## BLUE RIDGE ENVIRONMENTAL DEFENSE LEAGUE

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December 6, 2002

Mr. Brett Caswell Division of Engineering Services Bureau of Air Quality SC DHEC 2600 Bull Street Columbia, SC 29201 CASWELBM@COLUMB31.DHEC.STATE.SC.US

Re: Air Permit No. TV-0080-0041 Savannah River Site

Dear Mr. Caswell:

I write to provide additional comments on the SRS Title V permit. Attached please find a November 22, 2002 letter from Dr. Peter Rickards, a former member of the Centers for Disease Control Advisory Panel on INEEL. He recently submitted this letter to the DOE regarding the Modern Pit Facility. I have been in contact with Dr. Rickards on the issue of radionuclide contamination, the 10 mrem/year standard, and the lack of public health protection offered by HEPA filters. I am submitting his comments to you as part of the public record in reference to the SRS Title V air permit.

The SRS Title V permit as drafted may allow uncontrolled levels of radionuclides into the atmosphere. As outlined in the attached letter, HEPA filters are an unreliable means of controlling radionuclide emissions. The HEPA filter's failures include alpha migration, re-entrainment of particles, and alpha recoil through multiple filters. In his letter Dr. Rickards explains that alpha emitters like plutonium may "creep" through four HEPA filters in sequence. He states:

"Alpha recoil" is a DOE term, for the ability of alpha emitters, like plutonium, to "creep" through 4 HEPA filters in a row! Nobody knows how much plutonium comes out of the last filter. We need to make the DOE reveal the plutonium releases for normal operations, in a lab. The DOE has known of this problem since the 1970's, but has chosen to ignore it.

The waste burned in the Consolidated Incineration Facility (CIF) at SRS includes radioactive organic liquid waste; solid wastes such as gloves, suits, and contaminated soil; scintillation solutions, tritium-contaminated oils; and aqueous waste. In addition to incinerator stack air emissions from primary and secondary combustor chambers (H-WG0002), CIF emission points include waste storage room exhaust, kiln seal hoods, and Ashcrete vents. Also, the CIF Tank Farm stack (H-WT0020-1) and Solvent Storage Tanks (607-33H) emit a range of hazardous and

radioactive pollutants. Radionuclides listed in the waste feed data sheets include alpha-emitting Americium-241, Plutonium-238, and Plutonium-239.

The pollution control device utilized in all these emission points is the HEPA filter, sometimes in combination with other devices. For example, the pollution reduction efficiency claimed by WSRC/DOE is 98.99% for the Tank Farm Stack (SRS Part 70 Operating Permit Application, Vol. X, Book 2). We question the validity of emission reduction efficiencies of HEPA pollution control devices for the CIF and for all atmospheric emission points at SRS. Without further testing of HEPA filters as outlined by Dr. Rickards, DHEC cannot assure that SRS will meet NESHAP radionuclide emissions limits. SRS may be out of compliance and DHEC cannot permit an ongoing violation. We recommend that the state not issue this permit until such assurance can be determined.

Respectfully submitted,

Louis Zeller Blue Ridge Environmental Defense League PO Box 88 Glendale Springs, NC 28629 email <u>BREDL@skybest.com</u>

SRS TV comments 6dec02

Attachment

Letter of Dr. Peter Rickards to the US Department of Energy

November 22, 2002

To the DOE,

RE: public comment on the plutonium pit facility

I needed to share some vital information on HEPA filter problems and plutonium transport in water that effects the true plutonium emissions from the proposed pit facility and the plutonium laced low level and Transuranic waste generated. I am a podiatrist in Twin Falls , Idaho. As a citizen, and as a member of the CDC advisory panel on INEEL, I have gathered some vital documents on HEPA filter problems and Pu transport problems . I hope you will address these issues. The HEPA filter issues really effects almost all nuclear projects. Please contact me for more details if desired, but here is an overview.

To get an air quality permit, the project has to show they do not expose the public to more than 10 mrem of radiation from normal operations (and my memory says that there is a 100 mrem limit to anticipated accidents). The filters are bragged to be 99.97% efficient for 0.3 micron particles, and more efficient for both smaller and larger particles. This allows them to calculate a very low rate of release, qualifying easily for a permit.

Here are the 2 main areas of filter problems, that remain unquantified. I have called for testing the filters, in lab, for these problems, at all so-called Environmental Impact scoping hearings. To date, these questions have remained unanswered.

1) Most folks know that the filters can burn, but even if the fire is contained and put out by sprinklers, that humidity can ruin the filters. The DOE's May 1999 Defense Nuclear Facilities Safety Board(DNFSB/TECH-23) had this to say, on page 2-5, " When installed fire suppression systems are activated to protect systems, structures, and components inside confinement, the moisture-laden air carried downstream to the HEPA filters can seriously degrade filter preformance-at a time when high-efficiency filter performance is crucial." All this is "despite the fact that water repellents are applied to the medium during manufacturing." This does not stop the DOE from saying that the 3 HEPA filters (not in report) are also another unquantified accident, that could be quantified truthfully in lab settings. I have a great DOE paper from an FL Horn, replicating a criticality with plutonium. On day one, the particles were between 0.1 micron down to less than 0.005 micron. Plutonium is a heavy metal, and often a wind resuspension factor of 1 per million particles is assigned in the EIS. In this FL Horn experiment, the plutonium particles were so light, that in this windless closed cell, they floated for 3 days , bouncing around on the brownian motion of the air molecules! They slowly aggregated and precipitated, but that was in this closed cell.

2) "Alpha recoil " is a DOE term, for the ability of alpha emitters, like plutonium, to "creep " through 4 HEPA filters in a row! Nobody knows how much plutonium comes out of the last filter. We need to make the DOE reveal the plutonium releases for normal operations, in a lab. The DOE has known of this problem since the 1970's, but has chosen to ignore it. I have 2 papers from DOE on this. One is from WJ McDowell, from Oak Ridge. For the 14th ERDA Air Cleaning Conference, he writes a paper called " Penetration of HEPA filters By Alpha Recoil Aerosols." He says "Tests at Oak Ridge National Laboratory have confirmed that alpha-emitting particulate matter does penetrate High-efficiency filter media, such as that used by HEPA filters...Filter retention efficiencies drastically lower than the 99.9% quoted for ordinary particulate matter were observed with Pb-212, Es-253, and Pu-238

sources, indicating that the phenomenon is common to all of these..." It seems as if the alpha particle, from the radioactive decay, literally knocks the particles loose. As it creeps through any filters that is in it's way, the DOE thinks that smaller pieces of the plutonium particles, break off the original particle, increasing the joy of downwinders.

Another DOE paper comes from Arthur H Biermann, at Lawrence Livermore, from Dec,11,1991. His paper is called, "Alpha migration through Air Filters: A Numerical simulation." He says," It is obvious from the review of the literature that evidence exists of the migration of alpha radionuclide species through high efficiency filter media." Both papers have many DOE references, and both call for quantifying the true releases, in lab experiments. The experiments are do-able, but, so far, the DOE ain't gonna do it.

I have asked for Dr Liu, at the University of Minn. to be commissioned to study these issues. He uses a "total capture" technique for downstream particle counting. This is key to true efficiency detection ,or lack of. The present laser counter can detect down to 0.1 microns. Dr Liu can go to 0.007 micron. Seems the minimal efficiency size goes down from 0.3 micron, each time particle size detection ability increases...

The FL Horn experiment I mentioned replicates a criticality, and has Pu under the electron microscope. It ranges, on day one, from 0.1 to LESS THAN 0.005 micron, a bottomless scale! The Pu particles slowly aggregate, but much was still floating for THREE DAYS on the brownian motion of the air molecules, in this closed cell experiment. We need to quantify normal and accident filtering truefully, for the first time in nuclear history, and we should use this panel to do it. The DOE Beirmann paper mentions, as a theory, that the bigger pieces of Pu, that get caught in the first filter, may break off smaller pieces via this alpha recoil. That throws another flaw in the true dose to the public during normal operations, over 30 years. This effects all nuclear facilities, past and present.

While the DOE ignores this, a recent study was conducted in the UK. Y. Yamada et al published "Re-entrainment of 239PuO2 particles captured on HEPA filter fibres." (Radiation Protection Dosimetry Vol 82 No 1, pp25-29,1999). While I will present what I think are the shortcomings of the Yamada study, they clearly acknowledge the true efficiency of Pu filtering has NOT been quantified before. However, Yamada reported two different resuspension rates. The higher, dust loaded rate was a staggering resuspension of 1 particle per hundred per hour!

Firstly, it is significant that the Yamada study on the re-entrainment of PuO2, detected a PER HOUR rate of Pu resuspension. There is not supposed to be a PER HOUR rate of resuspension, of any kind. The DOE permit applications state that 99.97% efficiency is the MINIMUM, PERIOD.

This qualifies them to claim that the 10 mrem limit to public exposure will not be exceeded. This appears to be drastically contradicted by the continual plutonium resuspension rates, especially at higher dust loading , which replicates historical use of filters left in place for decades. Note p.28 states," For example, the dispersion rate at twice dust loading was calculated to have increased by 13 times. It was confirmed that re-entrainment was strongly affected by dust loading." My main criticism is that the experiment only lasted 20 days. The paper , ironically, does site and acknowledge, the 1976 McDowell paper I love. That McDowell paper notes that regular testing missed the alpha creep because of the short duration of their testing. McDowell left his test up for one year.

The Yamada test , however, seems to have enough sensitivity to detect alpha creep, at all flows, even in this limited 20 day experiment. I question their conclusion #1, which dismisses the lower rate of re-entrainment. They conclude, " Therefore, it was concluded that plutonium particles captured on fiber filters near the front surface hardly penetrate the filter."

I believe their dismissal misses the red flags I see. In a mere 20 day experiment, it is noteworthy that ANY plutonium gained full penetration of this filter, at this low rate. As McDowell notes, a longer time frame reveals more alpha creep. This 20 day experiment is unrealistic, since no where in the DOE are HEPA filters changed every 20 days. This low rate, short run, underestimates the true, long term penetration by alpha emitters. I noted Yamada's reference 4, the Fliescher study , that supports the probable fragmentation of smaller plutonium particles, from the larger original plutonium particles. This is the Bierman paper's theory , as well.

This clearly calls for Dr Liu's ultrasmall, ultrasensitive "total capture" technique, to capture ALL sizes of particles, to be done over an extensive period of time, that replicates actual normal use. How else are we going to determine the true efficiency, of this documented alpha creep problem?

Three important points come to mind.

1)Do the other beta and gamma emitters, that are impacted on the filter, with the alpha emitters, also leave the filters undetected? Does that not require further testing?

2) Do more radioactive alpha emitters, like the Pu-238, have even higher rates of resuspension? Does this not call for more testing?

3) Since this Yamada paper confirms alpha creep, why have the DOE downstream monitors not detect any whispering of this plutonium, through the filters? The CDC swears that the monitoring proves their is no alpha creep "footprint" on the monitors, declaring their faith in the monitors. I believe the phrase, "below detectable limits", applies to the downstream monitors, and their inability to reveal the true exposure to the public, of inhalable alpha emitters.

The second issue is the recent discoveries by DOE revealing plutonium transport in water is much easier than previously believed. The standard of 100 nanocuries per gram of waste material was created in 1984. The reason given to justify the change was a calculation that the 100 nano standard would give an acceptable dose of 500 mrem from animal intrusion and resuspension This definetly ignores the water pathway. More important, it ignores the total quantity of plutonium which will be left over the local water, buried as low level waste.

These decisions are only required to try to calculate radiation doses the public, in a thousand year time frame, if it is below 100 nano/gram. Unfortunately, as mentioned, the plutonium particles, which are potentially deadly and cancer causing, if inhaled and embedded in your lungs, remain radioactive for over 240,00 years.

We have been told for years that plutonium is an actinide, that binds to clay and rocks, immobilizing the plutonium, protecting the local aquifer. These decisions by the DOE have unfortunately ignored two recent, contradictory DOE studies, that both show how easily plutonium moves with water. Understanding these important contradictions is key to protecting local water supply and public health for centuries to come.

These two separate studies actually reveal a double trouble scenario, because both the soluble forms , and the insoluble forms of plutonium can move with water. The A. B. Kersting study , was done at the Nevada Test Site(1). This study found that insoluble plutonium had migrated 1.3 km (roughly one mile) bound to clay as a colloid and was suspended and floating in this sluggish aquifer, 30 years after being introduced to the underground environment. This is a profound, and dangerous discovery, that should change our nearsightedness about plutonium over our aquifer. These plutonium colloids ranged in size from greater than one micron, down to 0.007 microns. The DOE acknowledges that inhalation of plutonium is the most dangerous pathway of human exposure.

Plutonium colloids in our aquifer would be available for inhalation from the common use of sprinkle irrigation, and even canal irrigation that later dries, allowing newly surfaced plutonium to be resuspended in the wind. The fact that these are insoluble particles of plutonium, means that each particle contains millions of plutonium atoms. That makes inhalation more dangerous because , while the single strike alpha disintegration of a single radon gas atom is dangerous, an embedded plutonium particle provides a point of perpetual radiation and alpha destruction. The Kersting paper notes the old thinking of the DOE, siting the McDowell-Boyer paper. They say , "It has been argued that plutonium introduced into the subsurface environment is relatively immobile owing to its low solubility in ground water and strong sorption onto rocks." Kersting notes there are two previous studies of field observations contradicting that premise (2, 3).

I have heard the DOE, CDC, State, and ATSDR verbally dismiss the Kersting study as "due to the bomb testing." However, Kersting addresses the issue, stating that in the 40 years of bomb testing, previous testing only found that "radionuclides were detected at a maximum of a few hundred metres from the original detonation site. "Having isolated the specific isotope ratio of the Benham bomb test debris, there is no doubt of its origin. The Kersting team concludes, "The possibility that the Pu from the Benham test site was blasted and deposited greater than 1.3 km away, in two distinct aquifers separated by 300 m vertically and 30 m horizontally seems highly unlikely." Most importantly, Kersting concludes," Pu transport models that only take into account sorption and solubility may therefore underestimate the extent to which this species is able to migrate in ground water."

The second study I will refer to, is from DOE's Los Alamos lab, by John M. Haschke (4). While Kersting showed the mobility of insoluble plutonium, Haschke revealed that Pu in our environment can change oxidation states in the presence of airborne water vapor and become very soluble in water, enhancing mobility. This discovery contradicts the present textbooks, according Dr Madic (5) , who wrote the accompanying "Perspective" , when the Haschke study was published in Science. Textbook knowledge had only found Pu02 in the environment, in oxidation states III and IV. Madic writes how this must affect how we view everything, from the new plutonium laden MOX nuclear reactors, to nuclear storage. Madic states," Until now, it was assumed that plutonium would not be very mobile in the underground geological environment because of the insolubility of Pu(IV) compounds. But Haschke et al. demonstrate that water can oxidize Pu02 into Pu02+x, in which more than 25% of the plutonium can exist as Pu(VI), an ion that is far more soluble, and thus mobile, than Pu(IV). This new property will have important implications for the long term storage of plutonium."

So when will the DOE apply this information to protect our water and our health? We need above ground, inspectable and retrievable storage for the billions of plutonium particles dumped over our water. To ignore these studies is inexcusable.

There is one more paper I will quote, from Dr Runde. I went to the Wolfgang Runde article called "The Chemical Interaction of Plutonium in the Environment." It is from a Los Alamos conference on plutonium transport. That can be referenced at <a href="http://lib-www.lanl.gov/pubs/number26.htm">http://lib-www.lanl.gov/pubs/number26.htm</a> Runde acknowledges the colloid transport was fast, and concludes, "What is clear is that transport models to date have underestimated the extent of colloidal transport on plutonium mobility." Let me put his conclusion in context, and quote Dr Runde to a fuller extent. Dr Runde, on page 408 (or 17 of 20 on the computer download) says, "We are also trying to better understand the sorption/desorption reactions of actinides with colloids and the actinides' resulting transport characteristics. This area of environmental migration received attention with the discovery of plutonium in a borehole at the Nevada Test Site (Kersting et al. 1999). The plutonium had evidently migrated 1.3 kilometers in only 30 years." Runde continues," As discussed in the article by Maureen McGraw, we now believe that colloid transport was responsible for this remarkably fast movement of plutonium through the water saturated rock. It is not clear, however, whether the transport was facilitated by intrinsic plutonium colloids or natural (clay or zeolite) colloids. What is clear is that transport models to date have underestimated the extent of colloidal transport on plutonium mobility."

The only reference to the uniqueness of bomb testing is the initial time it takes to reach plutonium exposure to water. Runde notes that the underground explosion allowed the plutonium to be left in water, while a waste repository would differ, because the "radionuclides would be isolated, at least initially, from the hydrogeologic environment.".(p490) Runde also mentions a new concern for Pu migration, and that is microbes acting as " mobile colloids. " While they may act as a barrier, they may aid transport. Runde says, "As such, they act as mobile or even self propelled colloids. (p 409, 18/20). That is another reason we should simply re-barrel the plutonium waste, instead of shallow burial. Runde concludes, " More sophisticated models are needed to account for all the potential migration paths away from an actinide source. Theoretical and experimental scientists will be challenged for years by demands of developing these models.(p 410, 19/20)

Gee, I look forward to when they finish the job.

Sincerely, Dr Peter Rickards DPM 2672 E 4000 N, Twin Falls, ID 83301

1)A.B. Kersting et al. , Lawrence Livermore National Laboratory, Nature, vol 397 Jan 7, 1999, p56-59.

2)McDowell-Boyer, Environmental Science Technol., 26, 586-595 (1992)

3)Ryan et al, Physiochem. Eng. Aspects, 107, 1-56 (1996)

4)JM Haschke et al. ,Science 287, Jan 14 2000

5)C Madic, Science 287, Jan14, 2000