

#50

January 6, 2005

**PETITION FOR SPECIAL EXPOSURE COHORT STATUS FOR
HANFORD DUPONT WORKERS**

This Petition is submitted on behalf of a class of workers consisting of all former employees of Du Pont Company working at the Hanford Nuclear Reservation **during Dupont's operations from 1943 to September 1ST, 1946**, when Dupont Operations at Hanford terminated.

This petition is based upon three basic facts:

First, there are no individual Hanford Dupont worker records in existence **because they have all been destroyed**. This is an indisputable fact, as demonstrated by the attached enclosures in Exhibit A.

Secondly, there is not enough information available to either estimate the maximum radiation dose that could have been incurred under plausible circumstance by any member of the class, or to estimate the radiation dose to members of the class more precisely than the maximum dose estimate.

Thirdly, if one had hard data, then generic exposures might suffice in limited circumstance. But generic exposures developed from extrapolated data of doubtful reliability are not scientifically defensible. Such generic exposures are really just assumptions which cannot provide estimates of individual exposure with any semblance of reliability. It is not possible to make a dose reconstruction for an individual Hanford Du Pont worker based upon **generic** exposures that are necessarily dependent on assumptions. This is demonstrated by the following references.

I.
DUPONT RECORD DESTRUCTION
(Exh. A Enclosures)

a) Title page and page 2 from the Hanford monthly report for March 1950 states:

"Shipment to the Du Pont Company of copies of essentially all classified records accumulated by the Operation's Central Files during Du Pont's tenure at Hanford was made on March 29. This material, comprising 263 packing cases, covers the period from the fall of 1943 through August 31, 1946. Only about 4 packing cases, consisting of records presenting special documentation problems, remain to be processed."

b) Letter of April 2, 1990 from DuPont legal department stating:

"The Hall of Records has informed me that the only surviving records are payroll records of employees working at the site. All other records were either destroyed or turned over to the Atomic Energy Commission in the mid-1970's."

c) Letter of March 21, 2002 from the FOIA officer for the Hanford Department of Energy states:

"It is our understanding that employment, medical, and radiation exposure records of individuals whose employment terminated during the Hanford-DuPont contracting period (1943-1946) or who left Hanford and continued their employment with DuPont at the end of the contracting period were archived with DuPont and have subsequently been destroyed. You may wish to contact the organization that maintains Du Pont's historical records to obtain more detailed information. The address is as follows: Hagley Museum and Library, Business Archives, Attention Michael Nash, P.O. Box 3690, Wilmington, Delaware 19807-0630."

d) Letter of April 2, 2002 from Hagley DuPont Museum:

"Hagley's records relating to the Hanford Site are quite

fragmentary, as they primarily consist of contract files and in-house histories. We do not have any documentation about employment, medical, and radiation exposure."

e) Letter of March 11, 2002 from Pacific NW Laboratories (which keeps all the Hanford dosimetry records) states:

"As we have discussed in the past, when DuPont left Hanford, they took all their records with them. We have had no success in obtaining these records from them."

However, at least some DuPont operational records, as distinguished from individual records which were all destroyed, have survived. Reference is made to the Weekly H.I. Reports on the 200 Area for 1-5-45 thru 2-13-46, Parker, 7-1115-DEL. These reports indicate there is a reasonable likelihood that such radiation doses as revealed in these reports would have created a **Health Endangerment** for the Hanford DuPont workers but does not show levels of exposure similarly high to those occurring during nuclear criticality accidents. See Exhibit B.

II.

WHY THE NIOSH DOSE RECONSTRUCTIONS BASED ON ORAUT TECHNICAL BASIS DOCUMENTS ARE INVALID FOR DUPONT OPERATIONS WHICH TERMINATED SEPTEMBER 1ST, 1946.

It is important to recognize the development history of the inhaled exposure in any dose reconstruction (DR) attempted for DuPont workers at Hanford. The DR work has been done by Oak Ridge Associated Universities Team (ORAUT) under contact with NIOSH.

- 1) Since there are no individual dosimetry records, the ORAUT dose reconstructions for Hanford DuPont workers have attempted to establish generic inhalation exposures based on estimates of the various radionuclides being released into the air from the separations plants.
- 2) The generic exposures used are from the ORAUT Technical Basis Documents that characterize the Hanford site exposure conditions. (See

references at the end of each DR to these technical basis documents.)

3) The ORAUT Technical Basis Document for Occupational Environmental Dose is ORAUT-TKBS-0006-4, and the ORAUT Technical Basis Document for Occupational Internal Dose is ORAUT-TKBS-0006-5. These documents primarily concern inhalation exposures and are in turn based upon work done by Dr. Till in RAC Report No. 2-CDC, Final Report - Methods for Estimating Radiation Doses from Short-Lived Gaseous Radionuclides and Radioactive Particles Released to the Atmosphere during Early Hanford Operations, Till 2002.

4) Dr. Till's work for these exposures was based upon the work done by Mr. Heeb for the Hanford Environmental Dose Reconstruction (HEDR) project, Radionuclide Releases to the Atmosphere from Hanford Operations, 1944-1972, C. M. Heeb, May 1994.

5) In other words, Heeb's release estimates were adopted and modified by Till, and Till's estimates were adopted and modified by ORAUT site characterizations, which in turn were utilized as the basis of each individual DR.

6) Consequently, it is necessary to examine the validity of each stage of development upon which the following stage was based.

III.

WHY HEEB'S GENERIC RELEASE FACTORS CANNOT BE USED

Heeb's methodology was to first calculate the amount of each radionuclide created in the reactor fuel rods which he determined from the known basic physics of nuclear fission as a function of reactor power levels and the time fuel rods were in the reactor. Then he determined the amount of activity in the fuel rods being dissolved in the separation plant processing and the estimated amounts of the nuclides being released to the environment during dissolving and subsequent processing in the separation plant. This enabled him to establish a release factor or fraction for the nuclide in question as the ratio of the radionuclide activity released to the radionuclide activity processed.

The longer-lived nuclides created in the reactor available for possible release are not greatly affected by changes in the cooling times because rate of decay is not a significant factor. However, for short-lived nuclides, most particularly iodine-131 (eight-day half-life), the cooling period from time of discharge from the reactor to time of dissolving in the separation plants is a major factor in determining the amount of activity available for entrainment and possible release.

One problem with Heeb's **iodine** calculations is the difference in cooling times to be utilized to calculate amounts available to the dissolver. Heeb changed his cooling times from his 1993 report in which he recognized a three-day lag time error overestimation, which was omitted from his 1994 report. He also used linear averaging of monthly amounts when cooling time is actually an exponential factor, also causing cooling time overestimation. Further, he failed to account for the out-of-sequence dissolving order of fuel rods first in the cooling basis to be first sent out to the dissolver, (FIFO). (See Exhibits B and C for a full discussion by widely recognized and published researchers in nuclear science and iodine-131.)

The following citations were taken from Radionuclide Releases to the Atmosphere from Hanford Operations, 1944-1972, C. M. Heeb, May 1994. Heeb was the principal author of the HEDR project source term.

"Monthly releases of ruthenium-103, ruthenium-106, cerium-144, strontium-90, and plutonium-239, plus iodine-131 releases after 1949, were estimated as follows:" (Heeb, 1994 Pg. # 4.3)

"The release factor is defined as the ratio of the radionuclide activity released to the radionuclide activity processed. Release factors are calculated from known values over a short period of time. They provide an estimate of the fraction of radionuclide released from processing operations. **This estimate is then applied to time periods during which processing data, but no release data, are available.**" (Heeb, 1994, Pg. # 4.7)

"A monthly release factor was calculated for each of the six radionuclides under study in this report (iodine-131, ruthenium-103, ruthenium-106, cerium-144, strontium-90, and plutonium-239) for each separations plant in operation. Release factors based on measurements

from a given time period were assumed to apply to all time periods when the same release control equipment was in service. (Heeb, 1994, Pg. # 4.7)

“T and B Plants. Release factors calculated for the REDOX and PUREX plants were applied to T and B plants because the emission control devices at T and B plants were similar to those in place at the REDOX and PUREX plants.” (Heeb, 1994, Pg. # 4.17) (this is factually incorrect)

Heeb’s release factor for the **iodine** before installation of water scrubbers in May 1948 was 0.905. Water scrubbers were calculated to lower the release factor to 0.285, as per estimates made in the GE monthly report for May 1949, and when sand filters were installed in December of 1948, the release factor became 0.25. (Heeb 1994, Pg. # 2.2)

IV.

PARTICLE PROBLEM

One factor ignored by Heeb was the particle problem that was recognized as existing but unreported until 1947. Although this is after DuPont terminated, there is no reason to think it was not existing in the 2 years previously, since the emission control equipment was just beginning to be installed in mid-1948.

Health Instrument Section surveyors detected contamination in the form of discrete active particles on the ground in the region of the Hanford separations plants in late September 1947, HW-9259.

“Particle problem remains despite fan replacement: The active particle contamination in the Separations Plant Areas continued to command much attention. Since the replacement of one fan in each area, the rate of particle deposition has diminished by a factor of less than two, whereas the average activity of each particle has diminished by a factor of 5. Recently used photographic methods of detection indicate about ten times as many particles as were found by meter surveys... Biological monitoring to date has been inconclusive.” HW-9191, 2/29/1948.

“There is some evidence that apparent increase of concentration with increasing distance from the stack is real.” H.M. Parker, “*Review of the*

Stack Discharge Active Particle Contamination Problem” HW-9259,
3/22/1948.

Re: Dispersion. Assigned to Church, Meteorology. Unable to answer fundamental particle dispersion problems without extensive Hanford experimentation. Parker submits: “Small particles could ascend 10,000 to 15,000 feet in summer by thermal convection, and rarely to 3,000 feet in winter by mechanical turbulence.” HW-9529, 3/22/1948.

“... there is a backlog of tens to hundreds of millions of particles unaccounted for in the Plant vicinity. The responsibilities of the Company and of the Commission require the most energetic attempts to prove that no significant radiation hazard can arise from this source, or to correct an untoward condition regardless of cost.” HW-95259, 3/22/1948.

“The deposition of active particles during the month followed an erratic pattern, with no apparent correlation with wind direction.” HW-10166-K, 5/31/1948, H.I. May Monthly Report.

Active particles picked up in Benton City and Richland. Monitoring implemented to determine **whether deposition rate was related to processing operations**. “The only correlation found was that deposition rate was affected by wind direction.” HW-10166-K, 5/31/1948.

“Radioactive Particle Investigation” R. C. Thorburn: “The health hazard from such a source is apparent.... Our investigations indicate that the particles actually were coming from the corroded fans and fan breechings.”

“The appearance and shape of individual particles varied widely. However, all particles appeared rough and granular under a microscope and at least a spot of rust-brown color was common.”

“Similarity of structure between the specks found on air sample filters and fan and duct work particles noted.”

“Radioactivity of particles varied in a range of 2.5×10^{11} ^{neg-8?} to $3.2 \mu\text{c}$ per particle of beta and from $<5 \text{ d/m}$ to $3,800 \text{ d/m}$ per particle alpha was found. The possibility of even hotter specks exists.” HW-10261, 6/11/1948.

“There appears to be a general belief that the number of detectable active particles deposited per month on the ground in the 200 West and

200 East Areas is increasing. In round figures, 100 million such particles fell in each area during June. Preliminary measurements of particle size within the exhaust system showed that **much of the total emitted activity was concentrated in particles below 2 to 3 μ diameter.** These are the potentially dangerous ones with respect to inhalation, and **none of this size-range is being currently measured in the atmosphere.** Possibly 10^{10} particles above 3 μ diameter, and 10^{13} (trillion) below this size were emitted during this month." HW-10378, 6/30/1948.

May '48 - 270 million particles deposited in East Area and 190 million in 200 West. The highest deposition rate noted was 2 to 4 p.s.f. per week in both areas. HW-10378, 6/30/1948.

Second Meeting of Stack Gas Problem Working Group - Admits that very small particles may exist that can't be detected using present equipment or methods. The number of particles discharged per month to the atmosphere from the 200 area stacks is 10^{10} in the 10 micron size and 10^{12} to 10^{13} in 2 micron size. The highest density of particle distribution on the ground is 3 to 6 per square feet. Studies indicate that a resident of Richland would have a possibility of inhaling an average of .3 particles per month and that a worker in the 200 Area 10 to 12 particles per month. Recently, however, this probability of inhalation has increased as much as 100 to 200%. "The general pattern of distribution is a function of prevailing wind direction." Bates #122393 to 122408, 8/20/1948.

H.I. September '48 report: Fall of active particles in the reservation was "incorrectly reported to total 10^{13} . This should have been 10^{11} ." Active particles were readily detectible in the atmosphere in Spokane, 130 miles distant, and even at Mullan Pass, 200 miles away and at an elevation of 6,000 feet. Report states it is difficult to see why operations should be permitted without respirators. Further, in fact, it is difficult to justify further operations, pending completion of the sand filters. HW-11226, 9/30/1948.

H.I. October '48 Monthly Report: "Initial Results on active particle deposition, following completion of sand filter in the 200-W Area (T Plant) showed a reduction of a factor of 4 only. Conclusions from this would be premature, but it is tempting to surmise that trouble may still be anticipated either from particles emitted from the stack itself, or

redistributed after initial deposition." HW-11499, 10/31/1948.

"A total of 88 frames exposed on the reservation and at Benton City and Pasco showed a deposition rate of 4.4×10^{10} particles per month. Frame studies completed in the 200 Areas indicated deposition rates of 1.4×10^9 particles per month in 200-East, and 2.2×10^9 particles per month in 200-West." HW-11499, 10/31/1948.

H.I. November '48 monthly report indicates "insignificant" reduction in the rate of deposition. Off-site air samples show high readings for particle inhalation. HW-11835, 11/30/1948.

Paas to Singlevich, "Particle Deposition Summary" - Subtitled "Deposition in 200 Areas": indicates one billion particles deposited in 200 East Area between 10/7 to 10/20/1948. Bates #2004209, 6/15/1949.

Note it was acknowledged that none of these particles below 2-3 um were being measured in the atmosphere, see HW-10378, 6/30/1948 above, and they were not included in the stack release estimates. There is **no evidence that the additional exposures created by these particle were factored into Heeb's release fractions**, although they most certainly created an increase in the inhalation hazard. Dr. Till acknowledged that particles were released from B & T plants in early operations, but concluded that **"the process could not be reliably modeled."** Till 2001 Draft, Pg. # 2-22.

V.

GENERIC RELEASE FACTORS

The following contains the nub of Heeb's methodology for the early pre-1958 releases of cerium-144, strontium-90, ruthenium-106 and plutonium-239, with the plutonium and ruthenium particles presenting the major inhalation and ingestion exposures for DuPont workers.

There was no release data available contemporaneous with the DuPont operations. To create such data, Heeb used release data from later periods to calculate a release fraction and then back extrapolated this release fraction, as adjusted for lack of any emission controls, to the

DuPont operations. Data marked by an asterisk [*] indicates it was collected after DuPont terminated operations at Hanford.

“The release factors calculated for the REDOX and PUREX plants agreed with the monthly estimates of the release factor determined from the Roberts (1958) data for those periods of stable operation when no non-current-production particulates were being released. These periods were characterized by release factors ranging around 1.0×10^{-7} . A generic ruthenium release factor of 1.0×10^{-7} was used for all four separations plants after installation of emission-control equipment in May 1948.” Heeb 1994, Pg. #4.18. [*]

“A higher generic ruthenium release factor was applied to T and B plant operations for the period before installation of water scrubbers in May 1948. During this period, no emission control equipment was in operation. The water scrubbers were **assumed** to be at least 99-percent efficient in removing ruthenium (Uebelacker 1960). Therefore, a generic ruthenium release factor of 1×10^{-5} was used for T and B plant operations before May 1948. (The release factor for pre-scrubber operation was increased two orders of magnitude to account for increased release due to the absence of off-gas scrubbers.)” Heeb, 1994, Pg. # 4.18.

“**Cerium-144 Release Factors** - The only data available on cerium releases were found in Anderson (1958b), which provides data on cerium releases from the REDOX Plant during September 1958.” Heeb 1994, Pg. # 4.23. [*]

“**Strontium-90 Release Factors** - Anderson (1958a) provides average curies per day for strontium-90 releases from vent stack effluent in the “separations areas” (specific separations plants not listed) during 1957. Using this data along with information on the number of curies of strontium-90 processed, which were determined from the material tons processed and the curie inventory per ton from ORIGEN2, a generic release factor was calculated: .005 curies/day...” Heeb, 1994 Pg. # 4.25. [*]

“**Plutonium-239 Release Factors** - Records of plutonium release measurements from all four separations plant vent stacks were made available by J. K. Soldat.^(a) Generic release factors were calculated by dividing the curies of plutonium-239 released during a given time period by

the curies of plutonium-239 processed during that period. Release data and release factor data are provided in Table 4.8. A sample release factor calculation follows the table." Heeb, 1994, Pg. # 4.27.

Note that superscript (a) references HEDR Project Document No. 10930320, "Plutonium and Uranium Releases from Hanford Separations Facilities/Raw Data," from J. K. Soldat (Battelle NW) to HEDR Project Office, October 26, 1993. However this Soldat document gives data developed in 1952 and 1957 from the T & B plants and REDOX and Purex, and would not be applicable to the T and B plants before September 1st, 1946. [*]

"T and B Plants - The T and B plant plutonium-239 release factor was based on the combined plant average monthly releases of 3.9×10^{-7} (rounded to 4.0×10^{-7}) for operations after installation of emission-control equipment in May 1948. [*] Emission-control equipment **was assumed** to have been about 99-percent efficient, so the early period plutonium-239 release factor for the T and B plants was 4.0×10^{-5} . (The release factor for pre-scrubber operations (1944-1948) was increased two orders of magnitude to account for increased releases because of the absence of off-gas scrubbers.)" Heeb, 1994, Pg. # 4.27.

Plutonium-239 Release Estimates - Figure 4.8 shows that plutonium-239 releases were largest in the early period of separations plant operations, reaching a maximum of 0.0755 curies during the month of October 1945. Releases were reduced after water-scrubber installation in May 1948. The late-period maximum was reached in January 1966, with a plutonium-239 release value of 0.006456 curies for the month." Heeb, 1994, Pg. # 4.27.

It is clear that Heeb did not have any contemporaneous data to support a release factor (RF) for any of four radionuclides being considered in the particle inhalation exposures to DuPont workers. In each case Heeb used release factors developed in other facilities and in other time periods, different from the early conditions during DuPont tenure at the T and B plants when **no emission control** devices were being utilized. For **plutonium and ruthenium**, Heeb retrospectively adjusted these release factors to reflect an **assumed 99%** removal efficiency from the later developed emission control devices. The 99% efficiency assumption is not

supported by any hard comparison data from actual conditions existing at the T and B plants during the Dupont years at Hanford. The filtering equipment was subject to continuous changes for improvement and releases factors developed at one point in time were not applicable to other time periods with different equipment. For example, when **caustic** scrubbers replaced the **water** scrubbers the capture of iodine was greatly improved.

To back extrapolate a RF from a later date back to the early period before emission control devices were being developed, it is necessary to know the effectiveness of the devices as compared with the situation of no devices. (The release factor for pre-emission control devices (1944-1948) was increased two orders of magnitude to account for increased releases because of the absence of off-gas devices.) This effectiveness difference was set by Heeb at 100 times. It is interesting to note that Till used an increase factor of 150, which he called the "effluent treatment modifier" instead of Heeb's 100 (see Till 2002, pg.2-26), which emphasizes the basic uncertainty and raw assumptions in these so-called release factors. It is not known if ORAUT adopted Heeb's 100 multiplier or Till's 150 multiplier.

This basic uncertainty has been confirmed by a letter report by A.G. Blasewitz to the EPRP (External Peer Review Panel, see attached as Exhibit D) that was organized to provide a non-public review of HEDR, in addition to the public sessions and critiques by the Technical Steering Panel (TSP). This attached review make precisely the same points made by Petitioner in the foregoing paragraph, and concludes that the RF for Ce, Ru, Sr and PU should be 4×10^{-4} instead of 4×10^{-5} concluded by Heeb. See Exhibit E, page 3. With the estimated activity being released under no emission controls (DuPont era) ranging from the 100 times to 1000 times the release factors developed later under emission controls, the Dupont generic release factors are mere assumptions, at best, and cannot support an individual Dose Reconstruction.

In further support of the Till & Blasewitz disagreement with scrubber efficiency of 99% is the August 6, 1948 scrubber testing, finding a collection efficiency under **96%** and predominately under **90%**. See attached Exhibit F, Stack Contamination, Martel, 1948, HDC-611.

Heeb's methodology (and that of Till and ORAUT) to calculate release

fractions depends upon three factors:

First: All the emission control devices in operation at the time a RF is calculated must be identified. This was not done adequately.

Second: The efficiency of each device must be known along with the efficiency of two or more of the devices in combination. This was not fully applied.

Third: The sampling measurements of releases must be reasonably accurate. This is the Achilles heel of even the best efforts to calculate RF. Before a back-extrapolation is even made to calculate an RF, it is necessary to have reasonably accurate sampling measurements of activity being released. Such sampling measurements were not available. Not the least of the problems is that the sampling filters being used, from CWS paper to the Whatman 40 to the HV-70 to measure releases, were not adequate to "see" or capture all of the sub-micron size particles being released, which accounted for most of the number of particles being released.

"The first quarter 1949 report (HW-14243) states that the CWS filter paper used in the air monitoring program had been shown to be about 3.5% efficient for gaseous radioiodine." Till, 2002, Pg. #4-10 and Pg. #4-11.

See also Exhibit G, Use of Natural Airborne Activity to Evaluate Filters for Alpha Sampling, Lindeken, 1961, pg. 44, showing HV-70 filters at **just 18% to 28%** of alpha activity absorbed depending on the thickness.

Lastly, see attached Exhibit H, Notes of Interview With Bernard Saueressig on 8/15/96, confirmed with his signature and initials. **"We did not try to catch those particles less than 2 microns."**

It is clear the sampling results upon which RF were being calculated were results of dubious reliability, considering that much of the activity was not being captured by the sampling.

VI.

TILL'S MODIFICATIONS OF HEEB

Till, 2002, created a spreadsheet based on Heeb's figures to show total monthly releases from routine operational releases of each nuclide. He also described what he termed "Unusual Release Conditions" in which particles containing various radionuclides were released from B & T plants in early operations, but concluded that **"the process could not be reliably modeled."** Till 2001 Draft, Pg. # 2-22. (bold emphasis added)

The following citations were taken from RAC Report No. 2-CDC, Final Report - Methods for Estimating Radiation Doses from Short-Lived Gaseous Radionuclides and Radioactive Particles Released to the Atmosphere during Early Hanford Operations, Till 2002:

"Heeb (1994) made estimates of radionuclide input to the fuel dissolution process and those are tabulated in his report." Till 2002, Pg. #2-24.

"Monthly listings of the inputs of ^{90}Sr , ^{103}Ru , ^{106}Ru , ^{144}Ce , and ^{239}Pu for the T Plant, B Plant, and REDOX Plant were compiled for the period December 1944- February 1961 from tables given in the report by Heeb (1994)." Till, 2002, Pg. #2-24.

"Releases from T Plant. Monthly releases were estimated throughout the period of operation of T Plant, from late 1944 to early 1956." Till, 2002, Pg. #2-27.

"Deposition density is the number of particles detected per unit area of ground surface. Section 4.1.6 contains a review of available data." Till, 2002, Pg. #3-39.

"Beta activity. Document HW-9871 presents summary tables of beta activity collected on ambient (i.e., in the outside environment) air filters between January 1946 and April 1948 inclusive. Air was sampled continuously through a filter of about 1-1/2 inches diameter at a flow rate of about $2 \text{ ft}^3 \text{ min}^{-1}$. The filters were counted directly on thin mica-window Geiger counters. The sampling frequency was weekly (HW-12677, HW-13743), and the reporting frequency was monthly. The samplers were located under a protective cupola type roof (HW-13743). At that time, corrections to the measured count rates were made for geometry, collection efficiency, and radioactive decay, with the assumption that all beta activity

came from ¹³¹I. **Data that showed residual long-lived activity after the decay of I-131 were not included in this document.**" Till, 2002, Pg. #4-10. (i.e. no data for the rest of the nuclides of interest)

"Operations began at T Plant in December 1944 and at B Plant in April 1945. We have found no routine air monitoring data for 1945." Till, 2002, Pg. #4-11.

"Alpha activity. There was a limited amount of routine monitoring of alpha activity in air by the Hanford contractor during this time period. Reporting of these data began in the second quarter of 1951." [*] Till, 2002, Pg. #4-16.

"4.1.5 Particle Counts in Air. Beginning in the fourth quarter of 1948, Hanford's routine environmental reports included data on the number of radioactive particles present in a certain volume of ambient (outside) air (HW-13743)." Till, 2002, Pg. #4-33. [*]

"Chronological Review of Survey Data. Health Instrument Section surveyors detected contamination in the form of discrete active particles on the ground in the region of the Hanford separations plants in late September 1947 (HW-9259)." Till, 2002, Pg. #4-38. [*]

"In fact, H.M. Parker (HW-9259) stated that measured deposition was 1-10% of the estimated emission of active particles, meaning that **90-99% of particles were not detected within the surveyed area.**" Till, 2002, Pg. #4-42.

"A factor is included in the spreadsheet to account for particles not detected by the survey instruments used. A factor of 10-100 increase is suggested by comparison of surveys for large active particles released in the 1940's with more sensitive methods (HW-9141; HW-10941). However, if this were done, then the dose assessment from the particles would need to reflect the inclusion of smaller particles and their activity characteristics. **We have not made any adjustments to the measured deposition densities for the example calculations included with this report.**" Till 2002, Pg. #3-39 (In other words, Till's computer file, *scrlist.dat*, which provided ORAUT 0006-4 with its source term did NOT INCLUDE Till's suggested increases 10-100 times to account for particles not detected.)

VII.

ORAUT'S MODIFICATIONS OF TILL

ORAUT's calculation of dose estimates are based upon Heeb, 1994, and Till, 2002, as modified by ORAUT. ORAUT developed an individual dose calculator utilizing RATCHET, the HEDR air dispersion sub-program.

"5.2.1 Plutonium. By far the most serious intakes at Hanford involved plutonium and ^{241}Am . Routine urinalyses for plutonium started in September 1946." Technical Basis Document for the Hanford Site – Occupational Internal Dose: ORAUT-0006-5, Pg. #13. [*]

"Air sampling was performed in facilities from the earliest days of operation. Workers routinely exposed to areas with detectable airborne concentrations or performing high risk jobs that might produce airborne contamination were on routine bioassay schedules (except **prior** to about 1948)." [*] ORAUT-0006-5, Pg. #48.

"For routine releases from T and B plants the estimated monthly release is the product of the following factors:

- The monthly processing rates for each radionuclide compiled by Heeb (1994), which are the rates at which the radionuclide enter the separation plants as irradiated fuel from the reactors. **The processing rates were estimated by Heeb (1994) for ^{90}Sr , ^{103}Ru , ^{106}Ru , ^{131}I , ^{144}Ce , and ^{239}Pu and by Till et. al. (2002) for ^{137}Cs .**
- The release fraction which is the ratio of the rate that the radionuclide was released to the processing rate of the radionuclide.
- The **"effluent treatment modifier"** which accounted for increased release rates in the early years of operation of T and B plants when there were no scrubbers or filters on the effluents." Technical Basis Document for the Hanford Site – Occupational Environmental Dose: ORAUT 0006-4, Pg. #10.

"The computer file *scrlist.dat* (Till et al. 2002) provided the initial source terms in the spreadsheet. That file was based on

the extensive research preformed by Heeb (1994). The files from Till et al. (2002) provide monthly release estimates for the period from Hanford Site startup in 1944 through 1961.” ORAUT-0006-4, Pg. #15.

“Intake of Airborne Radionuclides - The computer program RATCHET and an Excel® spreadsheet were used to calculate claimant favorable but realistic intakes from airborne radionuclides as opposed to the computer program ‘Hcalc’ (Till et al. 2002) because ‘Hcalc’ calculates “worst case” or maximum hypothetical dose as opposed to a realistic dose.” ORAUT 0006-4, Pg. #14.

“The computer program RATCHET (Ramsdell, et al. 1994) was used to calculate annual average atmospheric dispersion coefficients. The RATCHET code was run for unit releases of 1 µm particles, noble gases, and iodine speciated as a combination of elemental iodine, particle-bound iodine, and organic iodine as described in Farris, et al. (1994).” ORAUT-0006-4, Pg. #16.

(Note: no basis given for assuming RATCHET could run the non-iodine particle dispersion correctly, or that 1 µm represents the average size of the non-iodine particles. See discussion below.)

The Till spreadsheet results were adopted by ORAUT and multiplied by air dispersion coefficients developed by RATCHET to develop the individual exposure from inhalation of the nuclides. Interesting to note that Till had developed an air dispersion model for particle air transport, Till 2002, section 3.2, but for unexplained reasons ORAUT utilized RATCHET.

There’s no full explanation as to exactly how RATCHET was modified. There are many difficulties with RATCHET, as follows:

1) It’s tendency to under-estimate. The Technical Steering Panel (TSP), a group of independent academics and scientists was organized to monitor and provide critical review of the HEDR project as it was being developed by DOE contactor, Battelle NW (a.k.a. Pacific NW Laboratories). The final report of the TSP upon completion of HEDR states, however, that:

“Key Issues Remain to be Resolved”

"The TSP identified seven priority tasks to be completed before the panel could say that dose reconstruction models and data could be used with confidence in Hanford health-related work. The CDC plans these tasks to be performed by outside contractors. The seven key issues are, in order of priority:

Air model concerns. Many people remain concerned about, 'disappearance factors' - the inconsistencies between measured and predicted deposition of iodine in the 1940's - and the role weather and topography may have played in the amounts and locations of radioactive iodine deposited." See Exhibit I, Reconstructing Hanford's Past Releases of Radioactive Materials: The History of the Technical Steering Panel 1988-1995, November 1996.

It will be noted that the adequacy of RATCHET was the number one concern needing correction. Even its author, Van Ramsdell, acknowledges under-estimates generally within a factor of four, but up to a magnitude at the more distant domain locations. See Exhibit J, Atmospheric Dispersion and Deposition of 131-I Released from the Hanford Site, J.V. Ramsdell Jr. et al., Health Phys. 71(4):568-577; 1996. However, although number one of the concerns of the TSP and the CDC, RATCHET was never changed! See Dr. Till's deposition admission, Exhibit K.

2) The code for RATCHET contains a mathematical error in the plume depletion equation which doubled the rate of plume depletion. According to Arthur Rood, a member of Dr. Till's scientific team, Ramsdell admitted this error in his code and that it was never corrected in his report, but he claimed, however, that the way he ran the code the error was compensated by another factor that exists in the code. See deposition Exhibit L for full elaboration.

3) The problem with ORAUT's approach is that using a one micron size for particles ignores the fact that many plutonium particles were less than one micron, and the average sizes of the ruthenium particles has not been characterized by any studies. Also, there is no data to support deposition rates for ruthenium or plutonium which were apparently just speculated as being equivalent to the combined deposition rates of the three species of iodine-131 (elemental, aerosol particles, and organic).

Clearly, ORAUT's atmospheric dispersion factors were developed on

insufficient and inappropriate data. It is also noted that ORAUT reported no effort to validate its model. Nor was any mass balance analysis made to determine if the modified RATCHET being used had been properly coded.

The generic data relied upon for DuPont exposures is based upon a series of assumptions from one step to another. First, Heeb's estimated DuPont release factors, for which no contemporaneous data existed, which he created by **retrospectively** applying release factors developed at later times from other facilities using efficiencies **assumed** for the facility from where release factors had been taken, and then back-correcting to account for the lack of the filters at DuPont. Secondly, these Heeb estimates were utilized by Till as the basis of his estimates of DuPont exposures which in turn were converted by ORAUT into individual exposures. DuPont generic exposures are necessarily dependent on extrapolated assumptions and cannot provide estimates of individual exposure with any semblance of reliability.

VIII.

THE EFFORTS TO CREATE A DR FOR DUPONT WORKERS AT HANFORD, HOWEVER WELL INTENDED, ARE SIMPLY NOT SCIENTIFICALLY DEFENSIBLE.

Even if one were to assume, arguendo, some plausibility for such generic data, it cannot be applicable without some knowledge of the individual dosimetry, the workers activities, the locations of his work, protective gear, if any, and any episodes of higher than normal air concentrations, and incidents of accidents or special individual exposures, **for which data no longer exists**. NIOSH cannot reasonably estimate a dose in a factual vacuum. The worker could have been at any location in the 100, 200, or 300 areas at Hanford and received an excessive exposure for which the records no longer exist. Recall also, this was start-up time, the period of the largest releases and the most prone to accidents, and also failures to adequately protect workers as the hazards were being learned.

IX.

NECESSARY CONCLUSION

The facts establish that there is **not** enough information available to

either **estimate the maximum radiation dose** that could have been incurred under plausible circumstance by any member of the class, or to estimate the radiation dose to members of the class more precisely than the maximum dose estimate. However, at least some DuPont operational records, as distinguished from individual records which were all destroyed, have survived. Reference is made to the Weekly H.I. Reports on the 200 Area for 1-5-45 thru 2-13-46, Parker, 7-1115-DEL. These reports indicate there is a reasonable likelihood that such radiation doses as revealed in these reports would have created a **Health Endangerment** for the Hanford DuPont workers, but does not show levels of exposure similarly high to those occurring during nuclear criticality accidents.

Consequently, there is no basis upon which to calculate generic or individual external or internal exposures to DuPont workers at Hanford, and since all the records of Hanford DuPont workers have been destroyed, their inclusion as a Special Exposure Cohort is mandated by this lack of any reliable data. The destruction by DuPont of Hanford worker records was a shameful act for which the workers should not be penalized. To give the worker the legally mandatory benefit of doubt, unencumbered by specious assumptions, would be to establish the class of all former DuPont workers at Hanford as a Special Exposure Cohort.

Respectfully submitted by undersigned Petitioner **on behalf of all the former DuPont workers at Hanford**, as authorized in the attached Petitioner Authorization Forms signed by the following named former DuPont workers or their Survivors:

1. _____, Survivor of DuPont employee _____
2. _____, Attorney-in-fact for _____, Survivor of DuPont employee _____
3. _____, Survivor of DuPont employee _____
4. _____, Survivor of DuPont employee _____
5. _____, Survivor of DuPont employee _____
6. _____, Survivor of DuPont employee _____
7. _____, Survivor of DuPont employee _____
8. _____, former DuPont employee.
9. _____, Survivor of DuPont employee _____

Presented By:

D
I

d

Attachments:

- Exhibit A: Letters Confirming destruction of DuPont Records
- Exhibit B: Jervis/McNeill 1999 report
- Exhibit C: Jervis/McNeill 2004 report
- Exhibit D: A.G. Blasewitz to the EPRP (External Peer Review Panel)
- Exhibit E: Drawing attached to Blasewitz letter
- Exhibit F: Stack Contamination, Martel, 1948, HDC-611.
- Exhibit G: *Lindeken, 1961, pg. 44*
- Exhibit H: *Saueressig on 8/15/96*
- Exhibit I: The History of the Technical Steering Panel 1988-1995, 11/1996.
- Exhibit J: J.V. Ramsdell Jr. et al., Health Phys.
- Exhibit K: Dr. Till's deposition admission
- Exhibit L: Rood's Deposition: Excuse for the RATCHET coding error in
Exh. M
- Exhibit M: Atmospheric Tracking Code (RATCHET)
- Exhibit N: Petitioner Authorization Forms

Exhibit A

DECLASSIFIEDTECHNICAL DIVISIONS

March 1950

4/10/50

SUMMARYPile Technology Division

The major effort of the Physics Section involved planning for the loading of the central 600 tubes of the H Pile for increased P-10 production. Experimental work in the DR Pile is being planned to determine the optimum loading arrangement.

A large development program for extraction phases of the P-10 program has been initiated.

Improvements have been effected in the method of canning P-10 fuel slugs.

An improved pile start-up procedure, which has significant advantages over the conventional "reduced power" start-up procedure was demonstrated.

Precise measurements of the curvature near the inner end of the gun barrel in top central tubes of the piles show that the worst condition prevails at the F Pile, with a radius of curvature of 660 inches.

Development of equipment for insuring complete transformation of production slugs is continuing.

Separations Technology Division

The first of ca. 450 MWD/ton metal is now being processed in the Separations Plants under close observation. The causes of higher-than-normal product hold-up obtained in recent B Plant Acid Wash Runs are being investigated. Metathesis time cycle shortening by production testing has been satisfactorily obtained and variations in the Isolation Building final product solution volume are being studied via production tests in an effort to improve product accountability and transfers between Bldgs. 231 and 234. Improvements in Bldg. 234 Dry Chemistry conversion yields are being attempted. Accumulated "scow sweepings" from Dry Chemistry operations have been separately reduced to plutonium metal buttons, which are being stored for future recovery operations.

In Redox and Metal Waste Recovery process development, sixty-one additional solvent extraction column runs were made during the past month, all on TBP process studies. Packed column performance with Shell Deodorized Spray Base as the TBP diluent was poorer than that with Stoddard Solvent previously reported. Additional pulse column runs under optimum conditions have produced waste losses of 0.1 - 0.2% for both the RA and RC Columns for effective "packed" heights of only 5.4 ft. Redox pump testing has advanced satisfactorily to the point where pump specifications for Production Plant design are now being established.

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In the research laboratory, Redox studies have eliminated previous concern about possible plutonium precipitation in product streams, and additional scavenging and ozonization data have been obtained. TBP process studies have included dispersion studies in liquid-liquid extractors, acid butyl phosphates formation and properties fluoride complexing, and decontamination behavior of various TBP diluents. Process scouting for Bldg. 234-5 operations has involved studies of nitric acid recovery, leaching of slag and crucible wastes, "electroless" plating of nickel on plutonium, and coupling of Redox solutions to Bldg. 234 operations. Various potential methods of separating aluminum and U²³⁵ are being investigated as a necessary requisite to the development of a more desirable process for the recovery of "25."

In the 234-5 process development laboratory, the study of substitution of peroxide for oxalate purification has been continued. A design basis for plutonium "skull" recovery has been issued. Considerable improvement has been made in producing plutonium cores that will pass through the "Go" gage in all directions. Improvements in radiographic methods and techniques have been obtained.

Investigation of sub-normal T Plant sand filter efficiencies has revealed the filter to be saturated with water from steam leaks into the ventilation duct and previous cell flushes. Unusually large quantities of iodine have been trapped in the filter and appear to be the cause of low "apparent" filter efficiency. The steam leaks have all been corrected and the filter bed is being dried out slowly. Pilot runs on the silver reactor for iodine removal and Fiberglass particle filters are being accelerated.

Technical Services Division.

Rala Laboratory design work continued in the Analytical Section on a 6-day week basis, and fabrication was initiated on several phases of the necessary mock-up in Bldg. 101. Sample sizes and analytical methods continued under study, and techniques were proposed for the adequate evaluation of total iodine and I¹³¹ in gases to be evolved during the dissolution of Rala slugs.

The spontaneous fission counter was received from KAFI and is being installed in the T Plant control laboratory for Analytical Section use with the alpha pulse analyzer in the determination of individual plutonium isotopes in Hanford process materials.

The Analytical Section has initiated a survey of all analytical procedures presently employed in the control of the separations process, with a view to introducing improved methods wherever possible. Particular attention is being given initially to the AT assay.

Shipment to the Du Pont Company of copies of essentially all classified records accumulated by the Operation's Central Files during Du Pont's tenure at Hanford was made on March 29. This material, comprising 263 packing cases, covers the period from the fall of 1943 through August 31, 1946. Only about 4 packing cases, consisting of records presenting special documentation problems, remain to be processed.

Initial steps were taken to centralize in the Information Group the control of code designations used at Hanford. Meetings were held with representatives from Manufacturing, E.I., Technical, and Design and Construction, and a draft of an Instructions Letter on the subject is being prepared.

AB Greninger:dg

165616



LEGAL

Wilmington, Delaware 19898

April 2, 1990

Mr. Shirley Gydesen
Battelle
Pacific Northwest Laboratories
P. O. Box 999
Richland, Washington USA 99352

Dear Mr. Gydesen:

RE: HANFORD SITE - DU PONT DOCUMENTS

As we discussed today, I contacted Du Pont's "Hall of Records" concerning the existence of certain documents relating to the operation of the Hanford, Washington Site as referenced in your letter of February 5, 1990. The Hall of Records has informed me that the only surviving records are payroll records of employees working at the site. All other records were either destroyed or turned over to the Atomic Energy Commission in the mid-1970's.

My apologies for the delay in responding. Please let me know if you have additional questions.

Very truly yours,

John W. Keiter

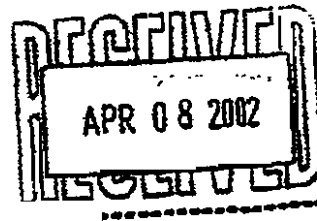
JWK:cjs

RECEIVED

APR 10 1990

S.E. GYDESEN

Hagley MUSEUM AND LIBRARY



April 2, 2002

Thank you for your letter of March 28, 2002. Hagley's records relating to the Hanford Site are quite fragmentary, as they primarily consist of contract files and in-house histories. We do not have any documentation about employment, medical, and radiation exposure.

If you have any further questions about our collections please feel free to contact me again.

Sincerely,

A handwritten signature in dark ink, appearing to read "Michael Nash". The signature is fluid and cursive, written over the typed name.

Michael Nash
Chief Curator,
Library Collections

MN/mcm



Department of Energy
Richland Operations Office
P.O. Box 550
Richland, Washington 99352

MAR 21 2002

FREEDOM OF INFORMATION ACT REQUEST (RL FOIA 2002-0114)

Your letter dated March 5, 2002, was received by this office on March 11, 2002. In that letter you requested the radiation exposure and employment records for _____ on behalf of his son. These records are protected under the Freedom of Information Act therefore we have interpreted your request under that statute.

Enclosed is a copy of _____ radiation exposure record. We have deleted the names and other personal identifiers of other individuals to protect their privacy.

According to the radiation exposure record, _____ was employed by DuPont. It is our understanding that employment, medical and radiation exposure records of individuals whose employment terminated during the Hanford-DuPont contracting period (1943-1946) or who left Hanford and continued their employment with DuPont at the end of the contracting period were archived with DuPont and have subsequently been destroyed. You may wish to contact the organization that maintains DuPont's historical records to obtain more detailed information. The address is as follows: Hagley Museum and Library, Business Archives, Attention Michael Nash, P.O. Box 3690, Wilmington, Delaware 19807-0630.


We have conducted a thorough search by name and Social Security Number (SSN) for employment records related to _____ and we were unable to locate any. Therefore, this portion of your request must be denied.

The undersigned individual is responsible for this determination. You have the right to appeal to the Office of Hearings and Appeals, as provided in 10 CFR 1004.8, for any information denied to you in this letter or regarding the adequacy of our search. Any such appeal shall be made in writing to the Director, Office of Hearings and Appeals, U.S. Department of Energy, 1000 Independence Avenue SW, Washington, D.C. 20585, and shall be filed within 30 days after receipt of this letter. Should you choose to appeal, please provide this office with a copy of your letter.

MAR 21 2002

If you have any questions regarding your request, please contact Sarah Prein, of my staff, at our address on the previous page or on (509) 376-2516.

Sincerely,


Marla Marvin, Director
Office of Intergovernmental, Public
and Institutional Affairs

IPI:SLP

Enclosure

Exhibit B

Report for the In Re Berg Litigation

Reference-Hanford Releases

March 1, 1999

by

**K. G. McNeill and R.E. Jervis
University of Toronto**

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A. Academic and Professional Experience: K.G. McNeill

A brief resume of my academic background is given here; a full CV is attached*.

I gained a First Class Honours degree in Physics from Oxford UK in 1947. My D.Phil (1950) was on the D-D (fusion) reaction. Fellowships at Yale and Glasgow UK preceded a lectureship at Glasgow, then Associate and then Full Professorships in Physics at Toronto. Honours included visiting scientist status at Melbourne, Livermore, Ottawa (in all cases more than once) Harwell and Saskatoon, and status as Professor of Medicine at Toronto and Special Staff Member at the Toronto General Hospital. During the Toronto years research has been equally divided between pure nuclear physics (photodisintegration) and nuclear physics applied to medicine (mainly in bone disease, particularly osteoporosis).

The research work has resulted in just less than 200 papers published in refereed journals. There have also been a couple of books, several published lecture notes, and several major reports on nuclear emergency planning. I have been a consultant for safety in uranium mining and for the Departments of Health and of Labour of the Government of Ontario, for the federal (Canadian) Auditor General's office, and for the Atomic Energy Control Board.

For the decade or so up to 1997 I have been the Technical Advisor to the Solicitor-General of Ontario with reference to off-site effects of accidents at nuclear power plants - this involved, amongst other things, chairing the Technical Advisory Committee and half its Working Groups, and some of its sub committees, dealing with source term estimations, meteorological modelling of distribution of radioactive emissions, dose prediction and protective actions. I chaired, for more than 10 years, the University's Radiation Protection Authority (the University has 230 professorial staff licenced to use radioisotopes).

I am a member of the Canadian Association of Physicists, of the Canadian Radiation Protection Association, and (until retirement) a Fellow of the (UK) Institution of Nuclear Engineers.

*

(see attached C.V. for details)

B. Academic and Professional Experience: R. E. JERVIS

I am presently Prof Emeritus, Nuclear Sci. & Eng., Univ. of Toronto, and a nuclear consultant**.

I gained a First Class Honours degree in Physics from Oxford UK in 1947. My D.Phil (1950) was on the D-D (fusion) reaction. Fellowships at Yale and Glasgow UK preceded a lectureship at Glasgow, then Associate and then Full Professorships in Physics at Toronto. Honours included visiting scientist status at Melbourne, Livermore, Ottawa (in all cases more than once) Harwell and Saskatoon, and status as Professor of Medicine at Toronto and Special Staff Member at the Toronto General Hospital. During the Toronto years research has been equally divided between pure nuclear physics (photodisintegration) and nuclear physics applied to medicine (mainly in bone disease, particularly osteoporosis).

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I am a member of the Canadian Association of Physicists, of the Canadian Radiation Protection Association, and (until retirement) a Fellow of the (UK) Institution of Nuclear Engineers.

**

(see attached C.V. for details)

C. Hanford Introduction

In this report we consider the validity and accuracy of the I-131 releases estimated for HEDR from the Hanford Site 200 B, T and REDOX plutonium extraction plants over their operating lifetime. While some of the HEDR calculations were based on historical records of fuel inventories and of various release measurements, a number of significant assumptions were made also and several of these were erroneous. The assumptions made were mostly conservative and led to (significant) underestimation of radioiodine releases during some of the early years of operation. Later, emission control devices and extension of fuel cooling times before processing were introduced to mitigate releases because of concerns about high emission levels of I-131 such as occurred during the years prior to 1951.

In order to evaluate the HEDR calculations we have had to review the timing of plant start-ups, fuel histories and processing records, the sequence of installation of the various emission control devices and their reputed efficiency and performance testing.

Iodine 131, a radioisotope of half-life 8 days, radioactive decay constant $\lambda = 0.0862 \text{ days}^{-1}$, is a fission product released on dissolving of irradiated U fuel. The route by which biological harm may have been caused to nearby resident and neighbouring communities by effluent iodine from the Hanford works can be broken up into different stages.

- (a) the production of the fission products in slugs and their ejection from the reactors
- (b) the "cooling time", in which I-131 partially decays
- (c) the emission of I-131 resulting from the dissolving of cooled fuel slugs
- (d) the filtering of the effluent iodine
- (e) distribution of effluent iodine
- (f) pick-up into the human system with a radiation dose being given, primarily to the thyroid
- (g) biological effect of that dose

It may first be noted that very great time and effort has been expended in the HED Reconstruction of I-131 emissions. Scoping calculations and knowledge of similar systems have been used by the present writers to show to their satisfaction that *ab initio* detailed recalculation is unnecessary even if it were possible; but this same approach showed that in particular parts of the whole reconstruction erroneous assumptions had been made, in all good faith. We concentrate our efforts in these parts of the whole in sections D and E, while consequences are discussed in G and H. D deals with "cooling times" and E with "fractional releases".

By the end of 1949, 90% or more of the total nominal emission of I-131 had taken place. In much later years the emissions were lower and so less hazardous. Nevertheless, the uncertainties in measurement and assumptions made in calculation of releases in the fifties led to the need for careful assessment, to check on biologically significant releases in the earlier part of that decade, for instance when Ag reactors were being "broken-in". However, releases after 1956 were so small that this period has been ignored.

D. Incorrect Cooling Times

D1. Introduction

Very early in our review of the Hanford problems, our attention was drawn to the incorrect, linear, averaging of cooling times from B and T plants to give a "average" cooling time which could be used to make calculations easier.

Linear averaging always and inevitably results in an appearance of a longer cooling time than is proper and therefore an underestimation of the actual I-131 in the effluent. That this is so is readily seen if, for instance, one takes equal masses of initial equal unit activity and lets one cool for 60 days and the other for 100 days. The two activities at the end of the cooling periods will be 0.0055 and .00018, total $0.0057 = 2 \exp(-0.086 \times T)$, and so $T = 68$ days where T is the effective cooling time for the two masses. For comparison, linear averaging, as used in Equation 1 of the HEDR report He92, would give $(60 + 100)/2 = 80$ days. The last part of the longer cooling period is effectively wasted, as there is very little left to decay. The 12 day difference represents for I-131 a factor of 2.8 in calculated activity.

Although the initial problem was the averaging of cooling times reported for different plants, it was quickly realized that cooling times given for each plant were themselves averages of the actual cooling times for individual dissolvings, and that these individual dissolvings were made up of slugs which may well have had different cooling times themselves. Again it is inevitable that linear averaging of these times will lead to a misleading estimate of an "average" cooling time which is always longer than the effective cooling time calculated properly, that is taking into account the exponential nature of the decay. This problem presents itself in three different ways, discussed below.

D.2 Exponential Cooling Times

For the very early days, when there were few dissolvings, it is possible to examine details and indeed for the early years HEDR does treat batches individually. [He94] (p 4.4) states that "Records were kept of the amount of uranium fuel dissolved, the burnup of the fuel, and the average cooling time (General Electric 1948a). Enough of these records survived to permit a daily reconstruction of iodine-131 from T and B Plant operations through the end of 1949. Thereafter, monthly summary records only permit a monthly reconstruction of the amount of fuel dissolved, the average burnup of the dissolved fuel, and the average cooling time of the fuel throughout the operating life of each separations plant."

For the years 1950-56 there are given the average cooling time for each month (a linear average) and, for 1953 to 1956, the minimum cooling time for any dissolving in that month. Considering first slugs from one reactor, going to one dissolving plant, as the rate of dissolving must be approximately equal to the rate of discharge of slugs from the reactors (otherwise there would be a build up of slugs or, alternatively, "days-off"); the most likely distribution of cooling times is uniform between the minimum cooling time T_{\min} for the month and a maximum of $T_{\text{average}} + (T_{\text{average}} - T_{\min})$ i.e. $T_{\text{AV}} - T_{\min} = 15$ days. This is obvious if there were 30 charge-worths of U pushed once

every month (i.e. 30 x 3.3 tons), as when each successive charge is dissolved the cooling time will have been determined to be one day longer than that of the previous one, and so for all 30. With fewer than 30 charges, some gaps may occur, days without dissolving, but this will still lead to a flattish, though irregular, distribution; it is not possible to have two charges at the same time, and therefore two identical cooling times. It means that the (linear) average cooling time is longer than it could otherwise be, the maximum cooling time is also increased and so is $T_{AV} - T_{min}$. With two reactors supplying U to the same dissolving plant, there is the possibility of two charges with the same cooling time, but still an inevitably flattish distribution of cooling times. (As will be discussed later, there may be by accident (or design) gross errors in the orderly dissolving routine - see First In First Out Protocol).

To illustrate the effect of distribution of cooling times, take a very simple distribution of 3 charges of initial activity equal to unity dissolved after cooling times of 40, 48 and 56 days respectively. The linear average cooling time is 48 days, suggesting a released activity of $3 \times (1/2^6) = 3/64 = 0.047$. However the actual release will be $(1/2^5 + 1/2^6 + 1/2^7) = 0.031 + 0.016 + 0.008 = 0.055$, 17% more than suggested by the linear average and with an effective cooling time of 46.2 d. The situation would be much worse if the difference between the minimum and maximum cooling times were about a month, 32 days. Then the cooling times for the three charges could be 32, 48 and 64 days, giving a real emission of $1/16 + 1/64 + 1/256 = 0.063 + 0.016 + 0.004 = 0.083$, 75% greater than predicted by the "average" cooling time of 48 days and giving an effective cooling time of 41.6 days.

This 75% increase is also much greater than the 17% calculated above: that is, the minimum value of cooling times is important as well as the linear average in the determination of the correct effective cooling time, T_E , (i.e. taking into account exponential decay). Furthermore, note that if there were 5 dissolvings of equal initial activity and individual cooling times of 32, 40, 48, 56 and 64 days the linear average cooling time is still 48 days, and the minimum still 32, but the real emission of $1/16 + 1/32 + 1/64 + 1/128 + 1/256 = 0.0625 + 0.03125 + 0.0156 + 0.00781 + 0.003906 = 0.121$. On the linear averaging basis, the expected dose would be $5 \times 1/64 = 0.078$. 0.121 is 55% greater than 0.078 (i.e. ratio is 1.55). Thus the number of dissolvings is also relevant. Thus with cooling times of 32, 36, 40, 44, 48, 52, 56, 60 and 64 days the linear average is still 48 and minimum 32, as before. But the real emission is now $1/16 + 1/26.6 + 1/32 + 1/45.25 + 1/64 + 1/90.5 + 1/128 + 1/181 + 1/256 = 0.0625 + 0.0442 + 0.031 + 0.0221 + 0.0156 + 0.0110 + 0.00781 + 0.0055 + 0.003906 = 0.2036$. On a linear average basis, one would expect 0.141. The correction ratio is $0.2036/0.141 = 1.45$.

For a large number ($N \gg 1$) of dissolvings the exponential average T_E may be calculated from the equation

$$T_E = T_{min} + \frac{1}{\lambda} \ln \left[\frac{2\lambda(T_{AV} - T_{min})}{1 - \exp(-2\lambda(T_{AV} - T_{min}))} \right]$$

T_E derived from this formula is slightly higher than that calculated using a finite (< 30) number of

dissolvings in a month. For instance, with $T_{AV} = 48$ and $T_{min} = 32$, the formula give $T = 44.5$ d whereas the calculation above for 17 dissolvings and the same 48 and 32 days is 44.9 days. Thus the I release calculated using this T_E is lower (by a few percent) than that calculated by the long method above. As either method is assuming, for instance, a regular dissolving (e.g. once every 2 days), small uncertainties are inevitable and acceptable.

From T_E and T_{AV} can be calculated factors to modify or correct the HEDR release; the correction is $\exp\{-0.0862(T_E - T_{AV})\}$. But for correction of T_{AV} to T_E , T_{min} is required. In the period 1950 - 52 these values are not available. If everything was so organised, it might be that in a month $\Delta T = T_{AV} - T_{min} = 14$ or 15 days; or considering new pushes every week, it is perhaps 3 days; of course it might be that in some months very old or very green fuel might make ΔT longer - and "off-days" could result in a smaller ΔT than usual or indeed in a longer ΔT than necessary. Taking as examples $\Delta T = 15$ and $\Delta T = 10$ and $\Delta T = 3$, $T_E - T_{min}$ equals 12, 8 and 1.9. The effects of these effective 3, 2 or 1 days less cooling are that 1.29, 1.19 or 1.10, i.e. 29%, 19% or 10% more I is released than predicted by T_{AV} .

Are these values of ΔT reasonable? From the above it seems so. Also in 1953 for the T plant the average monthly ΔT for the 12 months is 14.3 d, in the Redox plant 11 days, giving effects (correction factors of $\exp[-0.0862(T_E - T_{AV})]$) of 1.39 and 1.29 respectively.

Realising that the total dose is the important parameter, and that taking a reasonable average will smooth out month to month variations, it is reasonable to use a correction factor of 1.25 for the years 1950 - 52; prior to 1950, batch-by-batch calculations should have avoided the use of average cooling times and therefore its complications, and T_{min} values for 1953 - 1956 obtained from the T plant and Redox data of HW 89085 (Gydesen 1992) can be used. Correction factors for this exponential cooling time effect (ECT factor) are given in the Table 2 (column -).

D.3 First In First Out Protocol

A variation on the average cooling time problem is violation of the First In First Out (FIFO) protocol assumed by HED Reconstruction when batch-by-batch data is not available and so assumptions, such as of orderly queueing, had to be made to reconstruct the sequence of events such as cooling, dissolving, and extracting. To illustrate this, consider a series of batches of slugs which have all had different cooling times to this minute, and have activities in the ratios 1:2:4:8. If these are dissolved at 8 day intervals in the above order, the I release will be $1 + 1 + 1 + 1 = 4$ in some appropriate units. If however order is, by mistake, changed, so that the fourth one is dissolved first, the release will be $8 + 0.5 + 0.5 + 0.5 = 9.5$. Even if simply the second and third are interchanged, the release is $1 + 2 + 0.5 + 1 = 4.5$.

It is clear that a mistake, such as the one made in September 1963 [Soldat 65], will not only produce a very sharp spike of activity at and after the day of dissolving, but will also increase the total activity emitted.

In HW 83869, Aug. 31, 1964, it is stated that "From time to time a quantity of incompletely

aged irradiated fuel has been sent by mistake from the reactors to the processing area" where fission products may be released. The control systems to prevent this had a "weakness" as attested by the "occasional shipments of fresh fuel". HW 84001, Sept. 14, 1966 says that "The potential of shipping a bucket of metal that has not had sufficient decay time is a common problem ...". Thus violation of FIFO occurred from time to time. If I-131 emission were calculated on the basis of stack sample emissions the extra pulse of I from a FIFO violation would be noted in the sampling and taken into account. However if total emission be calculated from reactor power and other characteristics, including the assumption of FIFO, violation of the protocol would result in an underestimate of the actual I emission.

Ten documents [HW 32916 Aug. 56, 56029 Aug. 54, HW 33266 Sept. 54, 39260 Oct. 55, HW 58330 Nov. 58, HW 58305 Dec. 58, 2191065 May 62, HW 89067 Sept. 63, HW 83869 Aug. 64, HW 84001 Sept. 64] found and read refer to FIFO violation or its results between 1954 and 1964. In this period, the emission was usually of the order of 1 Ci per day; the reported excess emissions (as indicated by the above documents) were in each case less than 10 Ci except for HW 32916 (up to 75 Ci) HW 58305 (20 Ci), 2191065 (May 9 1962, 40 Ci) and HW 89067 (Sept 63, the "Horn Rapids" release of 60 Ci). The Dec. 1949 deliberate use of green fuel (the Green Run) which produced 7000 Ci of emission is not included above as it was deliberate.

Pre - 1954 there is need for computation. FIFO was violated pre-1950 - Jaech (FTS - CLVI-73) documents that metals from pushes in January 1946 (days 480 and 482) were dissolved in June, after metals pushed in February and in March (days 519, 533 and 538). But would the pre - 1950 batch-by-batch calculation be more likely to note and take account of FIFO violation; would the relative inexperience of operators lead to greater probability of FIFO or would have operators become more cavalier as time went on? It should be noted that the figures above (a total of ~200 Ci in an overall emission of 6000 Ci) indicate an increase of 3% in emission due to FIFO violation; and that a corresponding probability in the 1950 to 1953 period would lead to an extra $0.03 \times 40\,000 \text{ Ci} = 1180 \text{ Ci}$. And correspondingly, on the reasonable hypothesis of a linear relationship between output and dose (and effect) there would be a 3% increase in dose to all exposed.

We noted that in the period 1954 to 1964 there were remarked-on violations of FIFO. We also noted that these marked violations produced 3% increase in I output over expected. But the probability of violations resulting in individually small increases in output is much higher (with a train of 10 events, there is only one way one can transpose the beginning and the end, but 9 ways one can transpose next-door neighbours). We feel it very conservative to round-up the factor of 1.03 (3%) to 1.05, and this 5% figure is used henceforward.

Considering all the uncertainties, it may fairly be said that there is indeed evidence of FIFO violation, even until 1964, and that there is little reason to believe violation was less probable pre - 1954, and that therefore the HEDR estimations of emission may reasonably be thought to be a few percent underestimated for the period 1950 - 53. For the period 1944 to May 46, there was apparently [He(92) p 2.4] no knowledge of the push data for a particular batch and so, to reconstruct the operations, again an orderly queuing was assumed. As there may well have been violations of FIFO (which in that high output time for lack of experience would have gone unnoticed), the factor

1.05 (5%) is applied to this period as well as to the "no log book period" May to August 1946. After August 1946 the push date was known for a particular batch until the end of 1949, so no general FIFO correction should need to be made.

Summary for FIFO Factors (repeated in Table 2, column 3)

FIFO correction Dec. 44 - Aug. 46 of 1.05

No FIFO correction Sept. 46 - Dec. 49

FIFO correction Jan. 50 - Dec. 56 of 1.05

D.4 Lag Time

Another source of incorrect cooling time is the fact that although the major evolution of iodine is at the dissolving of the U slugs, and so the time that primarily matters is that between reactor shut down and dissolution, the time which is given (in the Metal Histories, for instance) is the time of extraction, not of dissolving. On the basis of early (1945) data, HEDR concluded (and used in He92) that the lag between dissolving and extraction was (around) 3 days. This is for instance indicated by the Figure 2.1 of [He92], where cuts are in storage for several days. Then the cooling time is taken by [He92] as time to extraction less 3 days (or some variation around 3, see [He92] page 4.23, where the mode is taken as 3 and variability 0-7 days). This would be reasonable if the lag time were always 3 days. Even within the bounds of [He92], however, this lag time is shown to be incorrect.

For Nov. 47 the Metal History [FTS 71] for the T plant gives push dates and extraction dates which (linear) average to give 96 days "cooling time". This is the value which Gy92 gives but He(94) gives 94 days in Table A.1. This looks as if something like the 3 days has been removed (and so it looks as if He94 buys into He92 in this respect). For October 47 there is the same story. We find the average time push to extraction is 87 days, [Gy92] gives 87 days and He(94) gives 85 days. However this is perhaps just fortuitous. Looking over the whole of the comparison between Gy(92) and He(94) for the period 1944-47, for the T plant, there is almost a random difference between the two, with differences ranging from 7 days (March 46) to -4 (June 45) and (neglecting the egregious Jan. 46 (32 day difference)) an average of 1 day longer for Gy92 than for He94. Thus the evidence that He(94) has adopted the 3 day lag seems weak to non-existent.

For Dec. 47, however, in FTS 71 two lots of numbers (of different physical size - the bigger ones are here called the "main" numbers, the smaller the "subsidiary") - are both in the "cooling time" column, a column which is under the general heading of "extraction". Again push dates and extraction dates are given. Doing the arithmetic of averaging the time of push to time of extraction averages for Dec. 47 to 96 d. for the subsidiary numbers. Gy92 also gives 96 d. (He94 gives, inexplicably, 99 days - did he add 3 days?) The main numbers however average 89 days. This must be interpreted as a noting down of both the time to extraction and the real cooling time, that is the days to dissolving, and it follows that the lag is 7 days, not 3 days. Taking a few other worked examples, for Jan. 48 plant B the subsidiary numbers give 100 days (Gy92 gives 99) and the main numbers 91.6, a lag of 8.5 d. For August 49B the numbers are 102 (Gy 92 - 102) and 93 days, lag

9 days. March 53 has 88 days (Gy-88), and 81, so lag of 7 d. The average lag time on the data is 8 days. Wider inspection of the data indicates that 8 is not far from the truth at all post 1947 times (to 1953). We might note here that, post-1947, Gy92 and He94 agree.

D.4.1 1944 - 1947

If there be evidence of a lag time of 3 days in 1945, the only other datum in this period, the Metal History for Dec. 47, gives 7 days in 1947 (a figure bolstered by later data). Reasonably one would assume a value of 5 for 1946. The use by HEDR of 3 d as a lag time, instead of 5 or 7, results in the use of longer than correct cooling times, and so an underestimation of activity of $\exp(-0.0862 \times 2)$ or $\exp(-0.0862 \times 4)$ i.e. 1.19 or 1.41. That is, the real I release has an increase of 19% or 41% respectively over HEDR for 1946 and 1947, (modified slightly as shown below).

In [He92] it is asserted that lag time need only be used from Aug. 46 to Dec. 47 ("as the Metal History Reports regrettably do not give the time of dissolving but do give the time of extraction".) For the blank period May 22 1946 to August 46 the reconstruction was so total that dissolving dates were as readily estimated as extraction dates.

Taking all this into account, no lag correction factor is used until the end of August 1946. For the remainder of that year 1.19 is used, and for 1947 a factor of 1.41 (modified slightly as discussed below).

D.4.2 1948 - 56

We use a lag time of 8 days for this period based on Metal Histories. This lag time of (fortuitously) one half-life produces a factor of 2.0 correction to the estimated I release in this period. We considered whether 3 of these 8 days had already been incorporated by He94, but came to the conclusion that it had not, based on

- (a) the production in He94 of a new Table (A.1) with a specific column for Cooling Time which in this period agrees very well indeed with the Cooling Time of Gy92 which itself gives push date to extraction date.
- (b) the lack of any clear indication that in this Table He94 had attempted to incorporate the lag time dealt with extensively in He92 and
- (c) the lack of any reference to lag time in He94.

We therefore use a factor of 2 (modified slightly); if it can be shown that a 3 day lag has already been incorporated, the extra factor is 1.54 rather than 2.0.

D.4.3 Modifications

the former using grasses and brush on a routine basis over a network of such locations, creating a large body of historical biomonitoring data.

To a lesser extent, measurements of selected nuclides including radioiodine were attempted in the 200 plants' exhaust stacks although few reliable results are available as part of the historical record of iodine releases. Monitoring of I-131 presented a number of serious problems: stack measurements were complicated by little-known differences in chemical state of iodine as it was released from fuel dissolvers and subsequent Pu extraction processes; air flows in the stacks were very great and difficult to sample representatively and bulk chemicals and other long-lived nuclides were also present. The I-131 released from fuel and dissolver solutions was mainly divided among the species: volatile iodine, inorganic iodide, iodate, Cl-I and organic-I, and the filters and liquid scrubbing devices deployed both for entrapment and stack monitoring had varying efficiencies to retain the airborne I-131 and associated aerosol droplets and particulate forms [Ma88]. Organic iodides in particular were not appreciably sorbed by water scrubbers or fibre filters. Records indicate that relatively little attention was paid to this problem of the chemical states of radioiodine other than to recognize that some fraction was particulate and aerosol spray that needed filtration; a few stack tests were made with hydrazine scrubbers which were thought to be capable of distinguishing organic-I, although this was not standard practice, then or since.

Direct counting of stack-monitor filters and of pellets of vegetation using beta counters was the principal means of detecting I-131 during the first five years [Ma89] but this procedure led to very large underestimates of iodine emissions mainly because of detector calibration uncertainties and 60-70% beta self-absorption of low energy I-131, complicated also by some interference from long-lived nuclides such as Ru. Correction factors of 3.3 and higher were required for radioiodine measurements before 1948 when a chemical extraction method was introduced but even then, iodine extractability and radiochemical yields were variable and uncertain and it was not until a non-destructive gamma-spectrometric method based on the 364 keV γ -line was adopted in 1958 that it was found that the prior methods, even after earlier correction factors were applied, had been yielding low results by a further factor of 1.6 [Ke94]. Documents show that many filters and monitor samples were measured by direct β -counting from 1948-58 because the chemical-extraction procedure was time-intensive. In addition to errors from counting, later tests showed that stack monitoring through long narrow sampling lines caused low I-131 levels to be found in the monitor filters and bubblers by as much as 50-70% (in addition to the 60-70% losses due to beta self-absorption from direct β -counting of filters.)

These observations about the difficulties encountered in monitoring I-131 emissions up until 1958 are highly relevant to HEDR estimates of I-131 releases from the B, T and REDOX plants after 1948 because attempts to measure the iodine removal efficiencies of the gas scrubbers, sand and fibre filters and silver reactors also depended on the accuracy of radiochemical assay procedures. During the first 3-4 years of Pu extraction at B and T plants when about half of the total fuel extracted there had been processed, most (~90%) of radioiodine escaped from the dissolvers and subsequent processing cells without any removal or containment devices installed. Subsequently, dissolver gas scrubbers('48), sand beds('48) and glass fibre filters('50) progressively reduced I-131 emissions to about 25% until the end of 1951 when columns containing layers of silver nitrate backed up by filters

The factors above have to be further, though only slightly, modified because although most of the emissable iodine comes out on dissolving, a fraction is emitted at extraction. He92 says that 86% of all I comes out at dissolving and after the lag (He92 used 3 days) 7.5% more is emitted - or $86 + 9.7\% = 95.7\%$ is available for emission, of which 86% comes out first, followed by the decayed 9.7%, that is, 7.5%, giving a total of 93.5%. The difference between 95.7% and 93.5% is equivalent to an effective cooling of all the I by 0.27 days. If the lag time were only 2 days, the effective cooling time would be increased by 0.19 days, whereas for 4 days, by 0.36 days and for 8 days, by 0.61 days

In the worst case, the last, we are saying that if the extraction time is, say, 96 days, and the lag time 8 days, most of the I is only cooled 88 days, not 96 days, giving an apparent correction factor of 2.0; however, taking into account hold-up of some of the I, a better cooling time is 88.61 days, giving a correction factor of 1.9 rather than 2.

For conservatism, we use these slightly corrected factors in the Results, as given in the following Table 1 and reproduced in Table 2 column 5.

Table 1 - Summary

Date	Lag time correction factor
1945 to Aug. 46	1.0
1946(Sept. to Dec.)	1.17
1947	1.37
1948-1956	1.9

E. Radiodine Releases During Fuel Processing:

From the start of U fuel processing at Hanford, very large, uncontrolled, radionuclide emissions occurred through the 200-site stacks, notably inert gas fission products (f.p.'s) and volatile forms of radioiodine. Lesser quantities of other f.p.'s (Ru, Cs), and of uranium and plutonium were also released.

Recognizing the potential health and environmental impacts of massive releases of radioactivity in the Hanford region, attempts were made to monitor such stack losses and to undertake environmental monitoring to assess whether significant levels of radionuclides were depositing over downwind areas both on-site and off-site, that is, outside the boundaries of the Hanford reservation. Vegetation, soil and water monitoring for I-131 and alpha nuclides was done,

reduced losses further, about tenfold according to various estimates [He92]. Iodine emissions had also been mitigated to some extent in the interval by increases in fuel cooling times from 30-40 days to 90-100 as detailed above in Section D. However, by that time, computations from fuel content indicate that more than 700,000 Ci. had been vented through the Hanford 200 site stacks.

That HEDR [He92, He94] neglected most of the historic data on I-131 stack release monitoring results and based most estimates of releases on computations of fuel content and on 'generic release factors (R.F.'s)' was presumably due to the considered unreliability of stack measurements (with implicit sources of error, detailed above). Also, because of the variability of early B and T plant emission measurements even after Ag reactors were commissioned, Heeb [He94] evaluated R.F.'s from a set of 1959 measurements at REDOX and applied them to 1951-56 and 1951-52 operations at the more primitive T and B plants, respectively. The continuous, on-line I-131 stack monitors were not independently calibrated and considered only as a qualitative indication of emission levels but did serve to alert operators about abnormal release events which occurred not infrequently and were related to breakdowns of the Ag reactors and filters [Bu91]. The inaccuracies of measured iodine removal efficiencies of the control devices, and the validity of assuming that 1959 release estimates from REDOX could be applied for calculating 1951-56 releases from the older plants, are further considered below.

In May, 1948, water scrubbers were installed over the fuel dissolver tanks of the B and T plants to treat the "off-gases" and in Nov/Dec '48, dry sand beds were installed to 'filter' an estimated 7.5% of I-131 escaping into the plants' 'canyon' ventilation system, mainly to trap particulate iodine with varying efficiencies, depending on the extent of condensation occurring. Deep bed fibre glass filters were later installed in Oct. (B plant) and Dec '50 (T) also in an attempt to reduce particulates and sprays. Finally, because emissions were estimated still to be over 25%, the filter beds were deployed in series with silver nitrate-coated reactor columns in Nov, 1950 (T plant) and Jan, 1951 (B). Radioiodine releases from each plant were estimated for HEDR by Heeb [He92, He94] on the basis of various measurements of the filtering/trapping efficiencies of the control devices used at different times. Heeb provides a summary of the 'best estimate' release factors:

$$R. F. = (\text{radioiodine released from plant}) / (\text{calc. fuel radioiodine content})$$

Release factors for the period 1944 to May, 1948 were based on a mean estimate of 86% I-131 released, with sparging, from dissolver gases, with a further 5-10% lost during subsequent processing steps and approximately 3% contained in stack condensate, or

$$R. F. = 0.86 + (5 + 10)/2 - 0.03 = 0.90_5.$$

Tests of the removal fraction of the 1948 water scrubbers yielded highly variable results which can be explained on the basis of the chemical form(s) expected in the oxidizing conditions of the dissolvers: estimates would be approx. 85% inorganic, 15% organic with a fraction of each contained in off-gas droplets and particulates [Ev89]. Water scrubbers are efficient for trapping the latter, less so for iodine and inefficient (< 20%) for organic iodides. Heeb [He92] reported that about 75% of I-131 was removed by the scrubbers, or,

$$\text{R.F.} = 0.86 (1-0.75) = 0.21, \text{ plus canyon air content } (0.075) = 0.28,$$

Efficiency tests on sand filter beds installed on B and T plant 'canyon' ventilation air, Nov/Dec. '48, also yielded widely different removal efficiencies ranging from 27-68% (depending on whether they were wet or dry). Considering that separate tests had shown that dry sand beds removed only small fractions of inorganic iodine, no organic-I and mainly particulates, it is our opinion that the lower figure is more realistic (whereas Heeb used a broad average value). Therefore, the appropriate, effective R.F. when both sand beds and scrubbers were operating (1948-51), was:

$$\text{R.F.} = 0.21 + (0.73 \times 0.075) = 0.26.$$

After their installation in 1951/52, the silver reactors considerably reduced emissions below the 26% levels but their performance was found highly variable. During early months, Paas and Soldat [Pa51] estimated 'ideal' R.F. = 0.01, but measured values (which may also have been appreciably underestimated because of the inaccuracy of release monitoring before 1958, cited above) increased more than tenfold within 6 months when 13% releases were reported. Rather than using these limited and inaccurate release data after initial commissioning, HEDR [He94] chose to base B and T plant silver reactor plus fibre filter efficiencies from 1952, on a documented set of 158 measurements from the newer REDOX plant. Heeb[He94] derived a median R.F. value of 0.012, from a range of 0.002 to 0.09 results. We contend that, based on a linear dose/response model for estimating biological effects, the appropriate estimate to use is the average or mean releases, namely R.F. = 0.022, a result yielding 75% higher radioiodine releases for the rest of the plants' operations until the 1956 last shutdown of T plant, compared to the HEDR calculated release estimates [He94].

Even these increased release figures are undoubtedly conservative underestimates, not only because of the inaccuracy of I-131 release monitoring before 1958, but also because the efficiencies of these control devices decreased particularly at breakthroughs when the Ag reactors became exhausted, were fouled with excess chlorine that acted to replace already adsorbed iodide, and when release episodes occurred such as the 'Green Run' and the accidental 72 Curie release from PUREX in 1963 [He94]. Also, there were frequent incidents reported when breaks, tears and corrosion of filters permitted excess fission products and actinides to by-pass stack filters and be released.

Considering (a) the different chemical forms of radioiodine, (b) differential removal of them by the several control devices and (c) deposition and condensation occurring in the ventilation and release stack ducting, the released I-131 would have been depleted in reactive I₂ and particulates that had a greater tendency to deposit locally, and result in enrichment of the volatile, less reactive, organic iodides which were not appreciably adsorbed on vegetation and remained airborne for periods of >200 h [Bu91], dispersing in a downwind plume to much greater distances off-site. Recently published research on the behaviour of fission product I-131 in releases from irradiated U and the relative absorbability of elemental I₂, iodides, iodates, IOH and organic-I on equipment surfaces including small diameter lengthy stack sampling lines, filters, charcoal, HEPA and various liquid chemical scrubbers [Ev94] provided quantitative information on the fate of 'carrier-free' radioiodine nuclides in these various chemical states. Based on information such as this, it can be estimated that the bulk of I-131 released from the first stage dissolvers had an approximate composition of 70%

inorganic, 20% organic and 10% aerosol and particulate iodide. After penetrating the control devices and ventilation, material released from the stack was probably altered in composition to 50-60% inorganic, 30-45% organic and 0-5% particulate. Owing to the reactivity and short lifetime of iodine in the atmosphere, dry and wet deposition processes occurring both on-site and off-site, the composition distribution among these iodine species would have been changing continuously with distance in a downwind plume, enriching the volatile organic-iodides.

Summary of Corrected I-131 Release Factors - reproduced in Table 2 column 7

Time period	HEDR	Corrected R.F.
Startup to April, 1948	0.90,	0.90,
May, 1948 to Oct, 1948	0.28,	0.28,
Nov, 1948 to Oct, 1950	0.25	0.25
Nov, 1950 to July, 1951	0.01-0.13	0.01-0.13
after Aug, 1951	0.012,	0.022

Typo-Corrected by 2004 Report

F. Summary of Corrected Emissions

We believe that the estimates of HEDR may be taken as a basis of I-131 source term, but that significant corrections need be made. In this paper conservative estimates of the corrections have been discussed. In Table 2 are given the appropriate correction factors due to cooling times and release factors, and overall correction factor (the product of the individual factors) and both the HEDR emissions and the corrected (HEDR × overall correction factors) emissions on a monthly (1944 - 1953) or yearly (1954 - 1956) basis.

G. Thyroid Dose Estimations

G.1 Doses within 5 years

Previously the source term for emission of I-131 from Hanford in the years 1944 to 1956 has been estimated based on modifications of the HEDR estimate. To obtain doses to individuals from this information requires knowledge of distribution of radioactivity, probability of uptake in animals and man, and distribution and metabolism in the body, as well as biological and nuclear parameters.

These (intermediate) steps have been taken by HEDR and others building on HEDR. Accepting the general correctness of these steps, (after ourselves doing scoping calculations) and assuming that general wind patterns and weather conditions and feeding patterns do not change markedly from five year period to neighbouring five year period, we can calculate the estimated dose as a function of site resulting from our corrected source term by a linear modification of the HEDR - based doses calculated by Shipler et al. (1996) and Farris et al. (1996), substituting the present emission estimates for those of HEDR.

To explain the steps taken here, we have to note that the radiosensitivity of thyroid tissue depends on age of the individual - young children are more susceptible than adults. We also note the concept of "doubling dose", the radiation dose level (at which, and) above which it is more likely than not that an existing thyroid cancer would have been caused by radiation rather than by any other normal source. While we do not necessarily subscribe to the particular dose criteria or Doubling Dose concept previously adopted by the Court, even under such standards and concept, thyroid cancer causation could be expected throughout much of the study domain. Judge Macdonald has taken that the relevant doubling dose is, for children of up to 0 - 4 years old at the time of exposure, five rads to the thyroid. For children 5 years to 9 years, the doubling dose is 10 rads, for those 10 - 19 33 rads, and for adults 100 rads.

Concentrating initially on the youngest children, we have calculated moving (or rolling) summations of source term emitted in 5 year periods starting 1940 to 1944, finishing 1952 - 1956. (See Tables 3 and 4) We have also tabulated (Table 5) these 5 year rolling totals divided by the 728 746 Ci estimated by HEDR to have been emitted in the 7 years + 1 month between Dec. 44 and Dec. 51 to aid (see below) in depicting dose distributions to persons.

We have also noted that for children born 26 December 1944 Farris depicts distributions of doses given from Dec 44 to Dec 51 (7 year plus 1 month, or more exactly 7 years plus 6 days). The HEDR emission in that period is 728 746 Ci. So we have here from Farris a relation between emission and dose to a child at a particular site. Use of the corrected emission over the 5 years after birth gives a corrected dose for that site to a 0 - 4 year old at that place.

As noted, Farris has given a drawing of dose distribution, with iso-dose contours. For this drawing to illustrate the corrected doses, the contours may be re-designated; thus a Farris 100 rad

contour may be now designated the 27.3 rad contour, if the emission in a particular 5 year period is 27.3% of the 7 year plus 1 month base HEDR emission of 728 746 Ci. This is discussed further in G.3.

G.2 Years outside the initial 5 years of life

If E be the normally expected incidence of a disease, and O the observed incidence (the increase being due to radiation) the Relative Risk RR is O/E. The dose which makes $O = 2E$ is the Doubling Dose DD. In general, if the dose be $\Sigma(0-4)$, the dose received in the first 5 years of life

the O/E will be given by $1 + \frac{\Sigma(0-4)}{DD(4)}$ where DD(4) is the doubling dose for this age group. If $\frac{\Sigma(0-4)}{DD(4)}$ is > 1 , it is more likely than not that radiation has caused the observed disease.

But now consider that dose after the age of 4 may add to the probability of causing the disease, though this, rad for rad, is less likely. If $\Sigma(5-9)$ were the dose for the next 5 years, and if at this age the DD is DD(9), the disease would more likely then not be caused by radiation if $\frac{\Sigma(5-9)}{DD(9)} + \frac{\Sigma(0-4)}{DD(4)}$ is > 1 . Similarly for older ages, with DD(4) = 5 rads, DD(9) = 10 rads, DD(19) = 33 rads and DD(>19) = 100 rads.

Then a child born in Jan. 1945, living at such a distance from Hanford that he/she does not receive 5 rad in 1945-1949, may move in 1950 or 1951 much closer to the site of emission and pick up additional rads in the next 5 years. Or a child born in 1941 may well have a sub-critical dose up to and including 1945 but still have an above critical dose for the first 10 years of life.

For instance, if a person born in 1947 received 4.5 rads in the first 5 years of life (to Dec. 31 1945) he/she could well, if living in the same place, receive $(0.25/0.80) \times 4.5 = 1.4$ rads in the next 5 years. The relevant probability figures are $\frac{4.5}{5} + \frac{1.4}{10} = 0.9 + 0.14 = 1.04$, i.e. the total excess relative risk is greater than unity, so if the person has thyroid cancer, it is more likely than not that the radiation dose caused the cancer.

These cases will have to be looked at case-by-case.

G.3 The Distribution of Dose

As noted above, Shipler et al (1996) and Farris et al (1996) have used computer codes (Ratchet and Descartes) to move from source term to dose as a function of position vis-à-vis Hanford works. In particular, they have estimated and depicted in a series of drawings the dose received by a target group (infants) as a result of 728 746 Ci of I-131 released. (Note these doses are median doses with large errors on them, as indicated in Table 1 of Farris's paper).

In the drawings given here which are adapted from Farris, we depict the doses generated by I emission proper for different 5 year periods; this has been done by modifying the isodose contours of Farris by multiplying the dose represented by a contour by the factor M, where M is the factor given in Table 5, the emission in Ci in a particular 5 years according to our estimates (Table 4) divided by 728 746 Ci.

Let us consider our Figure 1, for the first 5 years of life of infants born 1/1/41 and consuming "back-yard" produce. This is based on Fig. 6 of Farris which is for infants eating "back-yard" food. Our Table 4 shows that in the period 1/1/41 to Dec 1945 the I emitted is 585 090 Ci, which is 0.803 of the HEDR 728 746 Ci. So the isodose contour values must be changed by a factor of 0.803. For the 4 highest dose regions shown in Farris Fig. 6, the boundaries are given as 230 rad to 100 rads, 100 to 32, 32 to 10 and 10 to 3 rads. Our modifications to make the depiction applicable to the 5 years 41-45, are to change the region boundaries to 184 rads to 80 r, 80 r to 26 r, 26 r to 8 r, and 8 r to 2.6 rads. Further, we have sketched in the probable position of the (median) 5 rad isodose contour, because of the Doubling Dose being 5 rad for infants. It is seen that Richland, Spokane, Walla Walla and Pendleton are all within this contour, i.e. a child born in 1941 in any of these places, and living there for 5 years or more would likely receive more than 5 rads (and on the doubling dose picture would, if they later developed thyroid cancer, be more likely than not to have had the cancer caused by radiation from the I emissions).

Fig. 2 is for infants born on or about Dec. 26 1944. For the 5 years (plus a few days) to December 1949 a resident infant would receive (Table 5) a dose 103% of the 728 746 Ci on which the Farris drawing is based. So here we multiply the Farris contours by 1.03 to give boundaries of 238 r to 103 r, 103 r to 33 r, 33 r to 10 r and 10 r to 3 rads. Again the 5 rad contour is sketched in.

Furthermore, as the 5 year rolling summations are roughly the same from 1942-1946 to 1945-1949, this figure 2 shows, within 10%, the 5-year doses in this extended period.

We also show Figure 3 (1946-1950) [and also useful for 1947-1951] and Figure 4 (1951-1955) which also roughly (within 20%) portrays 1948-1952, 1949-1953, and 1950-1954. Beyond 1955 the doses fall off and are not illustrated.

For other circumstances, e.g. for adults, Farris Figure 7 could be correspondingly modified. For infants consuming "commercial" food and milk Farris Figure 8 applies; our Figure 5 indicate doses for infants born in 1942 to 1945. The general picture is the same as for our Figure 2.

H. Opinions and Conclusions

Although the HED Reconstruction has been carried out with diligence and overall care, there are a number of incorrect assumptions made or points neglected which make necessary correction of the I-131 emissions from Hanford. None vary greatly the massive output in 1945, but they do strongly alter calculations of later emission estimates.

In particular we note

- (a) incorrect averaging of cooling times, with correction factors ranging from unity to 1.67
- (b) violation of First In First Out Protocol, with correction factors of from 1 to 1.05
- (c) lag time (time between dissolving and extraction) correction factors ranging from 1 to 1.9
- (d) stack monitoring of I-131 being vented to the atmosphere was inaccurate and unreliable because of poor design of the long, narrow stack air sampling lines, poor calibration and variable collection efficiencies of the bubblers and filters connected to the lines, and improper radioassay procedures. The 'generic release factors' that HEDR applied to calculate fractions of iodine released were not well known from 1948 to 1956 and the few measured values were subject to the same errors of stack monitoring. The use of an R.F. estimated from 1959 REDOX plant performance as a basis for calculating 1951-56 losses from the older B and T plants is highly questionable, and in addition mean, not median, values are appropriate. The release correction factors range for 1.0 to 1.76

The overall effect is such that in one month (Jan 1953) the correction factor is 6.28, that is HEDR underestimated the emission by a factor of more than six.

Using the corrected emissions and the relationship between I emission and distribution of dose in the proximity of Hanford developed by Shieler et al (1996) and Farris et al (1996), depictions of dose received by e.g. infants drinking "back-yard" and commercial milk are presented, showing in particular areas within which doses of more than 5 rads to the thyroid may have been received.

Signed:

K.G. McNeill

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March 1 / 99

R. E. Jervis

R.E. Jervis

March 1, 1999

Date:

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Exhibit C

Academic and Professional Experience: R. E. JERVIS

I am presently Prof. Emeritus, Nuclear Science and Engineering., Univ. of Toronto, and a nuclear consultant^c.

After earning an undergraduate degree in Physics and Chemistry majors in 1949 at Toronto, I undertook master's and doctoral research in physical chemistry, programs that included formal instruction in nuclear physics and radiochemistry. As a research nuclear chemist at the Chalk River Nuclear Laboratories, Atomic Energy of Canada Ltd. starting in 1952, I engaged in radiochemical extractions of plutonium and heavy actinides, radioiodines and other fission products using Geiger counters, alpha chambers and rudimentary gamma-ray spectrometers (all techniques and procedures contemporary with and relevant to Hanford operations at the time). I participated in investigations into the monitoring of radionuclide releases, including iodine, from reactor and chemical processing stacks using various filter types. Other research done in cooperation with GE and Westinghouse involved at-power testing of the integrity of prototype power reactor and submarine fuel using rapid radioiodine detection for early fuel failure recognition. As developer in Canada of the applied radiochemical technique of RAA, radioactivation analysis, I have measured major and trace levels of many radioisotopes in solutions, air filters and biological materials.

In 1958 I accepted a faculty position at the Univ. of Toronto in the Applied Science and Engineering Faculty as Professor, Nuclear Science and Eng. Among academic research projects undertaken since (and published in 230 papers) have been: measurements of iodine and other inorganics in urban air filters; developing size - selective aerosol sampling with the aid of electron microscopy; application of different receptor models for air pollution source recognition and apportionment based on particle-size characteristics and correlated concentrations; studies of the chemical states of radioiodine as released from reactor fuel under accident conditions, their air/water/steam partitioning and the relative volatilities, solubilities and absorbabilities of I_2 , I^- , IO_3^- , organic-I and I-bearing aerosols. These latter studies were done for reactor safety in cooperation with AECL and the Ontario Hydro, Canada largest nuclear utility.

I have been active in developing and promoting environmental and nuclear safety standards with the CSA (Can. Stds. Assoc.), latterly as Chair, Advisory Comm. on Nuclear Safety of the Atomic Energy Control Board of Canada, regulator of all Canadian nuclear activities. I have been named Fellow: Can. Soc. Chemistry; Can. Nuclear Soc.; Royal Soc. of Canada; Amer. Scientific Affiliation; have been honored with 3 international medals and other of awards. A registered (licensed) Professional Engineer since 1963; consultant on research reactors and radioisotope applications to the UN Agency: International Atomic Energy Agency, Vienna; Associate Editor of an international radiochemical journal.

* (see attached C.V. for details)

Academic and Professional Experience: K.G. McNeill

A brief resume of my academic background is given here; a full CV is attached^δ.

I gained a First Class Honours degree in Physics from Oxford UK in 1947. My D.Phil (1950) was on the D-D (deuteron-deuteron fusion) reaction. Fellowships at Yale and Glasgow UK preceded a lectureship at Glasgow, then Associate and then Full Professorships in Physics at Toronto. Honours included visiting scientist status at Melbourne, Livermore, Ottawa (in all cases more than once) Harwell and Saskatoon, and status as Professor of Medicine at Toronto and Special Staff Member at the Toronto General Hospital. During the Toronto years research has been equally divided between pure nuclear physics (photodisintegration) and nuclear physics applied to medicine (mainly in bone disease, particularly osteoporosis).

The research work has resulted in just less than 200 papers published in refereed journals. There have also been a couple of books, several published lecture notes, and several major reports on nuclear emergency planning. I have been a consultant for safety in uranium mining and for the Departments of Health and of Labour of the Government of Ontario, for the federal (Canadian) Auditor General's office, and for the Atomic Energy Control Board.

For the decade or so up to 1997 I was the Technical Advisor to the Solicitor-General of Ontario with reference to off-site effects of accidents at nuclear power plants - this involved, amongst other things, chairing the Technical Advisory Committee and half its Working Groups, and some of its sub committees, dealing with source term estimations, meteorological modelling of distribution of radioactive emissions, dose prediction and protective actions. I chaired, for more than 10 years, the University's Radiation Protection Authority (the University has 230 professorial staff licenced to use radioisotopes).

I am a member of the Canadian Association of Physicists, of the Canadian Radiation Protection Association, and (until retirement) a Fellow of the (UK) Institution of Nuclear Engineers.

I am now an Emeritus Professor of Physics in the University of Toronto.

* (see attached C.V. for details)

**Errors in HEDR I-131 Release Estimates and Corrections
to Dose Estimates to Account For These Errors**

by

R.E. Jervis and K.G. McNeill

Report to the Hanford Radiation Litigation

Tom H. Foulds and Richard Eymann, Attorneys

Supplemental Report

March, 2004

University of Toronto

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Executive Summary

During the chemical separation of plutonium at Hanford in the state of Washington in the years 1944 to 1972, fission products, particularly I-131, were released to the atmosphere. A fraction of this I-131 was distributed in the downwind ecosystem and reached the human thyroid gland through various environmental pathways where became a potential for the induction of thyroid disease.

In this report we estimate the quantities of I-131 released, mainly during dissolution of irradiated uranium, calculate how that activity was distributed in the air, how much was ingested by cows and into milk as a source for human ingestion, and finally, how much was deposited in the thyroid of a typical person living at a particular place and how much dose was given to the gland.

The complex pathways for the iodine released from the Hanford Plant make accurate estimation of the final biological effects of the I-131 very challenging. However, certain facts are clear - for instance, the greater the release of I-131 the greater will be the potential for causing thyroid disease.

The HEDR approach (Heeb[1993]and Heeb [1994]) was reviewed by the present authors in 1999 (McNJ99). Corrections were made to the HEDR estimates of I-131 released into the plant - at times these corrections were of the order of a factor of two - and consideration was given to the fraction of I-131 which escaped into the atmosphere despite attempts to filter out this and other fission products.

The present report looks particularly at the Release Factors, the proportion of the I-131 in the plant which escaped into the atmosphere. One of the authors (REJ) has researched and published extensively in this field. The major difference between the HEDR estimations and the present work results from a new and better understanding of the role of organic-I in releases - the organic-I is much less reactive than, say, gaseous I_2 and I^- , and so was not trapped by most off-gas filters. After April 1951 the Release Factor estimated in this report is 11 times higher than that estimated by HEDR.

This large factor (of 11) makes a considerable difference to the estimated dose in practical cases. Thus the dose to an adult living in Ringold from 1950 through 1956 was twelve times greater than the dose calculated by HEDR (Napier 2002), while the isodose contour for a child born Jan 1 1945 and living on " back yard food " was significantly further distant from Hanford than for the equivalent contour based on the HEDR estimates. The HEDR Release Factors are too low, mainly because the removal efficiencies of filter devices

were appreciably overestimated and, the role of the organic-I was not properly taken into account.

A. INTRODUCTION

This report is supplemental to a previous document, McNJ99, in which we considered the validity and accuracy of HEDR estimates of I-131 releases from the Hanford Site 200 B, T and REDOX chemical separation plants over their operating lifetime (1944-1956). While some of the HEDR calculations were based on historical records of fuel radioiodine inventories and various release estimates, a number of significant assumptions and approximations were also made and several of these we consider to have been erroneous and to have led appreciable underestimates of iodine releases. In the period, 1944-1956, considered here, greater than 90% of total I-131 releases to atmosphere at Hanford occurred.

In McNJ99, correction factors were computed to be applied to HEDR [Heeb,1994] release estimates to allow for errors such as:

- (i) Exponential Cooling Times (ECT): correction was required for errors resulting from their incorrect linear averaging of fuel cooling (decay) times as a simplification for calculations. The McNJ99 ECT factors were based on true exponential I-131 decay over the time period from fuel discharge to processing.
- (ii) First-In-First Out Protocol (FIFO): corrections were required to correct for events in which average cooling times assumed by HEDR were not applicable because orderly queuing of fuel batches for cooling, dissolving and extraction was not always followed.
- (iii) Lag Time: another source of incorrect cooling times was the time difference between dissolution and extraction of fuel (so-called lag time). Cooling times assumed by HEDR were the period between reactor shutdown and extraction, not time of dissolving when the major evolution of iodine occurred.
- (iv) Radioiodine Release Factors (RF): corrections were applied in McNJ99 to HEDR iodine release factors to account for inefficiencies of the removal devices in the 1948-56 period. HEDR used best case scenarios based, in part, on unreliable stack release measurements to estimate a set of generic release factors which were quite low, at times by an order of magnitude.

Mathematical details of how the ECT, FIFO, LAG and RF correction factors were computed were given in McNJ99 and are not repeated here.

In this report, as a consequence of reviewing relevant Hanford and HEDR documents, and, critical reviews made by NRC, CDC [Anspaugh,2001] and other reputable experts, we have found compelling evidence that some of our earlier estimates of the correction factors

which needed to be applied to the HEDR estimates, deserved re-examination. In particular, evidence of additional FIFO violations, increasing attention to other chemical forms of

radioiodine, especially organic-iodides, which HEDR ignored, and the increased radiation doses to exposed populations that would result from our less conservative, revised corrections factors, were the impetus for this joint supplemental report.

B. FIFO:

In McNJ99 there were documented 10 cases, between the years 1954 to 1964, in which the emission of I-131 was greater than expected. These out-of-ordinary emissions were attributable to the processing of "greener" fuel than expected, and in its turn this was attributed to accidental violation of the First-In-First-Out protocol -- under this FIFO protocol the fuel first removed from the reactor was the first to be processed. (The Dec., 1949 "Green Run" was not included as one of these cases since, in the Green Run, the violation was deliberate [Harlan *et al*, 1950]. In the 10 cases above, six had emissions of less than 10 Ci, while in the other four, the emissions were 75 Ci, 28 Ci, 40 Ci and 60 Ci. This total of about 200 Ci occurred when the grand total (normal emissions plus these 200 Ci) was about 6000 Ci; the extra was thus about 3%.

Since McNJ99, three more extra-ordinary cases of greater than expected emissions that could be attributed to accidental violations of the FIFO protocol have been documented. These occurred in September 1959 [HW-62179], when the I emission was of 20 Ci, in April 1962 [HW-73514], 40 Ci, and Dec., 1963, 8 Ci [Keane, 1963]. All three of these cases occurred in the same period: 1954-64 as with the cases discussed above [HW-89067]. Adding in these 3 new cases gives a new total of about 200 Ci + 68 Ci. This totals to an extra of 268/6000 (Ci), or 4%. Just as McNJ99 rounded up the estimated (extra) emission from 3 to 5%, so it would be reasonable to round up the 4% to 6% as a result of these new findings. This value of 6% has been estimated on the basis of data for the period 1954 to 1964; however, McNJ99 justified its use over the whole range of reactor years of interest (except for the time period: Sept., 1946 to Dec., 1949 during which period the push dates were known for each particular batch, so no generalized FIFO correction should need to be made for that period).

A summary of revised FIFO factors is shown in Column 3, Table 2, as follows:

FIFO Correction Factors:

Dec 1944 to Aug 1946	1.06
Sept 1946 to Dec 1949	1.00
Jan 1950 to Dec 1956	1.06

C. ECT Corrections:

The exponential cooling time correction factors, ranging from 1.0 to 1.7, as shown in Column 4, Table 3 do not require any revisions here.

D. Lag Time Correction Factors:

Similarly, the lag time correction factors which ranged from 1.0, 1.17, 1.37 to 1.9 have not required further revisions here and are shown in Column 5, Table 3.

E. Radioiodine Release Factor Estimates:

The estimations of Heeb, [1993] and [1994], of emissions of I-131 from the Hanford Site 200 plants formed the basis of the calculation of doses for the HEDR94 and the Hanford Thyroid Disease Study [HTDS, 1999] and also, of Napier re-evaluation [Napier, 2002]. Questions have arisen since as to the accuracy and variability of these estimates. In a previous report [Jervis,1995], errors in I-131 stack and vegetation measurements were addressed in detail.

Napier [2002] reviewed the basic HEDR approach of using generic release factors for the various removal devices, presumably based on historical records, and proposed instead an alternative approach using stack measurements. Heeb[1994] had calculated the total I-131 inventory in processed U fuel from reactor power, irradiation and cooling time histories and then estimated the fraction of total iodine released to the atmosphere using RF, release factor values which he claimed were supported in part by historical stack data. Napier 2002 claims an independent approach to the calculation of the amount of radioiodine emitted to the atmosphere, based on historical stack measurements and verification of estimated emissions based on atmospheric monitoring at sites some kilometers from the Hanford site.

Initially, from the 1944 startup until water scrubbers and sand beds were used in 1948, HEDR estimated that most (~90%) of I-131 contained in the fuel had been released from the dissolvers and subsequent processing. After the scrubbers and filters were introduced, HEDR release estimates were reduced to about 30% and after silver reactors started to be used (1950-51), to about 1% of I-131 released from fuel.

This approach of HEDR had a number of substantial errors, including:

- (i) best performance estimates of water scrubber and sand bed removal efficiencies were used that were quite optimistic,
- (ii) silver reactor efficiencies for 1959-60 were applied back to the early 1950 during periods when records show that their performance had deteriorated drastically and regeneration was often required [Burger, 1961], and,
- (iii) there had been a complete neglect by Hanford scientists and HEDR of the existence of an appreciable proportion of organic iodides in the off-gases, forms of iodine that were not filtered or absorbed by removal devices and not measured in stack gas sampling.

Almost all of the I-131 of dissolved uranium fuel rods was released at two points: the dissolver off-gases (50% initially, 86% after ir-sparging [Warren, 1961] and most of the rest evolved during subsequent handling of dissolver solution. A small fraction (1-5%) was

entrained in condensate running down the stack walls [Smith,1946]. However for the first 4-5 years after startup, absolutely no engineered measures or devices were implemented to reduce I-131 discharges, notwithstanding evidence of high levels of radioactivity downwind and high levels on vegetation off-site and in distant communities [Healy,1946].

Among the media used in attempts to mitigate such releases were:

E.1 Fiber Filters:

Although there was little evidence that appreciable I-131 was evolving from the dissolvers and processing streams in particulates, a bank of fiber filters were installed in futile attempts to trap radioiodine from the off-gases. In current industrial use were the so-called ambridge absolute filters asbestos and other paper composite types such as the wartime CWS (gas mask) filters, some of which were used in sampling lines. However, glass fiber filters were selected for the stack gases out of concern for flammability and susceptibility to corrosive fumes. Probably, the composite filters would have filtered and absorbed more I-131 but the glass fiber filters were very ineffective (<5% removed) because the radioiodine was predominantly, 95-99%, in gaseous forms.

Filter tests done in the Chalk River radioactive exhaust stack by the author [Jervis, 1956] about 50 years ago using both CWS and Millipore membrane filters showed that very little iodine could be collected on filters. On the other hand, based on information published in 1934 [Robertson], granular charcoal filters would have been much more effective had they been selected, removing also organic-iodides.

E.2 Water Scrubbers and Sand Beds:

Wet scrubbers had been tried in unsuccessful attempts for n-line stack monitoring in 1945 and were also considered as a means of I-131 recovery for commercial, medical applications in mid-1946 [Healy,1946], rather than as a safety measure to reduce excessive vegetation contamination. However, by mid-1948 much concern was expressed by the AEC Reactor Safeguards Committee about Hanford I-131 stack emissions and GE was instructed that up to 99% needed to be removed [AEC, 1948]. Sand beds were reported to remove as much as 98% in early tests but this was for new beds. An October,1948 HEW report [HW-11449] indicated: widely varying efficiency reaching a maximum of 25% in one run, also, sand beds ranged from 27-68% [HW-13610]. Sand beds frequently became clogged by liquids and other chemicals, such as the January,1950 water clogging at the T plant. Further, in HW-67520 it was reported that bed efficiencies based on vent samples collected were not valid due to inadequacy of the sampling system.

Water scrubbers were considered to have efficiencies as high as 85% but, in other tests it was found that only about one-half of the I-131 was caught in water scrubbers [HW-7-6391]. The choice of water and soda ash (carbonate) for wet scrubber media was curious, if not negligent, because caustic scrubbers (having a much higher pH) were considerably more efficient for iodine removal, particularly from acidic fumes. Further, at

the time this decision was made, DuPont had had experience with caustic scrubbers at Oak Ridge where they had been used from 1944 at the RaLa (X-10) plant according to Hoffman [1999; see also, the attached schematics of the RaLa2 plant, 1945; Appendix].

Efficiencies of 80-90% for I-131 removal by water scrubbers were not realized in prolonged operation. Wichner [2004] reported that, from studies at Oak Ridge, their efficiencies were found to be unstable and were strongly affected by acidity when operated in the recycle mode. The kinetics of the I_2 hydrolysis reactions complicated removal processes in the scrubbers because, although the first step: $I_2 \rightarrow 2 I^-$ was rapid (reaction half-time ~ 5 min.) the second reaction: $HOI \rightarrow I^- + IO_3^-$ was very slow.

Herrmann and Herrmann [1996] judged the scrubber and sand bed efficiencies to be 40% and 30% respectively based on much research and operating experiences at German and French plants. Therefore, and with evidence of much lower operating performance at Hanford, these (Herrmann) values (40 and 30%, respectively) have been used here for revised RF estimates for the period May 8 up to Oct 0, when silver reactors were introduced.

E.3 Silver Beds:

Among the choices of media to trap iodine, it appears that silver was not initially selected although knowledge of classical iodine chemistry at the time would have indicated that silver ions and silver surfaces would tightly bind iodine from solution or from fumes. There was much experience from before the 20th century that thin layers of silver iodide on inert substrates such as were used in the photographic film industry, did not wash off even with strong acids.

At Hanford, it was discovered through a random observation in some laboratory tests around 1948, that silver and copper metal pieces in equipment exposed to radioiodine became highly radioactive. Following some small-scale development work, beds to treat the off-gases were made up of ceramic pieces (addles coated with silver nitrate. Initially in 1950 it was asserted that 99% of I-131 was removed by these silver reactors [Wolff, 1951].

However, early in 1951, shortly after their installation, it was found that they were not performing as expected and that there had been a substantial increase (more than ten-fold) in I-131 releases [Pas and Soldat, 1951]. These failing reactors were allowed to remain in operation from Feb 1 to Aug 1 before they were replaced. It was concluded that the operating temperatures had exceeded 370° F and the silver nitrate had melted and slid off the coatings [Blasewitz, HW-21959, Aug 1]. The bed failure was detected mainly by off-site vegetation monitoring. Also, the beds were subject to fouling and saturation by AgCl and AgI. Excess chlorine in the off-gases competed with and replaced iodine atoms for sites on the silver coating [duPlessis and Sutter, 1969]. It was concluded that the Ag reactors needed to be regenerated regularly after about 140-200 tons of U fuel had been processed [Burger, 1991]. However, there were times during the mid-50 when the beds were not regenerated until after 600-800 tons had been dissolved [Warren, 1961]. Burger also stated that the main function of the Ag reactors was to hold I-131 long enough to decay, not for the long term as long-term buildup was marginal and used beds contained mostly AgCl, (less than 2% was