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June 8, 1979

Mr. W. T. Crow  
Section Leader  
Uranium Fuel Fabrication Section  
Division of Fuel Cycle and Material Safety  
U.S. Nuclear Regulatory Commission  
Washington, D.C. 20555

Dear Mr. Crow:

Enclosed is an analysis supported by experimental data to justify that release of spent limestone for unrestricted use will have a negligible short or long term environmental impact. This is in response to Item 8 of the attachment to your letter dated January 18, 1979.

Very truly yours,

COMBUSTION ENGINEERING, INC.

H. E. Eskridge  
Supervisor, Nuclear Licensing,  
Safety and Accountability

/wg  
Enclosure

FEE EXEMPT

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JUSTIFICATION FOR RELEASE OF SPENT LIMESTONE FOR  
UNRESTRICTED USE

Release of the spent limestone for unrestricted use is to be based on the following proposed conditions:

- 1 - The spent limestone must have no detectable activity as determined by monitoring with an alpha survey instrument.
- 2 - A composite sample from 3 scrubber loads (one day's maximum generation) will be tumbled, sampled and counted in a proportional counter. The sample must have an activity level of less than 4 dpm. This procedure is described and the limit justified below.
- 3 - A sample of the spent limestone will be analyzed for parts per million total uranium on a monthly frequency. This analysis will be used as a control check on the tumbling procedure. To allow for statistical variation, the control limit will be twice the natural uranium content of limestone as received from the quarry.

An experimental program was conducted to determine potential radiological exposures that could result from inhalation of dust originating from abrasion of the surface of spent limestone rock, and to determine measurable criteria for release of the spent limestone for unrestricted use.

Airborne dust was generated by tumbling two-pound limestone charges in a tumbler resembling a small concrete mixer. Dust samples were collected on filter paper by placing a standard air sampler head in the mouth of the tumbler bowl. The filter papers were weighed before and after sampling to determine the sample weight. Dust loadings in the tumbler bowl were quite high, averaging 23 milligrams per cubic meter ( $\text{mg}/\text{m}^3$ ). After

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weighing, the samples were counted in a gas proportional counter with a window thickness of 150  $\mu\text{g}/\text{cm}^2$ , and a nominal efficiency of 30%. Counter background is typically 1 to 2 dpm.

Sampling new and spent limestone using the above technique showed no statistically significant difference in radioactivity levels:

New limestone	-	$3.1 \times 10^{-13} \mu\text{Ci}/\text{m}^2$
Spent limestone	-	$2.7 \times 10^{-13} \mu\text{Ci}/\text{m}^2$

However, the minimum detectable activity (MDA) for this sampling, based on sample volume and counter characteristics, was  $6 \times 10^{-13} \mu\text{Ci}/\text{m}^2$ . The relatively high MDA is largely due to the small volume of air having a high dust loading that can be sampled before excessive build-up occurs. A "thin" source that will minimize self-absorption in alpha counting is necessary, thus limiting sample volume.

Ambient air quality suspended particulate sampling by Union Electric Company shows concentrations ranging from 24.5 to 80.2 micrograms per cubic meter ( $\mu\text{g}/\text{m}^3$ ) at three stations in the St. Louis area. The annual average concentration ranged from 36.1 to 43.7  $\mu\text{g}/\text{m}^3$ . Using the radioactivity concentrations we observed in the above tests, an assumed dust loading of 40  $\mu\text{g}/\text{m}^3$  equates to  $4.4 \times 10^{-16} \mu\text{Ci}/\text{m}^2$ .

Since radioactivity concentrations in the air for both new and spent limestone are below MDA for the counter, it was decided to use contaminated limestone to better characterize the sampling and counting technique. Spent limestone having a contamination level of 600 dpm/100  $\text{cm}^2$ , as measured with an alpha survey instrument, was selected for tumbling. Eleven samples were collected ranging from 72.1 to 340.2 milligrams in weight. Analysis of the data from counting these samples shows the following:

- 1 - Sample activity ranged from 124 to 179 dpm, generally showing good agreement with the alpha survey instrument measurement (when adjusted for relative area).

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- 2 - Above a sample weight of 100 milligrams there is no appreciable increase in the count rate. Above this sample thickness (11 mg/cm<sup>2</sup>) only the upper layer of the sample is being counted due to self-absorption.
- 3 - Above a sample weight of 300 milligrams the count rate begins to decrease. This indicates that the activity is actually present on the spent limestone rock as surface contamination. The decrease is due to the sample becoming coated with cleaner under surface rock as the contaminated surface abrades away.

Experiments with the contaminated limestone show that the tumbling, sampling and analysis techniques are an effective way to measure surface contamination levels. By collecting a sample in the 100 to 300 milligram range a maximum count rate is obtained. Then, if an acceptable limit is set on the allowable count rate, it is not necessary to calculate a related air concentration for the actual sampling. Also, the concentration in the mouth of the tumbler is orders of magnitude greater than those projected from any conceivable use of the spent limestone rock.

Considering statistical variation in counter background and the count rates observed with new limestone, a limit of 4 dpm was set so as to have a better than 99% probability of not receiving a false indication of contamination. Using an assumed dust loading of 40  $\mu\text{g}/\text{m}^3$ , the 4 dpm limit is equivalent to  $7.2 \times 10^{-16} \mu\text{Ci}/\text{m}^2$ .

To determine if the spent limestone generated over the past few years would meet the 4 dpm limit, 20 samples were taken on a grid pattern of the spent limestone pile. These samples were tumbled and dust collected as described above. Activity ranged from 0 to 2.7 dpm, with an average of 1.0 dpm. The 1.0 dpm average is equivalent to  $1.8 \times 10^{-16} \mu\text{Ci}/\text{m}^2$ .

Continuous exposure to spent limestone dust at the 4 dpm limit, at a concentration of 40  $\mu\text{g}/\text{m}^3$ , would result in a maximum annual dose of 3.6 mr. The observed 1.0 dpm level would result in an annual dose of only 0.9 mr. As the doses are only a fraction of the proposed 25 mr annual limit, release of the spent limestone for unrestricted use is environmentally acceptable.

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