the coal because most of the radionuclides stay in the ash as compared to being released to the air during the combustion process. Therefore, burning off the organic content of the coal results in about a 10-fold increase in U, Th, and Ra concentrations in the ash as compared to the coal (UNSCEAR, 1993; USGS Fact Sheet FS-163-97). Based on the concentration process, the Ra-226 concentrations in ash could be on the order of 1-30 pCi/g. Analyses of various ashes and ash products produced at WE-Energies plants in 1993 and 2003 found Ra-226 concentrations in the range of 1 – 3 pCi/g. This is comparable to the concentrations in soil (0.2 – 3 pCi/g) and within the range of 1 – 8 pCi/g found in ash from analyses of other fly ash in the US (Cement and Concrete Containing Fly Ash, Guideline for Federal Procurement, Federal Register, Vol 48 (20), January 28, 1983, Rules and Regulations; Zielinski and Budahn, Fuel Vol.77 (1998) 259-267).

U. Given that the ash may be land filled or may be used in building materials as a cement substitute, the doses resulting from these applications have been studied to determine if there is any risk. The British Nuclear Radiation Protection Board (Smith *et. al*. 2001) conducted a detailed evaluation "Radiological Impact on the UK Population of Industries Which Use or Produce Materials Containing Enhanced Levels of Naturally Occurring Radionuclides, Part I: Coal-fired Electricity Generation" (NRBP-R327) of the doses from fly ash released to the air to people living within 500 meters (547 yards) of a plant stack, to landfill workers burying fly ash, to workers manufacturing building products from fly ash, and to people living in a house built with fly ash building products. The maximum doses determined from this evaluation were 0.15 mrem/yr for the person living near the plant, 0.13 mrem/yr from releases from the ash landfill, 0.5 mrem/yr for workers manufacturing building products, and 13.5 mrem/yr to a resident of a home constructed with fly ash building materials. The latter is not that different from the 13 mrem/yr from living in a brick/masonry house mentioned earlier.

V. Based on the preceding discussion, the radioactivity levels in coal and the slightly enhanced levels in coal ash do not constitute a safety hazard. The levels of radioactivity are within the range found in other natural products. The doses resulting from using the ash in various products are comparable to doses from other human activities and from other natural sources. These doses from the radionuclides in ash are much less than the 300 mrem/yr received from normal background radiation.