TSP

Technical Steering Panel
of the Hanford Environmental Dose Reconstruction Project

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Source Term Subcommittee

May 1988 - September 1994

SOURCE TERM SUBCOMMITTEE

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EXECUTIVE SUMMARY

The Source Term Subcommittee monitored the progress of Battelle Pacific Northwest Laboratory researchers on the Project's Source Term task. The Source Term is the amount of radioactive materials that were released from the Hanford Site. Subcommittee members also carried out independent research on the Source Term.

Research on the atmospheric Source Term initially focused on air releases of iodine-131 (I-131) from 1944-47. Very detailed and precise knowledge of the irradiation and cooling histories of the discharged fuel was necessary to allow calculation of the iodine releases. After the initial scoping part of the study, much more information on fuel discharges and dissolvings was discovered among the Hanford records. This allowed for greater accuracy in determining this information. Enough information was found to generate hourly release data for 1944-1949 (the periods of largest releases). From 1949 to 1971, iodine releases were determined on a monthly basis.

Air releases of ruthenium-106, ruthenium-103, strontium-90, plutonium-239 and cerium-144 were determined on a monthly basis from 1944 through 1972. Some radionuclides of public interest (tritium, carbon-14 and argon-41) were evaluated as yearly averages.

The source term for 11 radionuclides released to the river was calculated. Five of these contributed over 94 percent of the dose. The five were: sodium-24, phosphorus-32, zinc-65, arsenic-76 and neptunium-239.

Subcommittee members also calculated the source term for an experimental release in December 1949 called "the Green Run", and calculated the estimated maximum thyroid doses from iodine 129 releases.

INTRODUCTION

The Hanford Environmental Dose Reconstruction Project (HEDRP) was carried out by the primary contractor, the Pacific Northwest Laboratory (PNL), a National Laboratory operated by Battelle-Pacific Northwest Laboratories. To direct and guide the HEDRP, a management committee was formed. This committee, the Technical Steering Panel (TSP), was given the responsibility and authority to direct, modify and approve of the research methods and procedures.

The calculation of the historical doses received by people who were exposed to the radioactivity released from operations at the Hanford nuclear facilities required several steps. These progressed from determining the amount of radioactivity released through calculating the concentration of radioactivity distributed around the environment through determining the environmental contamination of foodstuffs to calculating human uptake and exposure. When there are no data on the amount of contamination that was in food and water or on vegetation, it is necessary to calculate the amount of radioactivity received by people.

The first stage in the sequence of calculations was the determination of the amount of radioactivity released from the Hanford facilities. This is called the Source Term of the calculation. The Source Term is then used as input to the remainder of the calculation of the impact on the public of Hanford operations.

The TSP organized itself into several subcommittees each with responsibilities in one of the critical tasks necessary for completion of the HEDRP. One of these subcommittees, the Source Term Subcommittee, was responsible for monitoring the progress of the PNL researchers on the Source Term Task of the HEDRP team. Members of the Subcommittee followed the work of and interacted closely with the Battelle staff members carrying out the source term research. Subcommittee members also carried out independent research on various aspects of the determination of the source term.

At the completion of the main part of the HEDRP and the end of the Battelle contract, the Technical Steering Panel was reorganized into a smaller group with fewer subcommittees. The Source Term Subcommittee was disbanded. This report represents a summary of the activities of the Source Term Subcommittee during its tenure from 1988 to 1994.

Meetings of the Subcommittee were carried out on a regular basis during the general meetings of the Technical Steering Panel which were held periodically at various locations around the Pacific Northwest. At each general meeting of the TSP a Source Term Subcommittee meeting was held. The proceedings of each Subcommittee meeting was reported to the TSP during the general meeting at which it was held. Each Subcommittee report is recorded in the official transcript of the TSP meeting at which it was held. Subcommittee reports are displayed as **Appendix D**.

The results of independent research on Source Term issues that were carried out by members of the Subcommittee have been published or are in the process of being published in the open literature. Copies of these papers are attached as Appendices.

SOURCE TERM SUBCOMMITTEE

The Source Term Subcommittee was formed during the initial meeting of the TSP in May, 1988. The members of the subcommittee and their areas of expertise were:

Maurice A. Robkin, Ph.D., P.E.: Chairman: Nuclear Engineering, Health Physics
Mary Lou Blazek: Health Physics
Glyn Caldwell, M.D.: Epidemiology
Pat McGavran, Ph.D. (appointed 1991): Toxicology
Bernard Shleien, Pharm. D., CHP: Radiation Dosimetry, Health Physics
John Till, Ph.D., CHP: Environmental Assessment, Health Physics

BACKGROUND FOR THE HANFORD DOSE RECONSTRUCTION PROJECT

People who lived in the Northwest were exposed to radioactivity during the time that the nuclear facilities at Hanford operated. They were mainly exposed to radioactivity from the chemical reprocessing plants that was released to the air and carried by the wind. To a lesser extent, they were exposed to radioactivity produced by activation of the cooling water of the once-through production reactors which was then discharged to the Columbia river. These reactors were called "once-through" because the cooling water was drawn directly from the river, purified and conditioned, passed through the reactor to carry away the fission heat, and then discharged back into the river.

THE NATURE OF THE DOSIMETRY PROBLEM

The largest radiation doses to the people affected by the radioactivity released from Hanford occurred during 1944-1947, the period of the greatest air releases of the iodine isotope I-131 from the chemical reprocessing plants. I-131 has a half-live of 8 days which is long enough for the wind to spread it around and for it to enter all pathways to people.

Radioactivity released to the Columbia does not spread out beyond the confines of the river. It is mixed into the bulk of the river flow and carried downstream. Its concentration decreases by radioactive decay, certain physical and biological removal processes or by dilution by tributaries entering the river. People who used the river for drinking, fishing, recreation or other activities were exposed.

During the early period of Hanford, there were few reactors operating. By the mid-1960's there were 8 once-through production reactors operating at much higher power than were the 3 original Hanford reactors during their earliest period of operation. The mid-1960s

was the time when the radioactivity released to the river was the largest. However, even during this period, the contribution to the radiation dose due to activity in the river was much smaller than that due to the I-131 releases to the air in the 1944-1947 period.

SIGNIFICANT ISSUES

Iodine

The Source Term research in Phase I, the first part of the HEDR project, concentrated on the 1944-1947 air releases of I-131. The release of I-131 depended on how much of it was present in the reactor fuel slugs when they were dissolved in the chemical plants. This depended on the power the slugs generated while they were irradiated in the reactor and the length of time they were stored after discharge from the reactor before being dissolved. The storage, or "cooling" time allowed the 8 day I-131 to undergo significant radioactive decay, thus greatly reducing the amount in the fuel before it was sent to the dissolvers.

A production reactor is very large, and the fuel in different parts generated plutonium at different rates. The amount of plutonium produced in a given batch of fuel is proportional to the amount of time it spent in the reactor and the local power density. The product of the time and local power density is called exposure or burnup, usually measured in units of megawatt-days per ton of uranium fuel, or Mwd/t. In the first few years of operation, when the local exposure reached about 200 Mwd/t, that batch of fuel was discharged. This criterion was set so that the plutonium would have desirable properties and be at about the same concentration in each batch of discharged fuel. As a result, fuel irradiated at the higher power level of the reactor center was discharged more often than fuel irradiated at the lower power at the edge of the reactor.

Because of its short half-live, the concentration of I-131 in the discharged fuel is proportional to the power at which the fuel was irradiated and not the burnup . The more frequent discharge of higher power fuel meant that overall the discharged fuel had a higher average power than the average power of the fuel in the reactor. The average concentration of I-131 in the discharged fuel was higher than what would be expected from considering only the average power of the reactor. The first approach was to divide the reactor into zones and use an analytical fit to the power distribution. This permitted a reasonable approximation to the enhancement of the I-131 due to the higher power level of the discharged fuel.

The initial transport model was a very complex deterministic one in which each puff of airborne release was followed around the countryside based on the records of the wind speeds and directions everywhere within the study area. This model used a monthly average time scale. At the end of the initial scoping study, it became clear that even this level of detail was not sufficient due to the great variability of the wind field. The variability meant that it was necessary to know what the weather was doing at any given hour and

how much iodine was being released during that hour. The atmospheric dispersion model was upgraded to include the variability of its parameters. The variability was accounted for by using a statistical computational method called Monte Carlo. It was necessary to consider the I-131 air release in the 1944-1947 period on an hour-to-hour basis, an enormously more difficult task. To keep track of the amount of I-131 in the fuel being dissolved required a very detailed and precise knowledge of the irradiation and cooling histories of the discharged fuel.

After the initial scoping part of the study was over, much more information on fuel discharges and dissolvings was discovered among the Hanford documents. The great increase in information allowed the Battelle source term researchers to take an innovative new approach for calculating the release of I-131 to the air.

The new model considered the records of fuel discharges amounts and burnups from the reactors, allowed for the distribution of power that the discharged fuel experienced, used the records of fuel dissolvings in the reprocessing plants to obtain the fuel cooling and dissolving times and solved the equation for the production and decay of the I-131. Account was taken of the recharging of fresh fuel to replace discharged fuel. The model predictions of the fuel discharges agreed extremely well with the records of the burnup of the fuel discharges kept by the operating personnel at the reactors.

To evaluate the iodine content in the discharged fuel, a computer code, called the Reactor Model (RM) was written by the Source Term Task researchers. The reactor model calculated the history of fuel irradiations using the statistical Monte Carlo technique. A code to evaluate the time history of iodine released from each fuel reprocessing was written. The output from this code, called the Do Iodine code (DOI) together with the output from the RM code were combined as input to a code that calculated the actual iodine release on an hourly basis. This code, the Source Term Release Model (STRM) is also a statistical code. The code sequence was run for the releases from B Plant and T Plant, the fuel reprocessing plants. The output from the STRM code for the chemical reprocessing plants was a time history of releases of radioactive iodine from these plants over the calculation period to use as input to the atmospheric dispersion calculation.

The model permitted the increase and decrease of the iodine concentration to be followed as fuel discharged from different parts of the reactor was dissolved. Experimental data on the fraction of the iodine in the fuel released to the air when fuel was dissolved was combined with the dissolving information to generate the complex pattern of the iodine emissions. Enough information was found to generate hourly release data for 1944 through 1949. The code sequence was run 100 times. The output from these statistical (Monte Carlo) models for the releases was 100 sets of hourly releases of I-131 for the period 1944-1949. The 100 sets of release values was then passed to the transport researchers calculating the dispersion of the radioactivity throughout the environment. From 1949 to 1971, the iodine released was determined on a monthly basis.

Other Significant Airborne Radionuclides

In addition to I-131, the other important radionuclides released to the air are ruthenium-106 and ruthenium-103, strontium-90, plutonium-239 and cerium-144. Air releases of these were determined on a monthly basis from 1944 through 1972. Some nuclides of public interest also were evaluated as yearly averages over this period. These are tritium, carbon-14, and argon-41.

River Activity

There are many radionuclides that contributed to the activity of the discharged coolant from a production reactor. They arose from activation of minerals carried in the river water, from radioactivity released from fuel element failures, and from corrosion of activated impurities in the aluminum process tubes and fuel cladding. Eleven of these were considered to be the most significant for contributing radiation doses to people using the Columbia River for fishing, agriculture and recreation. These are sodium-24, phosphorus-32, scandium-46, chromium-51, manganese-56, zinc-65, gallium-72, arsenic-76, yttrium-90, iodine-131, neptunium-239, and a combination of activities referred to as "gross beta". Of these, the five that contributed over 94% of the dose are sodium-24, phosphorus-32, zinc-65, arsenic-76, and neptunium-239.

For the initial scoping calculation of the doses from the radioactivity in the river, the starting point was directly from measured concentrations. The doses were calculated for 1964-1966 for people living along the River between Priest Rapids Dam and McNary Dam. This was the period when all eight of the production reactors were operating at their highest power. In the 1960's there were very good measurements of radioactivity in the environment. Since the Pathway part of the study starts with concentrations in air, water, plants, and animals that provide food and drink to people, this was a very direct way to do the dose calculation.

After the initial calculation, the evaluation of the impact of water-borne radioactivity in the river was extended all the way to the ocean. The time period was expanded to include 1944-1992. Unfortunately, during the early history of the Hanford operation the environmental measurements were much fewer and much poorer. For this period of time, a source term and a transport calculation were necessary. Not only were the environmental measurements poor, but the measurements of the activities released in the reactor coolant as it was discharged from the reactors were poor. Wherever measurements of the radioactivity in the discharged reactor coolant was reported, they were used. For the times when there were missing measurements, a statistical model (Monte Carlo) was developed to fill in the data. This model used the measured activity in the discharged coolant and reactor operating parameters to generate distributions of water borne activity to sample for the Monte Carlo calculation of the activity in the coolant at

times when there were no observations.

SPECIAL PROJECTS

The Green Run

In December of 1949, an experiment was carried out at Hanford in which I-131 was deliberately released to the air in order to test the ability of instruments located at great distances from the source to detect clandestine releases. M.A. Robkin, Chair of the Source Term Subcommittee, under instructions from the TSP undertook an independent analysis of the radiation dose due to I-131 released during the experiment. He used the existing declassified report of the Green Run and brought up-to-date the methods used by the original authors. He concluded that about 11,000 curies of I-131 were released. After the completion of this study, another document was found which indicated that the value is more likely to have been about 9,000 curies. His result has a standard error of about 30 percent. That is, the actual value is estimated to lie between about 3,600 and 14,400 curies with only a five percent chance that it lies outside of this range. The results of this study have been published in the open literature (Robkin 1993). A copy of this paper is included as **Appendix A**.

Other Releases

The major contribution to the radiation doses from Hanford were due to the radioactivity discharged to the river in the reactor coolant and to the air from the chemical reprocessing plants. However, there were other releases as well from a large number of other facilities at Hanford. To complete the HEDR project, estimates will be made to the extent possible for the releases to the air and water from all of the Hanford facilities for the entire operating period up to 1990. These will include all of the releases for which records can be found other than those to the river in the reactor coolant and to the air from the reprocessing plants. These other releases represent only minor contributors to the doses that people off-site would have received.

Iodine-129

There has been considerable public interest in the impact of releases of I-129 since it is a very long-lived isotope. Maurice Robkin, Chair of the Source Term Subcommittee, and Bernard Shleien, committee member, under instructions from the TSP undertook an independent analysis of the radiation dose due to I-129. Considerations of the behavior of iodine in the environment indicate that it should persist in the soil within the root zone of plants. The removal rate from this zone was estimated to be about 1% per year. Thus, when long-lived I-129 is released to the environment, it can contribute to the radiation dose of people for an entire lifetime.

The HEDRP results indicated that the individual maximally exposed to I-131 lived to the east of the Hanford site in the vicinity of Ringold, WA. Assuming that such an individual would also be the one maximally exposed to any release of I-129, a maximum dose from I-129 was calculated. The maximum dose was based on assuming that all of the I-129 in fuel reprocessed at Hanford was released to the air and that the maximally exposed individual had a rural lifestyle. There were about 55 curies of I-129 in fuel reprocessed at Hanford from 1944 through 1986. The maximum dose from I-129 occurred for an individual born in 1954. This person received a total of about 900 millirem to the thyroid from the year of birth through 1995. The greatest part of the dose comes from drinking milk from a cow eating fresh pasture grass. The small remainder of the dose comes from eating leafy vegetables, other vegetables, and cereals.

A paper on this maximum dosimetric impact of I-129 on the people affected by Hanford releases has been accepted for publication in the journal Health Physics. A reprint of the paper is included as **Appendix B**.

HEDRP REPORTS

The HEDRP research staff of Battelle have published many reports covering the various aspects of their work on the dose reconstruction. The HEDR reports relevant to the source term task are listed in **Appendix C**. Members of the TSP have undertaken various studies on HEDRP related source term topics. The reports on these studies are listed in **Appendix E**.

APPENDIX A

EXPERIMENTAL RELEASE

OF ^{131}-I: THE GREEN RUN

Paper

EXPERIMENTAL RELEASE OF ^{131}I: THE GREEN RUN

Maurice A. Robkin*

Abstract—**In December 1949, a large amount of ^{131}I was released to the air at Hanford during the dissolving of irradiated uranium fuel for a classified military experiment called the "Green Run." Reports of the release have varied from about 0.15–0.3 PBq. Using, as a guide, the reported measurements of ^{133}Xe released during the experiment, the amount of ^{131}I released has been reanalyzed. The results indicate that about 0.40 ± 0.12 PBq (11 ± 3 kCi) was released, somewhat larger than the largest previous estimates.**
Health Phys. 62(6):487–495; 1992

Key words: ^{131}I; contamination, environmental; emissions, atmospheric; radioactivity

INTRODUCTION

DURING its operations from 1944 to the present, the Hanford nuclear facilities released radionuclides to the environment. The releases were much larger in the early days before the development of control equipment and restrictive regulatory limits. These releases resulted in radiation doses to the surrounding populations. An extensive study of these doses is currently under way. This study, called the Hanford Environmental Dose Reconstruction Project (HEDRP), is being carried out by the Pacific Northwest Laboratories under the direction of a Technical Steering Panel (TSP). The study, which began in 1988, is a successor to the Hanford Health Effects Study carried out by the state of Washington and is the result of intense public interest and concern about the releases.

One of the singular events in the history of the Hanford releases, the Green Run experiment, occurred in December 1949. The experiment was part of the development of a monitoring methodology for intelligence efforts regarding the emerging Soviet nuclear program. The complete details of the experiment remain classified. A declassified description of the experiment has been released (Jenne and Healy 1950).

Declassified reports dealing with or mentioning the Green Run show considerable disagreement in the release amount (Jenne and Healy 1950; Paas and Singlevich 1950; Singlevich 1950; Roberts 1958; Anderson 1974). The values have ranged from 0.15 PBq (4,000 Ci) (Singlevich 1950) to 0.30 PBq (7,780 Ci) (Jenne and Healy 1950). There have been expressions of concern by various members of the public and by representatives of the states of Washington and Oregon about these disagreements.

It is important for the HEDRP that source terms be determined as accurately as possible. Although the earlier estimates indicated that the Green Run release represented less than 2% of the total ^{131}I releases from the Hanford facility, a defensible value is required for the HEDRP dosimetry. The purpose of the present study is to provide as good an estimate of the release from the Green Run experiment as is possible from the information contained in the historical record.

THE GREEN RUN

Examination of the *100 Areas Monthly Report* for November 1949 (Barnard 1949), the *P Division Monthly Report* for November 1949 (Lee 1949), and the *Daily Operating Conditions Report for the 100 Areas* (Anonymous 1950) indicates that the "F" Reactor supplied the irradiated fuel for the Green Run Experiment. The reactor outage began at 3 a.m. and reached nominal zero power at 4:50 a.m. on 16 November 1949. Fuel was removed (pushed) from the reactor on 17 November 1949 (Jenne and Healy 1950).

Approximately 1,000 of the discharged irradiated fuel slugs (two tons) were made available for the Green Run experiment. The radioactivity in the slugs was allowed to decay for only about 2 wk. The decay time was much shorter than usual to ensure that there would be sufficient ^{131}I ($T_{½}$ = 8.04 d) available for the experiment.

On 1 December 1949 these slugs were put into a clean dissolver at the reprocessing plant. The aluminum jackets were dissolved on the graveyard shift of 2 December (12 midnight to 8 a.m.). The dissolving of the declad and rinsed uranium slugs began at 8 p.m. that same day. The plant filters were bypassed to allow all of the evolved iodine to escape.

When a reactor is shut down in a normal way, the control rods are driven into the core. The neutron flux decreases quickly as the reactor goes highly subcritical. There is initially about 6.5% residual power that will slowly decrease with the decay of fission products. For a reactor that has been operating for a long time, the

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initial shutdown fission product power will decrease to about 1% of the full reactor power in a little over 3 h. The actual neutronic shutdown of the "F" reactor can be taken at 3 a.m. on 16 November 1949. The cooling time to the start of dissolving is 401 h measured from 3 a.m. on 16 November 1949 to 8 p.m. on 2 December 1949.

Not all of the ^{133}Xe and ^{131}I were discharged at the same time. The discharge of the xenon was spread out from the start of dissolving over 750 min while the iodine was emitted from about 50 to about 970 min after the start of the dissolving (Jenne and Healy 1950). Jenne and Healy (1950) give the time history of the evolution of these isotopes as the dissolving proceeded. The iodine release rate as a function of time is very symmetric around the midpoint of the discharge. The xenon release rate is nonsymmetric having a very rapid rise to a maximum at about 2.5 h after the start of dissolving followed by an extended decrease over a period of about 10 h (Jenne and Healy 1950). Using a discharge-rate- and decay-weighted average time-to-release gives average values of 4.8 h and 8 h for the average additional cooling from the start of dissolving for xenon and iodine, respectively. The total cooling times for these two isotopes are then 406 h for ^{133}Xe and 409 h for ^{131}I with an uncertainty of not more than 2 h.

The ^{133}Xe available for release from the dissolved fuel comes from the ^{133}Xe in the fuel at the time of reactor shutdown plus the ^{133}Xe produced by the decay of the 21-h ^{133}I in the fuel at shutdown. About 2.5% of the xenon activity comes from ^{131}Xe that was produced in the fuel during the cooling period from the decay of ^{131}I, and its contribution to the ion chamber current in the Jenne and Healy (1950) measurement is accounted for.

The ^{131}I available for release during dissolving comes from ^{131}I in the fuel at reactor shutdown plus the decay of the 1.35-d ^{131m}Te in the fuel at shutdown. The contribution from the reservoir isotopes amounts to about 15% additional for xenon and about 5% additional for iodine over the activities that would be present at dissolving from just the ^{133}Xe and ^{131}I inventory at reactor shutdown.

In this paper, the estimate of the amount of 8.04-d ^{131}I released from the fuel is based on the amount of 5.243-d ^{133}Xe released during dissolving as reported by Jenne and Healy (1950). All of the contributing isotopes were at radioactive equilibrium at the time the reactor was shut down for fuel discharge. The Bateman (1910) equations were solved for the equilibrium activities and for the activities of ^{133}Xe and ^{131}I in the fuel at that time of dissolving.

At the time of the Green Run experiment, the reactor fuel was made up of solid slugs of uranium metal clad in aluminum. The average specific power density in F reactors was about 1.1 MW Mg^{-1} (1 MW ton^{-1}) (Lockwood 1958). It is assumed that at this power density there is little migration of fission products within the uranium metal and only the fission products formed in the uranium that was actually dissolved are available for release. The amount of iodine discharged from the plant stack is proportional to the assumed value of the fraction evolved from the process solutions.

As part of the experiment, samples were taken of the gases and vapors that escaped from the dissolver and were carried up the stack of the reprocessing plant. A pump pulled about 0.472 L s^{-1} (0.5 cfm) through a sampling line at the 15.24-m (50-ft) level of the stack into the analysis building. The gas stream was passed through a scrubber solution and expelled out the building stack. Samples of the gas stream were collected about every 15 min into evacuated 1-L spherical flasks ("boiling flasks") containing an iodine scrubber solution (Jenne and Healy 1950).

After standing several days, the scrubber solution removed all of the iodine from the sample. After this time, each scrubber flask was connected to an evacuated 1-L spherical ionization chamber made from a boiling flask and the xenon was allowed to divide between the two flasks. The ion chamber was carefully bled to equilibrium with the atmosphere (Jenne and Healy 1950). The ionization current generated in the ion chamber was measured by a vibrating reed electrometer. Jenne and Healy quote Thorburn (1950) who describes the chamber as operated at 90 V in an air-conditioned room maintained between 22–24°C and 50% relative humidity. Thorburn (1950) reported constant ionization current per unit activity above 67.5 volts.

Jenne and Healy (1950) expressed the release rate of xenon as curies per minute by converting the chamber ion current to flask activity by use of a calibration factor (0.115 fA Bq^{-1}) (4.25 pA μCi^{-1}). The derived their calibration factor by assuming that: 1) the average specific ionization of beta particles was 120 ion pairs (ip) per centimeter (ip cm^{-1}); 2) the average path length in the ion chamber was 6 cm; and 3) the ionization from gamma rays was negligible. The results of the ion chamber measurement were reported as the activity discharge rate of ^{133}Xe assuming that: 4) the half-life of ^{133}Xe was 5.4 d and 5) that the flow rate up the stack was 11.8 m^3 s^{-1} (25,000 cfm).

The first assumption appears to be based on applying to the entire ^{133}Xe beta spectrum the specific ionization for an electron whose energy is equal to the average beta energy and assuming that the specific ionization was constant throughout the path of the electrons. Using the average energy of the main beta to determine the energy deposition is not adequate for the following reasons: 1) the specific ionization is not linearly proportional to the electron energy; 2) the average energy of conversion and Auger electrons is about one-third that of the beta particles; and 3) the fractional energy deposition of the lower energy discrete electrons is much greater than for the beta particles; more than one-half of the energy deposition from ^{133}Xe in the

chamber comes from conversion and Auger electrons that Jenne and Healy (1950) did not take into account.

The second assumption is incorrect. The average uncollided path length to the wall in a spherical volume of radius, R, for the particles emitted uniformly within the volume by an isotropic emitter is $0.75R$ (Lind 1928). For a 1-L spherical volume, R = 6.2 cm, and the average path length is 4.65 cm.

The third assumption is correct.

The fourth assumption is slightly higher than the current value of 5.243 d (Walker et al. 1989). For a standing time in the collection flask of as much as 5 d, the difference in the reported discharge rate of ^{133}Xe would be <2%.

Even in 1949, air flow rates could be accurately measured, so the fifth assumption is probably reasonable and good to within a few percent.

Thorburn (1950) reported ion chamber backgrounds on the order of 2–5 fA for a 2-L chamber. The 1-L chamber used in the Jenne and Healy experiment would have about one-half this background. For the smallest xenon release rate measured by Jenne and Healy (about 200 MBq s^{-1}), their calibration factor and assumed stack air-flow rate implies a chamber current of about 1 pA, at least 400 times the background.

Measurement of the iodine in the scrubber solution gave a value 2 orders of magnitude lower than expected (Jenne and Healy 1950). They discarded the results and took samples of the condensate in the sampling line. Based on these samples, Jenne and Healy (1950) reported a release of 0.29 PBq of ^{131}I. No description of the analytical methodology was given. Because Jenne and Healy's lack of methodological detail for obtaining the reported iodine release, this analysis was based on the release of ^{133}Xe.

METHODOLOGY

General overview

Two models to calculate the energy deposition in a spherical ion chamber were examined. The first was exponential attenuation for the beta particles and the transmission probability model of Kobetich and Katz (1969) for the discrete energy conversion and Auger electrons. The second was based entirely on the Kobetich and Katz model. Evaluation of the two approaches for beta particles was made by comparing the calculated calibration factors with experimental values for a set of ion chamber measurements of ^{85}Kr in air. ^{85}Kr decays by beta emission with negligible production of conversion or Auger electrons (ICRP 1983). Gamma rays and x rays deposit negligible energy in the ion chambers and were ignored.

The measured calibration factors for spherical ion chambers containing ^{85}Kr in air were kindly provided by the Victoreen Instrument Co.† The Victoreen data are shown in Table 1. Details of the exponential model and the Kobetich and Katz model are given in the Appendix (see also Table 2).

Table 1. Experimental calibration factors for spherical ion chambers[a].

Volume (mL)	nA × mL × μCi^{-1} (reported)	pA μCi^{-1} (equivalent)	aA Bq^{-1} (equivalent)
282	1.00	3.55	96.0
524	2.28	4.35	118
1020	5.44	5.33	144

[a] Data from Ulman;† values ± 20%.

Model choices

The agreement with the Victoreen experiment was much better using the exponential model for the absorption of the beta particles. This model was used in calculating the contribution of the continuous spectrum beta particles to the calibration factor for ^{133}Xe. The exponential model does not apply to electrons emitted with discrete energies. For the discrete energy conversion and Auger electrons emitted in the decay of ^{133}Xe, absorption fractions were calculated based on the Kobetich and Katz (K-K) transmission probability model. The energy deposition from gamma rays and x rays is very small and was neglected.

Evans (1955) shows that the exponential attenuation model holds well in the energy range from 100–3000 keV. A numerical evaluation of the K-K model was made by comparing the absorption fractions in a 6.2-cm air-filled spherical ion chamber for both allowed and first-forbidden spectrum betas with maximum energies from 100–700 keV. The results showed that the ratio of the absorption fraction from the adjusted exponential model to that given by the K-K model using a first-forbidden spectrum starts out at about 1.3 at the highest energy and decreases rapidly with decreasing energy. At the lowest energy, the ratio had fallen to about 0.75. When an allowed beta spectrum was used in the K-K model, the ratio decreased from 1.1 at the highest energy to about 0.7 at the lowest energy with a minimum (0.65) at 200 keV. An approximate average for the ratio for beta spectra of 100–200 keV maximum energy is 0.7. Further details of this calculation are given in the following uncertainty section.

There are large numbers of electrons emitted in these lower-energy spectra with energies similar to those of all of the discrete electrons emitted by ^{131}Xe and ^{133}Xe. As a result, there is considerable uncertainty in the absorption fractions calculated by the K-K model. For the discrete electrons, their energy depositions were calculated using the K-K model, summed and multiplied by 0.7. The scaled energy deposition was used with a large associated uncertainty. The details of the energy absorption calculation are described in the Appendix.

† Personal communication (1990), Ulman, R. Measured calibration factors for spherical ion chambers containing ^{85}Kr in air. Victoreen, Inc., 10101 Woodland Avenue, Cleveland, OH 44104.

Table 2. Experimental and theoretical absorption fractions, f, for ^{85}Kr in Victoreen air-filled ion chambers.

Volume (mL)	Radius (cm)	m	f_e[a]	f_{KK}[b]	f_v[c]	f_v/f_e	f_v/f_{KK}	k[d]
282	4.07	0.130	0.0912	0.0614	0.0820	0.899	1.32	15.3
524	5.00	0.160	0.110	0.0745	0.100	0.909	1.33	15.5
1020	6.25	0.200	0.135	0.0915	0.123	0.911	1.33	15.5

[a] Exponential attenuation model.
[b] Kobetich and Katz attenuation model.
[c] Calculated from Victoreen data.
[d] Adjusted value of coefficient.

DATA FOR ^{133}Xe

The 1-L ion chamber sphere had a radius of 6.2 cm. Assuming that the measurements were carried out at 22°C and 1 atm pressure, the density of air in the ion chamber was 0.0012 g cm^{-3}.

The average energy to produce an ion pair in air is taken as 33.85 ± 0.15 eV (ICRU 1979). The charged particles emitted in the decay of ^{133}Xe are given in ICRP Publication No. 38 (ICRP 1983).

ABSORPTION FRACTIONS FOR ^{133}Xe PARTICLES

The absorption fraction for each of the emitted particles from ^{133}Xe and ^{131}Xe was calculated based on the methods described above. For each kind of energetic electron emitted, the energy deposited in the ion chamber is given by the product of $f \times y \times E$ where f is the absorption fraction of the electron, y is the yield in particles per decay, and E is either the average beta energy or the energy of the conversion of Auger electron. Summing these products over all of the emitted discrete energy particles, decreasing the sum to 70% of the calculated value, and combining the result with energy deposition from the beta particles, gives an energy deposition of 44.8 keV per decay.

THE CALIBRATION FACTOR FOR ^{133}Xe

The corrected calibration factor for ^{133}Xe in a spherical ion chamber can now be calculated:

$$I = \frac{q}{w}\,\delta E, \qquad (1)$$

where $q = 1.602 \times 10^{-19}$ C $ip_{/}$. For $w = 33.85$ eV ip^{-1}, and $\delta E = 44.8$ keV per decay, the calibration factor to be used for the present analysis becomes: $I = 0.21$ fA Bq^{-1} (7.8 pA μCi^{-1}).

XENON AND IODINE RELEASE

Jenne and Healy (1950) reported their measurements of the xenon discharged up the plant stack in Ci min^{-1} of ^{133}Xe based on their calibration factor of 4.25 pA Ci^{-1} (0.115 fA Bq^{-1}). Fig. 1 is taken directly from their report and shows their data as they presented them, including the curve. They reported that the area under the curve was about 20,000 Ci (740 TBq). To examine that value for this paper, the indicated data points were read from the curve as data pairs of time and activity discharge rate. These activities were transformed to their natural logarithms. The resulting values are shown in Fig. 2.

To obtain the total release, the release rate was integrated over the release interval. Note that there appear to be four distinct time intervals in each of which the logarithms of the discharge rates vary linearly with the time. Total dissolution time was taken to be 750 min (12.5 h) representing the interval between the very small xenon discharge rates at each end of the dissolution process.

The logarithms of the data in each region were fit by linear regression to a straight line using the computer code SYSTAT (Wilkenson 1990). The results are shown in Table 3. In each time interval, the release rate is given by:

$$\text{Rate} = \exp(A + B\,T). \qquad (2)$$

The total amount released is estimated by integration of eqn (2) over all four time-intervals to give:

$$\text{Release (Ci)} = 750 \sum_{i=1}^{4} \frac{e^{A_i}}{B_i}\left(e^{B_i T_{i2}} - e^{B_i T_{i1}}\right), \qquad (3)$$

where T_{i1} and T_{i2} are the times at the beginning and end of the ith time interval, respectively. Evaluation of eqn (3) with the parameters of Table 3 gives a total ^{133}Xe release of 29,000 Ci (1.07 PBq). The uncertainty in this estimate is discussed in a later section.

The ^{131}I release, A131, is given by:

$$A131 = Xe\,\frac{CF_{JH}}{CF_R}\,\frac{AI}{AX}\,f_r, \qquad (4)$$

where Xe is the activity of ^{133}Xe measured by Jenne and Healy (1950), CF_{JH} is the calibration factor used by Jenne and Healy, CF_R is the calibration factor used in this paper, AX is the ^{133}Xe activity, AI is the ^{131}I activity contained in the dissolved fuel at the mean time of dissolution, and f_r is the fraction of iodine released from the process solutions.

The data shown in Fig. 1 are based on ion current readings. Jenne and Healy reported the activity release rate from the ion current based on their value for the

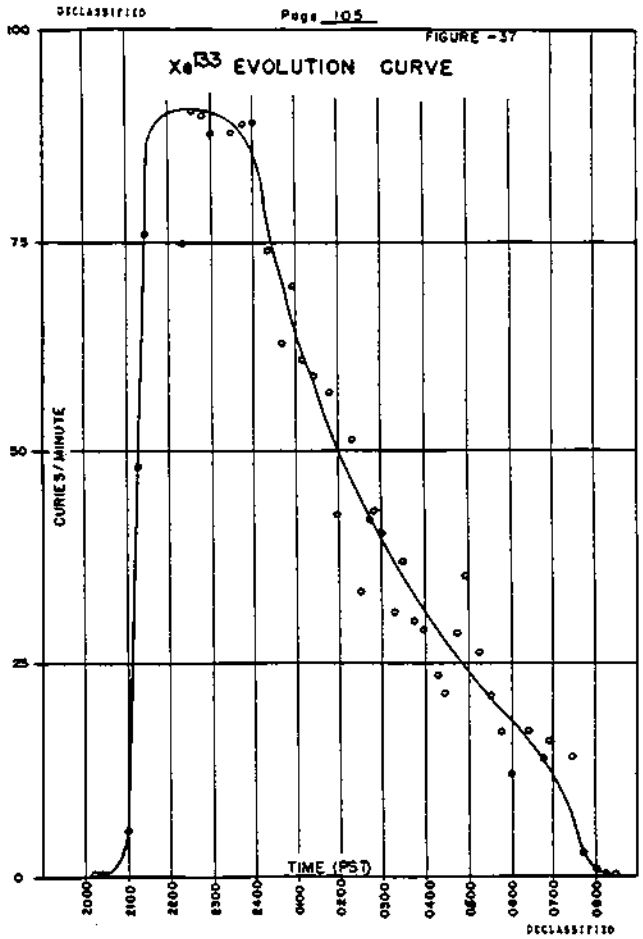

Fig. 1. Jenne and Healy (1950) data for ^{133}Xe release during the Green Run. PST = Pacific standard time.

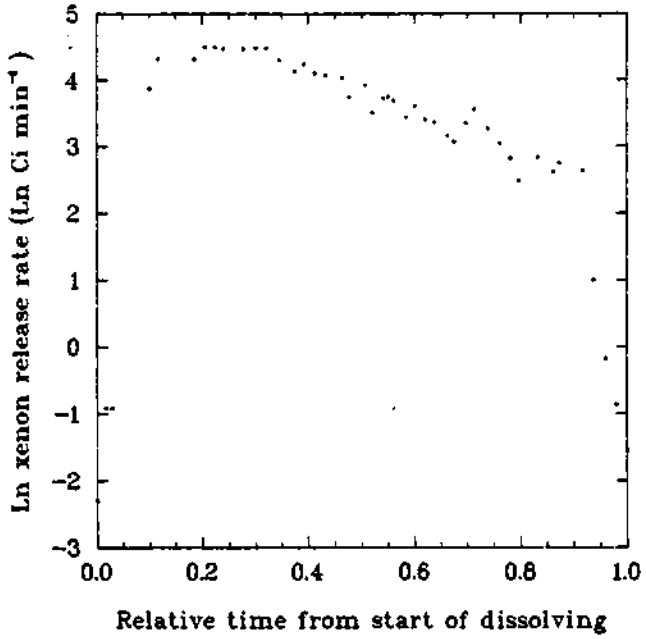

Fig. 2. Natural logarithm (Ln) of Jenne and Healy (1950) data for ^{133}Xe release during the Green Run vs. relative time. Relative time is unity at 750 min after the start of fuel dissolution. Linear regression fits were made in the intervals 0–0.117, 0.117–0.320, 0.320–0.917, and 0.917–1.

stack flow rate (25,000 cfm, 11.8 m^3 s^{-1}) and their calibration factor (4.25 pA μCi^{-1}, 0.115 fA Bq^{-1}). For a given ion current, the estimate of the amount of activity released is inversely proportional to the calibration factor. For a calibration factor of 7.8 pA μCi^{-1} (0.21 fA Bq^{-1}), the estimate of a release of 29,000 Ci (1.07 PBq) of ^{133}Xe obtained from the Jenne and Healy data becomes:

Release = (4.25/7.8) 29,000 Ci

= 16.000 Ci (0.6 PBq).

The solution to the Bateman (1910) equations gave the activities in the dissolved fuel at the effective time of release as the following (F is the fission rate in the dissolved fuel while it was irradiated in the reactor): $8.63 \times 10^{-3}F$ Bq for the 5.243-d ^{133}Xe, $2.17 \times 10^{-4}F$ Bq for the 11.9-d ^{131m}Xe, $1.56 \times 10^{-5}F$ for the 2.19-d ^{133m}Xe, and $6.91 \times 10^{-3}F$ for the 8.04-d ^{131}I. These values account for the contributions to xenon and iodine from the equilibrium activities at reactor shutdown of ^{133}Xe and ^{131}I as well as the equilibrium activities of ^{133}I and ^{131m}Te. These activities were then decayed to the effective time of release. The parameters for the Bateman equations were taken from Crouch (1977) for fission yields, from the General Electric Chart of the Nuclides (Walker et al. 1989) for decay energy (Q) values, from ICRP Publication No. 38 (ICRP 1983) for the particle energies and yields and from Sarantites et al. (1965) for the ratio of the independent fission yield ratios of the ^{131}Te isomers. The ratio of the activity of ^{131}I to that of ^{133}Xe at their effective times of release was calculated to be 0.8.

Estimates of the fraction of the radioiodine contained in the dissolved fuel released from the dissolver solution vary from about 65% to about 90% (Dreher 1945; PNL 1991; Jenne and Healy 1950). Kirkendall (1952a, b) reported measuring a release fraction of 85% during the actual dissolving step. Of the remaining 15%, he reports that approximately 5% is evolved during later processing. The Kirkendall reports are the only ones found of actual measurements of the iodine release fraction. Work (1946) reports that about 5% of the iodine released by fuel processing dissolves into the stack drainage and is recovered at the bottom of the stack. Starting from 90% of iodine released from the process solutions, the fraction emitted from the stack would then be 85%. For this paper, the release fraction was taken to be 85% ± 15%.

On this basis, the central estimate of the amount of ^{131}I released is 0.40 PBq (11,000 Ci). This value is somewhat larger than the estimate made by Jenne and Healy (1950) of 0.29 PBq (7,780 Ci) based on their

Table 3. Regression analysis of ^{133}Xe release[b]: Ln rate = $A + B\,T$[a].

T[a]	A[b,c]	B[b,c]	R[d]
0–0.117	−2.28 ± 0.31	56.7 ± 4.2	0.989
0.117–0.320	4.25 ± 0.08	0.826 ± 0.333	0.711
0.320–0.917	5.40 ± 0.11	−3.15 ± 0.18	0.960
0.917–1.0	47.21 ± 5.2	−49.0 ± 5.5	0.982

[a] T is unity at 750 min after start of dissolving.
[b] Regression coefficients for Ln-transformed Jenne and Healy (1950) data with SYSTAT code (Wilkenson 1990).
[c] Dimensions of regression coefficients are Ln(Ci min^{-1}). To convert regression coefficients A and B to SI units [Ln(Bq s^{-1})], add 20.24 to A. B remains unchanged.
[d] R = correlation coefficient.

analysis of the condensate in the sampling line from the plant stack to the laboratory building.

UNCERTAINTY

None of the parameters used in this analysis are known exactly. For most of them, the uncertainty in the values is given in the literature as a standard deviation. For some, the error is due to lack of exact knowledge and may be either positive or negative. In this work, these errors are treated as random with an expected value of zero. For some, the error is bias due to model errors, but the dominant contributors to the uncertainty are random errors.

Bias errors

Using the Kobetich and Katz model for the discrete energy particles gives an unknown bias for their absorption fractions in the Jenne chamber. The observed variation in the ratio of the beta absorption fractions based on the exponential model to those based on the K-K model suggests that the K-K-derived absorption fractions for the discrete energy particles may be too large, but the amount of the overestimate is unknown.

For the case of an allowed beta spectrum with maximum energy of 100 keV (average 26 keV), the exponential model gives an average absorption fraction of about 0.6 while the K-K method gives a value of about 0.85; a ratio of about 0.7. For a maximum beta energy of 200 keV (average 55 keV), the values are 0.38 and 0.58, respectively. The ratio is still about 0.7. Taking an uncertainty of 25% around a ratio of 0.7 gives a reasonable range for the ratio. To adjust the apparent overestimate in the energy absorption of the discrete electrons given by the K-K model, the absorption was scaled by 0.7 ± 25%. The uncertainty spans the estimates for the absorption fraction given by the exponential model for betas in the energy range from 0–200 keV. The central value is consistent with the absorption fraction calculated with the exponential model for a beta spectrum with average energy of 55 or 26 keV. In order to estimate the uncertainty of the ^{131}I release, the uncertainty in the absorption fraction was combined with the other errors as though they all were random and independent.

The contribution to the fissions by plutonium has been neglected. This neglects the differences in the fission yields for ^{133}Xe and ^{131}I from plutonium fissions. The fuel for the Green Run experiment was discharged at about 400 MWd Mg^{-1} (Barnard 1949). At this exposure, the contribution to the total fissions from plutonium is very small.

Random errors

The estimate of the release of ^{131}Xe involves a sequence of connected functions. The error associated with each of these steps is propagated assuming that the various physical parameters are independent.

The quantity, Q, that is functionally dependent on other parameters in the sense of $Q = Q(x_1, x_2, - - - -, x_n)$ has a variance that can be estimated by (Bowen and Bennett 1988):

$$\sigma_Q^2 = \sum_{i=1}^{N} \left[\left(\frac{\partial Q}{\partial x_i} \right)^2 \cdot \sigma_{x_i}^2 \right]. \quad (5)$$

Each of the x_i parameters in Q may, itself, be functionally dependent on other parameters with a variance estimated in the same way as that for Q.

Numerical values

The counting errors in the Jenne and Healy measurements were small. That is, if one uses their calibration factor the assumed values for the average energy expended per ion pair and the specific ionization of the beta particles, then the number of ion pairs that they observed was large and Poisson counting error very small.

The values obtained by reading the data points as given in the Jenne and Healy graph have a random error that was assumed to include the random experimental error in the Jenne and Healy data. These errors are reflected in the variances in the fitted values for the linear regression on the log transformed release values. It is these variances that are included in the variance propagated to the final result.

The uncertainties in the fission product chain yields were taken from Crouch (1977). No data were available for the errors in the fractional yields of isotopes in a mass chain. The parameters for the particles emitted by ^{85}Kr, ^{131}Xe, and ^{133}Xe were taken from ICRP Publication No. 38 (ICRP 1983), but this reference does not give the associated errors. The coefficients of variation (COV) for the yields and energies of the particles emitted from ^{85}Kr and ^{133}Xe were taken from the National Nuclear Data Tables (NNDT 1991). These two isotopes are the major contributors to the energy depositions considered.

^{131}Te is only a small contributor to the ^{131}I activity at the time the fuel was dissolved and its contribution to the overall error is ignored. ^{133}I contributes about 16% and ^{133m}Xe contributes about 3% to the ^{133}Xe

Table 4. Victoreen ion chambers parameter values.

Parameter	Reported value	Reported standard deviation	Reference
Victoreen chambers			
Volume	282 mL	—	Ulman 1990
Calibration	96 aA Bq^{-1}	20%	Ulman 1990
Volume	524 mL	—	Ulman 1990
Calibration	118 aA Bq^{-1}	20%	Ulman 1990
Volume	1020 mL	—	Ulman 1990
Calibration	144 aA Bq^{-1}	20%	Ulman 1990
Jenne chamber	1000 mL	—	Jenne and Healy 1950
w	33.85 eV ip^{-1}	0.15 eV ip^{-1}	ICRU 1979
K [from eqn (A7)]	15.4	3.4	This paper
Air density	1.2 mg mL^{-1}	3%	Assumed
^{85}Kr decays			
Half-life	10.72 y	0.19%	NNDT 1991
$\beta 1$ energy — max	687.4 keV[a]	3.0 keV[a]	NNDT 1991
ave	251.4 keV	0.8 keV	NNDT 1991
yield	99.563%	0.010%	NNDT 1991
$\beta 2$ energy — max	170.4 keV[a]	5 keV[a]	NNDT 1991
ave	46.6 keV	1.4 keV	NNDT 1991
yield	0.437%	0.010%	NNDT 1991

[a] Calculated from Q value and decay scheme.

inventory at the time the fuel was dissolved. Their contribution to the overall uncertainty is also small. The ratio of the ^{131}I to the ^{133}Xe activities at the time of fuel dissolving gives one of the scale factors for converting the observed (Jenne and Healy 1950) xenon release to the corresponding iodine release. The contributions to the relative error in the overall predicted value of iodine release from the relative errors in these activities are small compared to the those of the uncertainties in the iodine release fraction, the calibration factor, and the regression fit to the experimental xenon release data. Thus, the uncertainty in the iodine-to-xenon activity ratio is adequately calculated using only the values for the relative errors in the overall mass chain yields.

The regression on the experimental xenon release data and the integral of the release rate gave a result with a COV of 10%. The assumed COV of the iodine release fraction is nearly 20% (0.85 ± 0.15). The uncertainty in the calibration factor arises from the uncertainties in the calibration factors reported in the Victoreen data,[†] in the air density in the ion chambers, and in the fit to the constant in the exponential model. The Victoreen data were reported with an uncertainty of 20%. The air temperature and pressure were assumed to be at 22 ± 3°C and 101.1 ± 3.4 kPa, respectively. The resulting COV of the calibration factor was 14.5%. The regression on the experimental xenon release data gave a COV of 10% for the total xenon release. The ratio of the 131 to 133 chain yields, which were decayed to the time of dissolving, has a COV of about 3% based on the data from the National Nuclear Data Tables (NNDT 1991). A COV of 10% was assigned to the iodine-to-xenon activity ratio to encompass all of the uncertainties in the parameters that make up that ratio. Based on these uncertainties, the net COV of the iodine release was calculated to be 27%. The value is rounded off to 30%.

CONCLUSIONS

The amount of ^{131}I released to the atmosphere during the Green Run experiment is calculated to be 0.40 PBq ± 30% (11 kCi ± 30%). This result is based on a number of improvements over the analysis used by Jenne and Healy (1950), who reported a release value of 0.29 PBq (7,780 Ci) based on measuring the sampling line condensate. The difference between the Jenne and Healy value and the value calculated in this paper is not statistically significant. Without further experimental evidence to test the model used in this analysis and more information about the analytical procedure used by Jenne and Healy, the similarity in the values should be considered fortuitous.

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APPENDIX

The exponential model

It has been observed (Evans 1955) that beta particles with a continuous energy spectrum appear to attenuate exponentially through matter. For attenuation in aluminum, a reasonable fit to the mass absorption coefficient ($cm^2 g^{-1}$) for a beta spectrum with maximum energy, E, in MeV is given by (Evans 1955):

$$\frac{\mu}{\rho} = \frac{K}{E^{1.14}} = \frac{17}{E^{1.14}} \text{ cm}^2\text{g}^{-1}, \quad \text{(A1)}$$

where E is the end-point energy of the beta spectrum in MeV. It is assumed that the ion chambers were operated at 22°C and 1 atm pressure. For any reasonable range of temperature and pressure, the density of the air in the chambers will not vary by more than about 3%. The assumed density was 1.2 mg mL^{-1}. The maximum beta energy for ^{85}Kr is 0.6874 MeV (Walker 1989). Assuming simple density scaling, the linear beta attenuation coefficient calculated for air with eqn (A1) is 0.031 cm^{-1}.

Consider beta particles emitted isotropically from an arbitrary radial point within the spherical volume into a unit solid angle in an arbitrary direction. The shape of the energy spectrum remains nearly constant throughout the absorption (Evans 1955). Thus, the amount of energy deposited by the betas before they escape is proportional to the fraction of the particles that do not strike the wall averaged over the entire volume of the ion chamber. Treating the betas as exponentially attenuating, we can use the expression of Price et al. (1957) for the current, j ($cm^2\ s^{-1}$)$^{-1}$, at the surface of an absorbing sphere. The absorption fraction, f, is then the complement of the ratio of the total leakage, $4\pi R^2 j$, to the total source in the sphere:

$$f = 1 - \frac{3}{4m}\left[1 - \frac{1}{2m^2} + \left(\frac{1}{m} + \frac{1}{2m^2}\right) e^{-2m}\right], \quad \text{(A2)}$$

where m is the optical distance from the center to the surface of the sphere, μR.

The Kobetich and Katz model

Kobetich and Katz (1969) give an empirical equation for the characteristic distance of an electron of

kinetic energy, T, in a medium of atomic number, Z. Their eqn (4) is:

$$R = AT\left(1 - \frac{B}{1 + CT}\right), \quad \text{(A3)}$$

where

$A = (0.81\ Z^{-0.38} + 0.18) \times 10^{-3}$ g (cm^2 keV)$^{-1}$

$B = 0.21\ Z^{-0.055} + 0.78$

$C = (1.1\ Z^{0.29} + 0.21) \times 10^{-3}$ keV^{-1}.

Eqn (A3) can be inverted to give the residual energy corresponding to a given residual distance:

$$T = \frac{-[A(1 - B) - CR] \pm \{[A(1 - B) - CR]^2 + 4ACR\}^{1/2}}{2AC}. \quad \text{(A4)}$$

^{85}Kr has a first-forbidden-unique beta decay whose energy spectrum is given by (Siegbahn 1955):

$$\frac{dn}{dw} = K(w_0 - w)^2 w^2[(w^2 - 1) + (w_0 - w)^2][\alpha(Z) + \beta(Z)\sqrt{w - 1}], \quad \text{(A5)}$$

where $\alpha(Z)$ and $\beta(Z)$ are given by Schenter and Vogel (1983):

$\alpha(Z) = -0.811 + 4.46 \times 10^{-2}\ Z + 1.08 \times 10^{-4}\ Z^2$, and $\beta(Z) = 0.673 - 1.82 \times 10^{-2}\ Z + 6.38 \times 10^{-5}\ Z^2$, for $T < 1.2\ mc^2$; or $\alpha(Z) = -8.46 \times 10^{-2} + 2.48 \times 10^{-2}\ Z + 2.37 \times 10^{-4}\ Z^2$ and $\beta(Z) = 1.15 \times 10^{-2} + 3.58 \times 10^{-4}\ Z - 6.17 \times 10^{-5}\ Z^2$, for $T \geq 1.2\ mc^2$, and where w is the total relative beta energy $(1 + T\ (mc^2)^{-1})$ and K is a normalization constant.

For each beta of kinetic energy, T, in the beta spectrum of ^{85}Kr, emitted at the radial point, r, into the unit of solid angle defined by $d\Omega = 2\pi \sin\Theta\ d\Theta$, eqn (A4) is applied to find the residual range: R(residual) = $R(T) - d(r,\Theta)$, where the distance to the wall is given by:

$$d(r, \theta) = r\cos\theta + [R^2 - (r \sin\theta)^2]^{1/2}. \quad \text{(A6)}$$

The energy corresponding to this residual range, E(residual), is weighted by the probability of reaching the wall and subtracted from T. The difference is the amount of energy that the particular beta particle deposited in the ion chamber before it struck the wall. These energy depositions in the spherical ion chamber are numerically integrated by Simpson's rule over all energies of the beta spectrum for all angles and radii. The ratio of this energy deposition to the total energy emitted as beta kinetic energy in the ion chamber by the ^{85}Kr is the average absorption fraction.

Testing the models

The Victoreen data for the ion chamber measurements of ^{85}Kr were used to test eqn (A2). More than 99.5% of the decays of ^{85}Kr are by a beta with E_o = 0.6874 MeV (from Q value and decay scheme) and E_{av} = 0.2514 MeV (ICRP 1983). There is a very small amount of energy produced by a low probability decay of a much softer beta. With the absorption fraction computed with eqn (A2), the average exponential model absorption fractions calculated for the three Victoreen ion chambers are compared with the absorption fractions, f_v, corresponding to the experimental data assuming the average energy to produce an ion pair is 33.85 ± 0.15 eV (ICRU 1979).

The Kobetich and Katz model was programmed in QuickBasic[‡] and the absorption fractions for the three Victoreen ion chamber measurements of ^{85}Kr were calculated. The 170-keV (max) beta of ^{85}Kr is emitted with an intensity of 0.437% and is ignored.

The results of these calculations are shown in Table 2. As can be seen, the exponential model gives an absorption fraction, f_e, about 10% higher than f_v, for each of the volumes while the Kobetich and Katz model values for f_{KK} are 33% lower. Thus, the exponential model seems to be the better representation for the absorption fraction of beta particles.

The parameters for the attenuation coefficient in eqn (A1) were obtained by fitting observed data on beta absorption in aluminum (Evans 1955). For absorption in air, one might consider either a different constant or a different exponent on the energy term. For low-Z materials, the energy dependence is relatively constant in Z (Evans 1955). What might be expected to change in the stopping power are the terms characteristic of the material such as mass, electron density, and atomic weight. The constant term can be considered to capture the characteristics of the absorber material.

The mean free path, m_v, was evaluated for each ion chamber volume by numerically inverting eqn (A2) to find the value for m that yields the observed absorption fraction, f_v. An adjusted value for the constant, K, in eqn (A1), is then found as shown in eqn (A7):

$$K = \frac{m_V \times E^{1.14}}{R \times \rho}. \quad \text{(A7)}$$

These results are also shown in Table 2. The average value for K from the three measurements is 15.4. This value was used in the exponential model for evaluating the fraction of the ^{133}Xe betas absorbed in the Jenne and Healy ion chamber.

[‡] QuickBasic V. 4.5 —Programming in BASIC, Microsoft Corporation, 1 Microsoft Way, Redmond, WA 98052–6399.

■ ■

Correction

$d(r,\theta) = r\cos\theta + [r_0^2 - (r\sin\theta)^2]^{1/2}$ (A6)

r_0 = spherical ionization chamber radius

APPENDIX B

ESTIMATED MAXIMUM THYROID DOSES

FROM 129-I RELEASES FROM THE HANFORD SITE

FOR THE YEARS 1944 - 1995

ESTIMATED MAXIMUM THYROID DOSES FROM ^{129}I RELEASES FROM THE HANFORD SITE FOR THE YEARS 1944-1995

by

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ABSTRACT

^{129}I, a very long lived fission product, persisted in the environment after discharge from the chemical reprocessing plants at Hanford. The significance to human health is evaluated by considering the maximum possible impact it might have. Allowing the total production of ^{129}I to be discharged to the air and considering the location where the maximally exposed individuals reside, the lifetime cumulative thyroid dose is calculated as a function of birth year. The maximally exposed individual at the maximum exposure location was born in 1954 and accumulates a thyroid dose between 1954 and 1995 of slightly over 9,000 μSv (900 mrem) at an average rate of 220 μSv (22 mrem) per year. Over the same period of time, the soft tissue dose from background radiation due to both external and internal radiation is about 30,000 μSv (3,000 mrem).

INTRODUCTION

Radioiodine may have been a potential environmental and health hazard in the area around the Hanford site because of its production during the process of making plutonium, and its subsequent discharge during fuel dissolution. Prior to the 1950s, methods for trapping radioiodine released from the chemical reprocessing plants at Hanford were relatively inefficient compared to later techniques. Until 1951, silver reactors, which efficiently trap radioiodines and prevent their release to the atmosphere, were not routinely operative or suffered from periodic malfunctions.

Once in the environment, radioiodine is transferred through the food chain, particularly through the air-plant-milk-human pathway, and concentrates in the human thyroid gland. Because of its relatively abundant production and short half-life (8.04 days), most attention has correctly been focused on ^{131}I. However, ^{129}I, with a long half-life (15.7 million years), persists in the environment and is only slowly removed by natural processes such as leaching downwards through the ground. Its long half-life means that its specific activity (activity per unit weight) is about one billion times smaller than ^{131}I. Hence, its persistence in the environment, weighed against its low activity per unit weight, is one consideration in judging its potential hazard. The ICRP metabolic model for iodine (ICRP 1979) indicates a clearance half-time for the thyroid of 80 days and a clearance half-time for the rest of the body of 12 days. The dose to the thyroid gland from I-129 is thus entirely influenced by the biological behavior of iodine rather than its long physical half-life. Essentially all of the dose from ^{129}I is delivered during the first year after its intake.

^{129}I is a fission product that, because of its very long half-life, steadily accumulated in the fuel of the production reactors during irradiation. In the earliest days of Hanford operation before there were any filters in the reprocessing plant system, all of the gases and most of the volatiles were discharged to the atmosphere through the plant stack. Of these, the radioiodines were the most significant for radiological impact.

The release of radioactive iodine to the atmosphere depended on the efficiency of the off-gas filters. These evolved from none, to water scrubbers and sand filters to packed-column silver reactors. The silver reactor columns were packed with ceramic berls coated with silver nitrate solution. The silver reacted chemically with iodine to trap solid silver iodide on the berl surfaces. When they were working correctly, the recorded efficiency of the silver reactors for iodine generally ranged from 99% to nearly 100%. The isotope of concern, 8 day ^{131}I, decayed within the silver reactors so that the amount that built up was limited. Long-lived ^{129}I, on the other hand, continued to accumulate.

The silver nitrate charge in the silver reactors would periodically become depleted resulting in deterioration of their trapping efficiency and requiring recharging with fresh silver nitrate solution. In addition to the decreased trapping efficiency at the end of a duty cycle it was suspected that the silver reactors occasionally became poisoned with chlorine arising from impurities in chemical reactants. The chlorine competes with and displaces iodine from the silver. With sufficient chlorine poisoning, the iodine captured in the filter would be driven off (Burger 1955, duPlessis 1969, Strachan 1978) . There were also occasional filter failures when overheating melted the silver nitrate from the berls or when mechanical failures occurred. These sporadic failures resulted in bursts of iodine release.

During the periods when a silver reactors could not retain iodine such as when it was poisoned with chlorine, the accumulated ^{129}I could be released and escape out of the stack. A model for accounting for all of the phenomena that modify the effective efficiency for trapping ^{129}I over a duty cycle has not yet been developed. In this paper, consistent with the evaluation of maximum possible impact, we assume that all of the ^{129}I produced in the reactor fuel, while it originally may be trapped in the silver reactors, was discharged from the reprocessing plant stack when the silver reactor efficiency was compromised by one of the failure mechanisms. The determination of the release source term then becomes one of determining, from the fuel irradiation records, the inventory of ^{129}I in the fuel when it was discharged from the production reactors.

The ^{129}I release is combined with the results of a model that gives the annual committed thyroid dose to the maximally exposed individuals from the residual activity in the environment from a unit release during the commitment year or a prior year. Location and lifestyle parameters are selected to give a reasonable maximum thyroid dose. The dose commitments are then summed to give the cumulative dose from birth to the year 1995. Individuals born between 1924 and 1995 are evaluated. An individual born in 1924 would reach 20 years of age at the start of releases. That is, any individual born after 1924 would not be an adult during some part of the release period.

METHOD

^{129}I is the last beta-decaying member (isobar) of the mass-129 fission product chain, decaying to the radioactive isomer ^{129m}Xe which then decays to the stable ground state, ^{129}Xe. Except for the isomer, ^{129m}Te ($T_{1/2}$=33.6 d), all of the precursors of ^{129}I have half-lives of an hour or less. Most of the mass-129 fission product chain appears as ^{129}I in the fuel at the time it is discharged from the production reactors. We assume that all of the mass-129 chain is in the form of ^{129}I. To examine the reasonableness of this assumption we estimate the fraction of the chain in the form of ^{129m}Te at the time of dissolving the fuel.

In the very early days of Hanford operation when fuel was discharged after irradiation for a relatively short time and also cooled for a short time, of the order of the half-life of the 33.6 day Te isomer, the fraction of the mass-129 fission product chain in the form of the

isomer would have been relatively large. Any tellurium in the fuel at the time of reprocessing would have been sent to high-level waste storage where decay of ^{129m}Te would generate ^{129}I. The liquid wastes in the storage tanks at Hanford do, in fact, contain ^{129}I (Droppo, et al. 1991).

The amount of ^{129}I in the fuel at the time of reprocessing is given by where f is the

$$A_{I129} = K P y \left[t - \frac{(1-f)}{\lambda_{Te}} \left(1 - e^{-\lambda_{Te} t}\right) e^{-\lambda_{Te} T} \right] \qquad (1)$$

branching ratio for the decay of the short-lived precursor of Te, λ_{Te} is the decay constant of the 33.6 day ^{129m}Te, K is a conversion constant, P is the average power density in the fuel, y is the yield of the mass-129 chain (atoms/fission), t is the irradiation time in the reactor and T is the cooling time for the fuel from the time of discharge from the reactor to the start of reprocessing.

It has been assumed that conversion to other nuclides of members of the mass-129 decay chain by neutron capture during irradiation is very small. The fraction of the total mass-129 chain production stored as ^{129m}Te at the time of dissolving is given by

$$F = \frac{(1-f)}{\lambda_{Te}\, t} \left[1 - e^{-\lambda_{Te} t}\right] e^{-\lambda_{Te} T}. \qquad (2)$$

Over the history of the Hanford production reactors, fuel irradiation times ranged from about 30 days to about 300 days while cooling times ranged from 30 days to several years (for N-Reactor fuel). For some illustrative cases, the fraction of the mass-129 chain sequestered as ^{129m}Te at the time of dissolving is:

Irradiation time days	Cooling time days	^{129m}Te fraction
30	30	0.145
30	90	0.042
100	100	0.021
300	90	0.0091
300	300	0.00012

There was very little fuel processed with irradiation time and cooling time as short as 30 days. Assuming that all of the mass-129 chain is in the form of ^{129}I at the time of dissolving is sufficiently accurate for the purpose of estimating the potential hazard from this isotope and is slightly conservative as well.

Heeb (1991) has evaluated the nuclide inventory in Hanford fuel as a function of burnup using both the N-Reactor spectrum and the CANDU reactor spectrum in the ORIGEN2 code. The neutrons in the N-Reactor core are considerably more energetic (harder) than those in the CANDU core (softer). The neutron reaction rates of the component atoms in the core of a reactor is strongly dependent on the energy distribution of the neutron spectrum. The ORIGEN2 code accounts for fissions from all sources and tracks all of the fission products with time and burnup. He has shown that the inventory of ^{129}I produced over a burnup range of 100 to 1000 megawatt-days per ton (Mwd/ton) varies by an average of only 4 percent from the graphite moderated N-Reactor harder neutron spectrum to the D_2O moderated CANDU reactor softer neutron spectrum. Thus, the ^{129}I inventory calculated as a function of burnup for an N-Reactor spectrum should be quite similar to what would be computed for the other Hanford production reactors whose neutron energy spectra lie much closer to that of the N-Reactor than to that of a CANDU reactor.

Heeb (1993) has kindly made available his results for the inventory of ^{129}I as a function of burnup obtained from the ORIGEN2 code. His values for the amount of ^{129}I contained in the 7.85 pounds of uranium in a standard slug of naturally enriched Hanford fuel are given in Table 1 as a function of the burnup in Mwd/ton.

Since the inventory of ^{129}I is so insensitive to the neutron spectrum, it has been assumed that the production of ^{129}I per Mwd/ton in both depleted (0.3%) and enriched (0.95%) uranium will be similar to the production in natural (0.72%) uranium fuel. That is, the production of ^{129}I as a function of burnup does not depend on which Hanford production reactor the fuel came from.

To expedite the estimate of the ^{129}I inventory, Heeb's inventory results were fit to the analytical form

$$M_{I129} = 2.086 \times 10^{-5} (w_T - 300) + 6.258 \times 10^{-3} e^{-0.00125 w_T} \quad g/slug \qquad (3)$$

where W_T is the burnup in Mwd/ton.

The values predicted by the analytic fit differ from the ORIGEN2 results by no more than about one percent up to 4000 Mwd/ton.

The half-life of ^{129}I is so long that one gram of the isotope generates only 6.55 MBq (177 microcuries). Converting the fitted yield of ^{129}I to its activity in the fuel per metric ton (1 ton = 1.1 ton), Bq/Ton, as a function of specific burnup, W_T, gives

$$A_{I129} = 7.64 \times 10^{4} (w_T - 300) + 2.30 \times 10^{7} e^{-0.00125 w_T} \quad Bq/tonne \qquad (4)$$

The dosimetric impact of the release of ^{129}I on the population surrounding Hanford was addressed by considering the radiation dose received by a maximally exposed individual. The results of the Hanford Environmental Dose Reconstruction Project (Farris et al. 1994) indicated that the individual maximally exposed to the releases of ^{131}I in the period from 1944 through 1947 would be an infant residing around Ringold, Washington. This is a rural area about 19 km east of the chemical reprocessing plant in the 200 East area. This individual consumed fresh milk from a cow grazing on fresh pasture during the growing season. The pasture was contaminated by deposition of airborne ^{131}I released from the Hanford fuel reprocessing facilities.

Since an individual does not remain an infant forever, in this paper we consider the cumulative dose received by a rural individual born in each year from 1924 through 1995 and exposed to the persisting environmental contamination from releases of ^{129}I from the year of birth through 1995 while living at Ringold. Food consumption patterns for both urban and rural individuals and for male and female consumers as a function of age for the years 1945 to 1957 have been evaluated by Anderson, et al (1993). They give values for fresh and stored milk consumption as a function of the age of both urban and rural consumers. In this study, it is assumed that fresh milk is equivalent to milk produced from cows on fresh pasture. Fresh milk consumption ranged from 70% to 95% of total milk consumption for rural consumers. Because of the long half-life of ^{129}I, the differences between fresh and stored whole milk are not important.

The peak year for ^{129}I discharge is 1964, and the 1957 food consumption estimate was the closest year for which the Anderson report gives data. Based on these age dependent food consumption data for rural individuals averaged over males and females, the thyroid cumulative dose equivalent due to the release of a unit of ^{129}I activity (1 Ci) was calculated for the maximum individual at 19 km east of the fuel reprocessing area at 200 East as a function of age for the age groups for which the food data were available. These groups were <1 yr, 1-4 yrs, 5-9 yrs, 10-14 yrs, 15-19 yrs, 20-34 yrs, and >34 yrs. The calculations were done using the GENII computer code (Napier et al. 1988).

The GENII code allows for removal of radioactivity in the soil by mechanisms such as leaching down to greater depths than those near the surface where plants grow. The GENII default value for the removal rate of iodine from the soil is 0.8 per year (80% per year). However, Kocher and Killough (1986) have analyzed the global cycling of ^{129}I and found, based on data from Savannah River, that the mean residence time in the upper 30 cm of soil was 81±12 y based on a multi-layer diffusion model. For the upper meter of soil, the mean residence time was found to be 900±140 y. Leafy vegetables have shallow roots while some root vegetables and forage plants may have roots that extend below the 30 cm depth. Since only a single leaching rate is allowed in GENII, a value of 0.01 per year was selected for ^{129}I. This value is based on assuming that 80% of the plants are shallow rooted and 20% deep rooted. Based on the average leaching times, the average soil residence time for iodine with this assumption is 99 years. The resuspension option in

GENII was included using the built-in mass-loading model. All of the GENII parameters other than leaching were unmodified.

The largest contributor to the dose from iodine isotopes is from the dose to the thyroid, even when the effective dose equivalent is considered. The GENII code does not allow for variation in the dose conversion factor as a function of age. The National Radiation Protection Board computer code SR245 (Birchall and Hutton 1991) and ICRP 56 (ICRP 1990) both give the ^{129}I thyroid dose conversion factors for variously aged persons. Values in these two data sets are given to two significant figures. The NRPB dose conversion factors are slightly larger (more conservative). Given the variability and uncertainties in the biological and dosimetric parameters, using more than 1 significant figure for the purposes of this paper is not justified. However, both indicate that the ^{129}I thyroid dose conversion factor (Sv/Bq) for young persons is about a factor of 2 larger than that for adults. The unit release thyroid doses from GENII, which has a default value based on the dose conversion factor for adults, were increased by a factor of 2 for the ages from infant through 14 years.

The burnup history of the Hanford fuel has been tabulated in a spreadsheet by Jungfleisch (1992) and kindly made available by Heeb (1993). The Jungfleisch spreadsheet gives the burnup and tonnage discharged from each of the Hanford production reactors for each month from startup in 1944 until 1971 when N-Reactor was the only remaining operating reactor. The activity of ^{129}I discharged from each reactor each month was calculated from the fit and the discharged mass and summed over the reactors. The monthly totals were then summed to give annual values. None of the once-through Hanford production reactors operated after 1971. Discharges from N-Reactor after 1971 are not contained in the Jungfleisch spreadsheet. N-Reactor fuel was not reprocessed until the 1980's. From 1983 through 1986, about 3000 tons of N-reactor fuel irradiated to about 2000 Mwd/ton were reprocessed (Heeb 1993). This fuel contained about 185 GBq (5 Ci) of I-129. We assume that this fuel was reprocessed at a uniform rate and discharged ^{129}I into the air at the rate of 46.3 GBq/y (1.25 Ci/y) . A considerable amount of un reprocessed N-Reactor fuel is still in storage.

RESULTS

There was about 1900 Gbq (51 Ci) of ^{129}I generated in the fuel that was discharged from the production reactors and reprocessed between 1944 and 1986. The amounts of this ^{129}I used as a source term as a function of year of operation is shown in Figure 1. The annual intake from the activity in the environment in a given year from birth onwards from the ^{129}I discharged in that year and previous years was multiplied by the age dependent dose commitment per unit intake and summed over the years of the exposure period following the birth of the maximally exposed individual. The resulting cumulative dose as a function of birth year showed a maximum for people born in 1954/1955. There is very little difference in the cumulative doses using either the 1951 or 1957 food consumption data of Anderson, et al. (1993). The values based on the 1951 data are slightly larger.

Figure 2 shows the result using 1951 food consumption data.

The maximum thyroid dose accumulated up to 1995 of over 9,000 μSv (900 mrem) occurred for individuals born either in 1954 or 1955. Note that the people born in the year of maximum release are not the ones who accumulate the maximum cumulative dose. However, the maximum individuals were still children in the year of maximum release. The dominant pathway at all ages is the milk pathway. The balance of the dose is mostly distributed between leafy vegetables, other vegetables and cereals.

DISCUSSION

Rural individuals whose birth year was 1954 are estimated to have received the largest cumulative thyroid dose equivalent from the ongoing release of ^{129}I. This maximum dose, 9,000 μSv, is based on the assumption that all of the ^{129}I in the fuel dissolved in the chemical reprocessing plants from 1944 through 1986 was released to the air, a very conservative assumption. The ^{129}I thyroid dose to the average member of the population would be much smaller since most of the population around Hanford live at considerably greater distances from the point of release.

To put the cumulative dose from ^{129}I into perspective, the average annual natural background dose to soft tissue from both external and internal irradiation is about 720 μSv (NCRP 1987). Over the same 42 year period this dose rate would produce a cumulative soft tissue dose of about 30,000 μSv, or about 3 times more than the maximum dose estimated due to the total release of all of the ^{129}I produced in the discharged reactor fuel dissolved at Hanford.

Brauer, et al (1973) and Soldat (1976) estimated the thyroid dose due to ^{129}I based on measurements of concentrations in the environment and in food and water in the Richland, WA area. Soldat (1976) estimated that a maximally exposed adult would have a dose rate of 0.5 mrem/y (5 μSv/y) while Brauer, et al (1973) estimated 1.5 mrem/yr (15 μSv/y). For the maximum individual evaluated in the present study who was born in 1954 and exposed through 1995, the average annual dose equivalent rate to the thyroid is about 220 μSv (22 mrem) per year. Brauer's result is 15 times smaller than our upper limit maximum value.

The two most sensitive parameters in the calculation of the thyroid dose due to an air release of radioactive iodine are the direction and distance from the point of release to the location where food is grown and consumed by the exposed individual and the amount of and source of milk consumed. Richland is approximately twice the distance from the source as is Ringold. Thus, the airborne concentration at Richland due to a unit release to the air is much smaller than that at Ringold. The actual distribution between milk from cows fed stored feed and milk from cows on fresh pasture that is consumed by Richland residents is not known. However, there is no radioactive decay decrease of ^{129}I in stored feed. The doses calculated by Brauer, et al and by Soldat are based on measured radioiodine concentrations in milk.

Shleien (1994), also assuming that all production of ^{129}I was released, found somewhat smaller values than those found by us. In his case, the location of the maximum individual was in Richland at 24 km ESE of the source. Shleien did not specify the fraction of fresh pasture in the feed of the cows supplying milk, but as was mentioned above, this is a relatively unimportant point.

The considerably larger dose for a Ringold resident estimated in this study is not unreasonable considering the assumptions for parameter values and supports considering it a maximum value. This maximum dose is considerably smaller than the dose due to natural background and considerably larger than the maximum value estimated by Brauer (1973).

Acknowledgment

We are grateful to Calvin Heeb of the Battelle-Pacific Northwest Laboratories who generously made available to us the results of his computations and his reactor data files and to Bruce Napier of the Battelle-Pacific Northwest Laboratories for his assistance in applying the GENII code.

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Table 1
ORIGEN2 Prediction of ^{129}I Production

W_T MWd/ton	^{129}I g/slug
500	7.589E-3
1000	1.634E-2
1500	2.573E-2
2000	3.552E-2
2500	4.561E-2
3000	5.594E-2
3500	6.644E-2
4000	7.709E-2

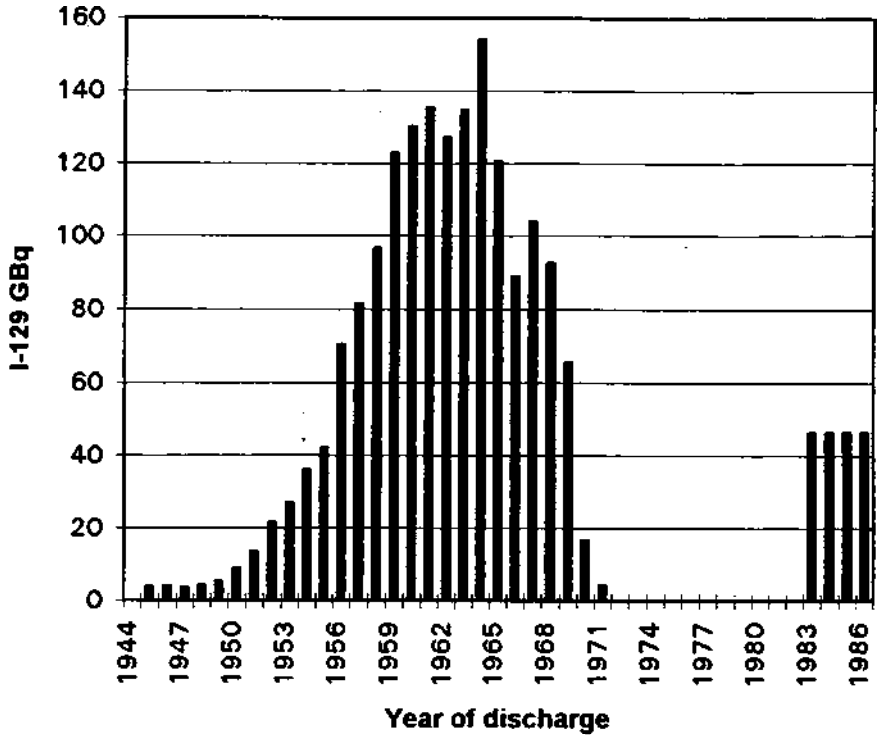

Figure 1
Maximum possible activity of ^{129}I discharged
to the air from fuel reprocessing at Hanford

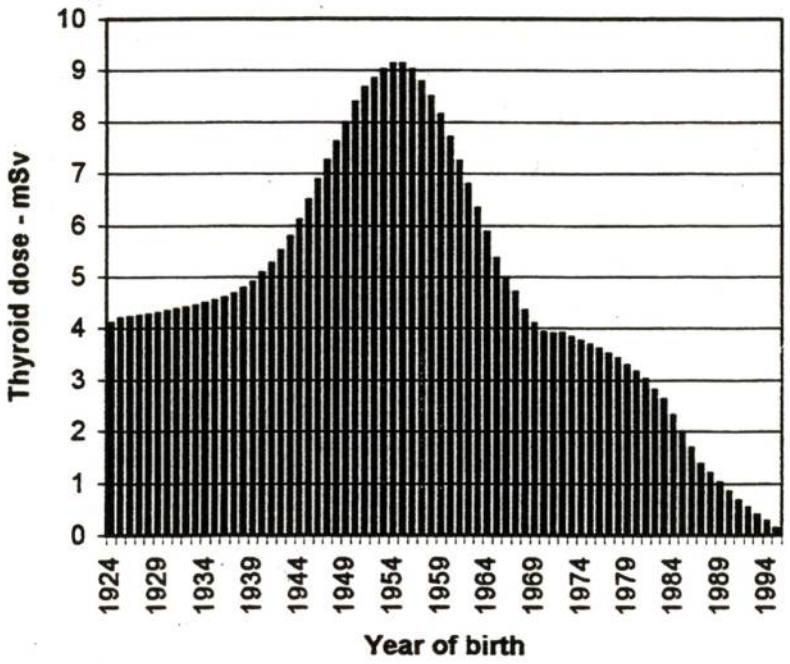

Figure 2
^{129}I lifetime cumulative dose by birth year for
maximum individual at 19 km East of 200E

APPENDIX C

BATTELLE REPORTS OF THE HEDRP

FOR THE SOURCE TERM

BATTELLE REPORTS OF THE HEDRP FOR THE SOURCE TERM

PNL-7177 HEDR Radionuclide sources and Radioactive Decay Figures Pertinent to the HEDR Project. C.M. Heeb March, 1991

PNL-7210 HEDR Fission-Product Iodine During Early Hanford-Site Operations: Its Production and Behavior During Fuel Processing, Off-Gas Treatment, and Release to the Atmosphere. L.L Burger, May, 1991.

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PNL-7253 HEDR I-131 in Irradiated Fuel at Time of Processing from December 1944 Through December 1947, C.M. Heeb, L.G. Morgan, March, 1991.

PNL-7717 HEDR Radionuclide Sources and Radioactive Decay Figures Pertinent to the HEDR Project, C.M. Heeb, May, 1991.

PNL-7868 HEDR Letter Report: References for Radioactive Releases to the Atmosphere from Hanford Operations, 1944-1957, R. B. Hall, November, 1991.

PNL-7869 HEDR Letter Report: References for Radioactive Releases to the Columbia River from Hanford Operations, 1944-1957, R.B. Hall, November, 1991.

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PNWD-2033 HEDR Vol.1 Iodine 131 Releases from the Hanford Site 1944 Through 1947, Vol.1-Text, C.M. Heeb, September, 1992.

PNWD-2033 HEDR Vol. 2 Iodine 131 Releases from the Hanford site 1944 Through 1947, Vol. 2- Data. C.M. Heeb, September, 1992.

PNWD-2161 HEDR Fuel-Element Failures in Hanford single-Pass Reactors, 1944-1971, S.P. Gydesen, July, 1993.

PNWD-2222 HEDR Radionuclide Releases to the Atmosphere from Hanford Operations, 1944-1971, C.M. Heeb, May, 1994.

PNWD-2223 HEDR Radionuclide Releases to the Columbia River from Hanford Operations, 1944-1971, C.M. Heeb, May, 1994.

PNWD-2254 HEDR Sources of Secondary Radionuclide Releases from Hanford Operations, C.M. Heeb, S.P. Gydesen, May, 1994.

APPENDIX D

MINUTES OF SOURCE TERM

SUBCOMMITTEE MEETINGS

SOURCE TERM
SUBCOMMITTEE REPORT

July 27, 1988

The Source Term Subcommittee inspected the data bases. FoxBASE, a dBASE type data base, is being developed by the source term subtask group of Battelle. The ASK SAM bibliography reference base is being developed by the Battelle technical library. We looked at the way the data base operated and we opened some modular communication to the data base.

The BNL subtask personnel briefed the subcommittee on their subtask responsibilities which included collecting time line information on the accidental releases, on reviewing documents for effluent data, and estimating iodine-131 releases based on the records of plant operations and processes. The iodine release estimates that were obtained or compared to some earlier release estimates, in comparison were found to be very good.

The inventory of isotopes, the amounts and identity of the isotopes that were in these early reactor fuels were analyzed. These are being analyzed by different codes to find the code that gives the best result. A code called "ORIGEN" has been determined to be the better of the codes which were examined. This code permits different isotope characteristics to be used in various isotope libraries so that the calculation can be directed very closely to the particular reactor. Each reactor class is somewhat different, and the are able to narrow the isotopics down to a class as well.

We discussed a model for the quality assurance of source term data in terms of how the data are characterized and classified and how one might go about reducing uncertainties and unknowns in these data.

We established direct lines of communication between he subcommittee and the subtask personnel so that technical questions could be addressed on an individual-to-individual basis to expedite the Panel's work in keeping track and directing the scientific work of the Battelle staff.

We addressed how the source term study interacts with the environmental transport question, and on the dose to the impacted populations through issues of specific life style, utilization of contaminated foods, and we showed that there would be a very close interaction between the various subcommittees to permit us to define a range of, or a level

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of releases below which, the contribution to the total dose to this population would be very small so that a lot of energy and effort wouldn't have to be spent in chasing something which is relatively unimportant rather than spending it on making sure that the important exposures, the important releases are correctly and accurately attended to.

SOURCE TERM
SUBCOMMITTEE REPORT

September 24, 1988

The committee met. Bruce Napier demonstrated the GENII code. The committee felt that this was an excellent code, a significant improvement on the state of the art, with the great advantage of being operable with a PC type of personal computer, menu driven with great flexibility and allows concentration of many isotopes, transport paths and pathways and organ doses. We were quite impressed by that effort.

Larry Morgan of Battelle reported on working progress to the group. They are continuing to compile accidental releases and expect to be finished the first quarter of 1989. He feels that all unplanned released have been documented, which, in my mind, raises an interesting question. That is, for the TSP to verify this may require enormous effort. It basically would require reproducing all of the effort that they have done. However, some spot checking may be certainly worth while to do. Members of the subcommittee perhaps could think about that for discussion at a later time.

He observed that progress on surveying documents is manpower limited because of the varied and extensive nature of the various tasks that the Battelle team is working on. Specifically , the assigned manpower for reviewing documents is limited due to limiting the rate of documents which can be cleared and classified, by which I mean sorted in terms of their content.

He reported that Cal Heeb has supplied Bruce Napier with input for reactor ORIGEN runs. He reported that the demographic data base is being revised to improve the field sizes and referencing capabilities, increasing the flexibility of the kinds of interrogations that the FOX BASE system will be able to use. All of his group's data base is also provided to Shirley Gydesen for incorporation through the summary Battelle data base at the Battelle library.

Their working group, Larry Morgan and his working group, will be examining how much of the classified data will contribute to the dose. That is, how significant will the set of releases which are classified be to the overall dose evaluation.

In the next six months, the working group will produce three documents. The first will be with Bruce Napier on selection of nuclides of significance and identification of facilities, processes, and emission control technologies which were in place in those days.

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The second will be a draft document which will be a compilation of accidental releases which will be submitted for TSP review. This is scheduled for May of 1989 and that will include data from classified source, if any such data can be determined. Up until now, they have not found any significant classified data relating to dosimetry.

The third document will be a description of the data base systems and the record format for the numerical emission data which are in the data base which will enable someone to extract specific numerical information.

Walt Haerer reported that they are still looking for good vegetation activity data and correlations with the source term and the meteorology to use as guidance to rationalize the early-on data where this correlation is either sparse or nonexistent. He didn't give a date they expected that effort to be completed. If he wants to comment on that particulate one, I didn't have it in my notes when he though that task of subtask would be finished.

Walt Haerer: The task is underway. What we're not certain about is the success of that task.

Maurice Robkin: One way or the other, you'll have some statement to make. I didn't recall what your goal was in terms of timing if, in fact, it was stated.

Walt Haerer: I had not stated one.

Maurice Robkin: continuing:

The subcommittee recommends that the TSP request a status report on the overall project at every TSP meeting which will summarize and update the results. The subcommittee also recommends that a study of all methods of analysis for vegetation be evaluated and calibrated so that the Panel can decide about vegetation as a continuing monitor for outside contamination. K. This recommendation relates to the report from the PNL working group that they are looking at the earliest set of consistent data of quality that they can define to do this sort of thing. It was moved and the subcommittee did pass a motion to recommend that to the TSP as an official subcommittee. That's all I have to report.

SOURCE TERM
SUBCOMMITTEE REPORT

January 19-21, 1989

Application of the recommendations from the Uncertainty and Sensitivity Workshop to the Source Term Task

"Art of the Possible"

1. What time period should be studied?

   Perhaps two time periods should be studied. The first time period would be when high quality data were available so that models and time independent parameters such as deposition velocities could be evaluated and validated well. The second would be the 1944-1947 period during which the largest releases of I-131 occurred.

   For initial dose estimation, use earliest period of highest releases. Obtain the time dependent parameters from these data. Make best estimate of doses possible at this stage recognizing that there may be wide uncertainties. Improve estimate and reduce uncertainties as project progresses. Therefore, source term data is needed for both time periods.

"Evaluate and Rank Models, Minimize Set of Models Useful for HEDR work."

1. Note: Although there are data and parameter uncertainties with respect to releases, they are less than those for any of the other parts of the dose calculation.

2. The number of candidate models is very limited. For example, for the radioisotope inventory, ORIGEN with a production reactor specific cross-section library would correspond to the industry standard. Release fractions uncertainties are probably expressible as simple range values.

"Better Characterize Necessary Parameters to Define 'Objectives"

1. The two main objectives are: i. Find the doses to individuals so that epidemiology can be done, and ii. Find the population average and maximum credible individual dose for any study population.

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2. The contribution of the Source Term Subtask to the objectives is straightforward. The number of parameters is limited.

    a. Inventory calculated with ORIGEN, the industry standard.
    b. Development of the production reactors cross-section library is reasonable straightforward. The theory is well developed and well validated from light water reactor work.
    c. The calculation is designed to follow all or nearly all of the isotopes in the fuel.
    d. Reaction cross-sections are known to within a few percent for most isotopes.
    e. Largest uncertainties are associated with sources, such as releases of activated film, that contribute much less to the early doses than the iodine releases.

"Probability Distribution is Preferred Description of Uncertainty"

1. PDF representation does not apply to source term for the major part of the releases. The isotope inventories and chemical processes are highly deterministic, but chemistry may not be well known for the earliest reprocessing, which may introduce some uncertainty into the releases. There will be some uncertainty in the activation isotopes source due to poorer knowledge of the complete mineral content of the water entering the reactors and the identification and the residence times of the elements trapped in the film that was built up in the cooling channels.

2. There will be some uncertainty in the inventory in the fuel slugs that burned on discharge from the reactors unless their irradiation histories are well documented.

    Overall conclusion: The source term uncertainties should not be a major contributor to the overall dose uncertainty.

<u>A meeting was held with the members of the PNL Source Term Subtask Group. The following items were reported to the TSP Source Term Subcommittee</u>

1. Programming changes have been made in the FOXBASE database code to increase keyword capacity. Some 450 references have been entered with about 20-30 more to be entered.

2. No development of the numerical data base has been done.

3. The WIMS multigroup transport theory code is being used to generate new cross-sections for the ORIGEN cross-section library for the production reactors.

4. There may be some data on I-131 releases for specific early dissolver runs to give handle on release fractions.

5. There are some good data on coolant channel film activation.

Items to be provided to the Source Term Subcommittee include

1. Details on how the iodine releases are being calculated.
2. Description of facilities and processes in use versus time.
3. Non classified data on release fractions and chemical form of releases.
4. Tabulation of how much source information has been generated.
5. Status report on the rebuilding of the ORIGEN library.
6. Identification of all parameters significant to the source term calculation over the entire time period of concern.
7. No longer classified production reactor parameters to be used in the source term computation.

TECHNICAL STEERING PANEL
RESEARCH DIRECTIVE

Directive Number:	**89-4**
Date:	**January 29, 1989**
Subject:	**ACTIVITY IN THE COLUMBIA RIVER FOLLOWING REACTOR PURGING**

Action to be Taken:

Documents cite "purging" of reactors to remove film deposited in pipes. This action was performed by sending a slug of diatomaceous earth or other materials through the cooling channels. The resultant purged material was emptied into the Columbia River and could have raised the level of activity in the River.

Battelle staff must investigate the effects of purging the production reactors and the resulting impact on radioactivity content in the Columbia River.

John E. Till, Ph.D.
Chairman, Technical Steering Panel

SOURCE TERM SUBCOMMITTEE REPORT

March 17, 1989

The Source Term Subcommittee had a technical working session with the Battelle Source Term group. We discussed the assembly that Battelle is making of the details of the early facilities. This is a historical collection. They are collecting the history of the processes, the materials, in some detail. The report on this tabulation will be submitted to the TSP by the end of March. They are collecting accidental release data which they will tabulate. There is work ongoing on the behavior of iodine in the processing facilities. They are tabulating the chemistry to describe the way iodine behaves in the process itself, and also the way it behaves in the air following a release through the atmosphere. The ORIGEN-II code, which is designed to calculate the evolution of the radioactive isotopes in the fuel, as the fuel is exposed in the reactor, is being evaluated. The cross section library, in particular, is being examined in order to determine whether a special library is needed for the production reactors. At present ORIGEN-II has available libraries describing the CANDU reactor and N-reactor. The N-reactor library is the closest to the production reactors. There are still some questions whether the results from an N-reactor calculation are accurate enough. What was suggested to Battelle is that a detailed multi-group calculation be made, comparing one of the typical production reactors to N-reactor to examine the neutron spectrum. Examination of that will give, I think, a much better picture as to whether it will be necessary to go through the full scale evaluation of a production reactor rather than using perhaps just the N-reactor library as it currently exists. The reason for this consideration is that the neutron spectrum is combined with the detailed multi-group cross-section library to produce a one group average cross-section value for the isotopes so that the evolution of isotopes during an irradiation can be followed.

The calculational spreadsheet model is being developed to evaluate burnup and to include an evaluation of the I-131 production for the 1944 to 1947 period. These calculations are based on the collected data estimates for the burnup, the power levels, and the time for each unit of fuel in the reactors as best as can be determined from individual historical records. The detailed spreadsheet is designed to follow each unit of fuel as it resides in the reactor, and to collect all those data forward in time to discharge through cooling and then through the dissolution.

The detailed spreadsheets have been compared to approximate methods, which are based on average quantities of burnup and cooling times and the amount of fuel dissolved. These have slightly different usages of the average values. One is based on averages

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obtained from the spreadsheet detail and the other is based on averages from the record's estimated averages. The two calculations that have been done indicate that the detailed spreadsheet result fails for almost all periods between these two approximate methods so that the approximate methods may in fact be adequate for calculating the inventory burnup coefficients, the times, the burnup, the mass, for use for iodine production estimation. There is some mention also of continuing work with source term data base, which is something that the source Term Group is maintaining, analogous to the other data bases but specifically for source term input data.

SOURCE TERM
SUBCOMMITTEE REPORT

May 20, 1989

Larry Morgan reported that the major Phase I activities will be completed by June 1. The data will then be delivered to the Transport Task by June 1.

There were a series of reports by the source term working group of Battelle, which are going to be published separately as well as included in the annual report. Some of these will note be published separately, but will be reported in the annual report as appendices.

Pete Jackson reported that the fuel element tracking data for the last half of 1945 to 1947 no longer exists. The spread sheet which runs '44 to mid '45 will be completed by June 1. There are a few railroad log sheets for the second half of '45. The proposal is to use the Anderson's annual tabulation data. I found out this morning that there are, in fact, weekly tabulations of data for 1946, which may be either primary or secondary data, but in any event usable data for forwarding to the transport sub-task.

Michelle Ballinger is doing the final stages for the internal review of the facilities history. She has written a very nice document which describes the early history of the Hanford project facilities. When that has gone through internal review, we will get a second draft, presumably by the end of May. She has also identified dates and quarterly release amounts for 1984-1986 for input to the atmospheric transport, I hesitate to use quality assurance, but at least the calculation/verification procedure that is being done by the atmospheric transport group.

Cal Heeb reported that there has been no particular progress on numerical data base since the last meeting, but they are working on the integration of all HEDR wide data bases into a single data base so that all of the information can be accessed in an efficient way.

He has also prepared an outline of the fuel heavy metal actinide inventories found in the irradiated fuel, and identified the most important activation isotopes and the important fission products. He did this by a decay scheme, so there is some information there.

They also ran the WIMS code, which is a multi-group transport theory code for calculation the actual neutron spectrum in the old production reactors to compare with N-reactor spectrum and what effect that would have on cross-sections and on inventories. The result

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was that the absorption cross-sections of most isotopes are affected by about a factor of 2, but for the fission products it really has very little effect, since the cross section doesn't have a big effect. There are a few which are affected. It does have a significant effect on the activation products by about a factor of 2. In fact the production reactor spectrum can be used in the ORIGEN code if it is felt, and I don't think it probably will be, significant for the fission product evolution. It is a separate calculation for the activation products. Probably this difference can be handled without having to do a great deal of rework with the ORIGEN library.

Lee Burger reported that there is as yet no hard data on iodine speciation in the dissolver and in atmospheric reactions for the iodine species. The report, when they get these into data, will be appended in the annual report.

Karen Wiemers is identifying the separation plants stack emission data, using Hanford documents, and documents prior to 1950, for which the first reliable information if T Plant in February of 1945, and B Plant in November of 1945. She is collecting raw data, which is direct instrument readings. She is collecting formatted data, which are readings which have been translated into amounts, that is curies or grams, et cetera, which were sent up the stack; calculated data, that is based on the amounts of fuel, of metal which were irradiated, the power levels they were irradiated at, and what the theoretical yields of the various isotopes would be; and then knowing something about the dissolution step in the release fraction and how much would go up the stack.

These data are estimated to be raw data for the 1944 period. That is all of the yearly data are raw data. In 1945 30 percent of the early data are raw data, 75 percent are formatted data, that is specific amounts, and 30 percent is in calculated form. These don't add up to 100 percent. These are just what kind of information is available by month throughout the year. Obviously there will be duplications. That is, some information will be in all three forms, some will be one form and some in another. In 1946 the breakdown was 60 percent as raw data, 30 percent as formatted, but 100 percent is calculated. So they calculated everything. In 1946 there are no raw data, 18 percent formatted, 50 percent calculated and there are data missing for some of the months. That is, some of the months have no information, some other months have more than one kind of information.

I reviewed the Green run and presented that report at the meeting. If I left something out, if one of the other members could add to that. Maybe I could just find out if I left anything out.

SOURCE TERM
SUBCOMMITTEE REPORT

July 22, 1989

The Source Term Subcommittee had discussions with PNL scientists to review the completion of the Phase I work. The production reactor, ORIGEN2 libraries for computation of fission product inventories, to get the iodine inventory were discussed. The ORIGEN2 code came with CANDU and Water Reactor libraries. An N reactor library was developed for comparison of cross sections. The N reactor spectrum cross sections were compared to CANDU reactor spectrum cross sections and the differences, generally were small for the most part except for a few isotopes. The production reactor spectra were computed by another code, called the WIMS code, and showed similarities to the N reactor spectrum with somewhat smaller fission flux. Since the iodine-131 production figures for N verso CANDU are similar, no advantage will be gained in developing ORIGEN2 libraries for the production reactors for Phase I.

Phase I source will be limited to iodine-131 for the air pathway and to measured isotopes for the river pathway. It's recommended that PNL evaluate other isotope contributions when the Phase I research is finished. This, of course, is part of what they will do. The estimate we had is approximately two man-months per nuclide. Effort will be required for evaluating the other isotopes.

The source term numerical data base will contain only early iodine-131 data during Phase I. Further input to the data base will occur in Phase II.

Karen Weimar has a draft report on the time line for existing separation plants stack emission data, which shows releases versus time, that is for the data that could be obtained. There have not been any correlations with projected releases based on calculated inventories and release fractions for times when the data overlap. The subcommittee feels that it would be of considerable value that, where the release measurements were made at a time when there is information based on the inventory and release fractions, these comparisons which provide independent check should be made.

The iodine chemical species study of Lee Berger is under PNL internal review. He has evaluated the species formed during fuel dissolving, processing, and atmospheric interactions and finds that elemental iodine dominates with about 80 percent, perhaps, of

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the total iodine; that methyl-iodide may contribute up to about 10 percent, and that particle bound iodine may represent between 1 and 10 percent. This is iodine absorbed on particles which are airborne and which may range in the 0.1 to 1 nanometer range.

Some confusion exists regarding the extent of independent iodine-131 data for the '44 to '47 period. Our initial impression was that there are production data for start up, that is 1944 to mid '45. From mid '45 to '47 there are no surviving production data. There are release data available from a report by Anderson, and we feel that more than a single datum is needed for reach aggregate period in order to justify setting the parameters of distribution for the iodine-131 source term. This may well be a misunderstanding in terms of what does or does not exist by way of data, and PNL will clarify this and respond by a memo to indicate the extent of independent iodine-131 data covering the Phase I time period.

SOURCE TERM
SUBCOMMITTEE REPORT

October 14, 1989

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1. The list of released isotopes still has not been received. Battelle has presented the list of dominant isotopes at this meeting. Battelle has committed to providing the TSP with a full list of released isotopes in early November, 1989.

2. Missing I-131 release data for '45-'47 period will be estimated by PNL staff from reactor irradiation histories. TSP Subcommittee members will interact with PNL staff on an individual basis to confirm procedure.

3. The question was raised of the effect of limiting source to a single point. This assumption is for Phase I based on closeness of major source stacks in 200E and 200W. Multiple sources will be addressed in Phase II.

4. Significance of using Kr85 dispersion data to test air dispersion model was addressed. Using a noble gas model for dilution with a particulate specific deposition velocity for the various forms of *-131 gives a conservative result for I-131. Effect of treating iodine chemistry will be addressed in Phase II.

5. The question of using a puff model (MESOI) for an extended release was addressed. Dr. Haerer pointed out that the MESOI code has been modified to account for time extension (:MESOI-LT).

6. The question of the impact of fuel element failures was discussed.. Apparent absence of rise during 1955 on fig 2.4 of PNL work plan raised question as to whether releases from fuel failures were measured. Graph gives Anderson Report of 1974 and site monitoring reports as sources. Anderson report is for atmospheric releases from separations plant and not relevant to the river. We had no data on hand from the monitoring reports, but since I-131 and gross beta are apparently combined into the total release, it would be difficult to see the rise due to fuel failure on the scale of the graph. A separate plot for gross beta alone may show the effect of fuel failure on gross beta in the river.

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7. Finally, the question was raised of why 1964-1966 was chosen for the river pathway when by 1966 3 reactors were shut down. It was pointed out that although the source term was somewhat smaller than, say , the 1962-1964 period, the monitoring data are better so that model testing is more reliably done. This is one of the main purposes of the Phase I part of the study.

SOURCE TERM
SUBCOMMITTEE REPORT

December 11, 1989

The Source Term Subcommittee met and discussed the items in the Source Term Subcommittee's annual report. Some additional items were brought up by members of the committee and will be included in that. As in the case of Dr. Klingeman's subcommittee, that will be submitted to Mary Lou Blazek for inclusion in the TSP's annual report.

The subject of defining possible activities and goals for Phase II was brought up. Discussion of various possibilities was held, and the decision was made that we should coordinate with Battelle's proposals and integrate the subcommittee, TSP view and Battelle's and resolve this for a conclusion at the next TSP meeting.

The issue of the mission basic data in the period from the mid-1945 to 1946 was addressed. It was resolved by observing that the Anderson Report notes, that is the transcription of the basic data that was made by Mr. Andersen of those notes, are available, so that although the original information has been destroyed as part of the pruning of old records, there are still enough notes available to recover details of the radiation histories of the fuel elements that were used in the interval of time from 1945 to 1947.

As part of this, the early data, the 1944,1945 data will provide some scaling factors, ratios of production, power levels, such that the information available in that period of late 1945 to 194 on power levels can be used to infer production, as they provide an independent check of the information being used for measurements.

The question of accidental releases and their importance was addressed. The resolution of this question was open, we have to examine the importance of various accidental releases and whether or not hey may be significant compared to the systemic releases fro particular nuclides which may be important, that may be distributed in a n important way by accidental releases. We had no information on this, and we will have to resolve this in the future.

Battelle will be at this meeting, meaning tomorrow and Wednesday, presenting some of their proposals for Phase II work. The subcommittee, although we discussed the possibilities, feel that we really cannot recommend anything at this stage, but would rather

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wait and integrate with the Battelle proposals and come to a conclusion at the next meeting.

It was brought up that information is needed on chemical and physical forms of many of the radionuclides, bedsides radioiodine, I order to evaluate what the potential distribution and transportation, and so forth, of those nuclides might be. This is an open issue for resolution later.

The question of whether or not it would be necessary for the ORIGEN II library to be developed explicitly for the production reactors was discussed.. The conclusion was that for fission products the existing libraries are sufficiently accurate. For activation products, it is a question as to the utility of up-dating the ORIGEN II library for activation products because there are enormous uncertainties in the amounts of activatable isotopes which are present in the river and in the film on the cooling channels. There is enough uncertainty that the improved calculation of ORIGEN II really isn't going to help the overall uncertainty since the basic amounts present are not well known.

The hot particle question was brought up and needs to be addressed. Not only the emission of particulate releases from the stacks which carried these radioisotopes, but also hot spot information where there might have been enhanced deposition at very specific locations around the environment, which may have resulted in locally enhanced doses. This topic needs to be addressed. That concludes my report.

JOINT REPORT OF THE SOURCE TERM AND ENVIRONMENTAL TRANSPORT SUBCOMMITTEES

February 16, 1990

Phase II planning involves issues of environmental transport and source term. These two topics are very intimately connected. The two subcommittees felt that it would be much more productive to hold both subcommittee meetings jointly. Therefore, the two subcommittees met jointly on Friday, February 16, 1990, to discuss several items of mutual concern. These included:

1. Declassification of documents
2. Phase I report
3. Phase II planning
4. Proposed atmospheric transport workshop

I. Declassification of Documents

The subcommittees noted with appreciation that the TSP request for declassification of documents was approved by Admiral Watkins and has expedited the Panel's work. There was some discussion about whether the 1960 cutoff date for declassification would present any difficulties for obtaining needed data for dose estimates after 1960. The subcommittees decided that considerable unclassified data are available from the later period. Therefore, any problems from accessing classified data would be addressed as they arise.

This concern over post-1960 classified data arose because the Phase II work will be based on monitoring data but source term information may be needed for independent validation of the results.

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II. Phase I Report

We learned that the draft reports currently being received and reviewed by the TSP will not be included bodily in the Phase I report. However, they will be included by reference. The Phase I report will have extensive supporting material in its appendices.

The subcommittees requested a detailed outline (e.g. an elaboration of the Table of Contents) for the Phase I report so that we can better anticipate the contents of that report. Walt Haerer agreed to our request. A detailed outline will be given to the Panel on Saturday (today) for its use.

The subcommittees discussed issues that could lead to delay in release of the Phase I report. These included the need for additional time to validate the source term and the atmospheric transport model, as well as issues that other subcommittees may identify. The question of the delay in the release of the Phase I report may be addressed by the full Panel. The subcommittees did not make any recommendation regarding delay of the Phase I report.

III. Phase II Planning

The subcommittees reviewed Walt Haerer's letter of January 22, 1990 to John Till in which Haerer proposed Phase II objectives. Each of the activities involving Source Term, Environmental Transport and Environmental Monitoring Data were discussed by the subcommittees. We understand that the letter proposed will later be expanded into a detailed workplan for TSP consideration. Rewording of several of the items for greater clarity is recommended.

Item 0301

Conduct Sensitivity Calculation Using ORIGEN2: This activity would generate neutron cross-section libraries for a few radionuclides to determine if Hanford Production Reactor libraries are needed for all key radionuclides. The need for this activity will be determined in Phase II.

After some discussion, this item was approved as appropriate for Phase II work.

Item 0302

Conduct Comparison of Original Data with Anderson's Data Used to Calculate Estimates of I-131 in Fuel Processed in Phase I Time Period: This comparison is needed during Phase II to determine the sensitivity of Anderson's source term estimates to assumptions concerning average cooling times and other parameters.

This activity also was recommended by the TSP (memo, December 11, 1989).

This item will be addressed at a work session between Robkin, Shleien, and the PNL Source Term Task in March. This work session will also address the question of the iodine inventories in the dissolved fuel.

Item 0303

Extend source term I-131 through 1955.

This items is recommended to provide input to the Thyroid Morbidity Study. These data are desired by the epidemiologists to correspond to the period of major exposure of the individuals evaluated in their study.

Item 0401

Atmosphere - Data Base Preparation: Meteorological data for the period 1944-1947 are currently being entered into the HEDR computer data base. These data will be needed during the Phase II to compare 1983-1987 data obtained from a dense grid to 1944-1947 data obtained from a sparse grid to determine the relative sensitivity of air concentrations to these two data sets.

After some discussion this item was approved as appropriate. The activity statement will need clarification when the workplan is developed. It was suggested that PNL reexamine the question whether other meteorological parameter will need to be considered, e.g. precipitation. Dr. Ramsdell pointed out that the existing model can handle precipitation, although precipitation is not now included.

Item 0402

Surface Water - Conceptual Model: The need for a detailed conceptual model of surface-water transport will be determined in Phase II. Actual development of a detailed conceptual model, if needed, will be postponed to Phase III.

In the discussion, a distinction was made between a preliminary conceptual model and a detailed conceptual model. A preliminary conceptual model will be completed by March 15. There may not be enough time to include this in the Phase I report if the report is released in April, but it will be included if the Phase I report is released in July. Instead of postponing a detailed conceptual model until Phase III, the subcommittees recommended that a more detailed conceptual model for the Phase I study area be prepared during Phase II. It was also recommended that a preliminary conceptual model that goes from Hanford to the ocean be developed during Phase II. A conceptual model describes the river processes for transporting and storing

radionuclides from various sources; this forms the basis for the mathematical model of the river pathway.

Item 0403

Atmosphere - Sensitivity Studies / Computer Model Development: The sensitivity of dose calculations to uncertainties in air concentration that are due to such factor as the selection to diffusion coefficients, spatial variation of stability, dry deposition, precipitation, surface roughness, meteorological station locations and procedures for calculating dry deposition and plume depletion will be determined during Phase II and could result in proposals to modify the model in Phase II or Phase III.

After discussion, this item was approved as appropriate. A distinction should be made between sensitivity analysis and model refinement, both of which should be included in this activity.

Item 0404

Surface Water - Mass Balance Routing Calculations: The results of the simple routing calculations of Phase I will be compared to Phase I monitoring data during Phase II to determine how much additional modeling might be required to provide calculated estimates where monitoring data are inadequate.

Item 0405

Surface Water - Sensitivity Study: The sensitivity of Phase I results to simplifying assumptions used in Phase I calculation will be determined in Phase II.

This item was approved as appropriate.

The Haerer letter of January 22, 1990 does not mention ground water transport. Discussion of ground water transport should be included at some point during the HEDR Project. However, no specific recommendation was made regarding ground water for Phase II.

The subcommittees discussed the relative merits of using source term data vs measurement data as input to surface water transport. The general discussion favored the use of measurement data, since the uncertainties associated with these are expected to be much less than those from source term data in the period after 1960.

Item 0501

Examine the Magnitude of the Effect of Phase I Assumptions on 1954 - 1947 Vegetation Concentrations: The sensitivity of Phase I dose estimates to simplifying assumption in reconstructed vegetation concentrations will be determined during Phase II. These simplifying assumptions will be modified in Phase III if necessary.

Item 0502

Determine Accuracy and Precision in Historical Radionuclide Measurements in Fish and Water: The sensitivity of surface-water pathway doses calculated as part of Phase I to estimated uncertainties in historical measurements of radionuclide concentrations in fish and water will be determined as necessary in Phase II.

Item 0505

Extended Terrestrial Media Data Base: During Phase II, the extent of terrestrial media data (space, time, media) needed in Phases III and IV will be determined.

Item 0506

Green Run Air Data: The suitability of "green run" air data will be determined during Phase II for use in later phases.

These were discussed briefly and approved as appropriate. However, some wording changes are needed for clarification of the proposed activities. For example, item 0501 refers to simplifying assumptions. These have been addressed in a report by Eva Mart but should be listed here as example.

IV. Proposed Atmospheric Transport Workshop

Dr. Murphy proposed holding a workshop on atmospheric transport and diffusion modeling be held in the near future. The purpose of this workshop would be to assess the strengths and weaknesses in the transport / diffusion models being used in the HEDRP and to identify the ways in which these models must be refined in subsequent phases of the project to ensure that the models will yield aporationally credible and scientifically defensible results. Participants should include "outside" experts on atmospheric transport / diffusion modeling members of the TSP, and Battelle staff members.

Suggestions for the workshop format included:

1. Invited papers on atmospheric modeling
2. Review of the Battelle model
3. Critique of the Battelle model
4. Diffusion
5. Dry deposition
6. Washout
7. Limitations of models and the reasons for limitations
8. Use of personal computer to demonstrate models
9. Possible use of Geographic Information System for illustration
10. To be held for experts but open to interested non-participants
11. Workshop summary to be prepared to communicate conclusions to rest of TSP at the non-expert level

SOURCE TERM
SUBCOMMITTEE REPORT

July 13, 1990

The Source Term Subcommittee discussed planning for Phase II. It is clear to the Subcommittee that organization of tasks is driven in what we can call reverse order. The first decision to be made is a TSP global decision as to what are the next doses to be added to the 1944-1947 iodine doses and the 1964-1966 water pathway doses. This decision will establish the expansion of the time periods, study area and nuclides.

In addition to the primary decision about the next set of doses, a list of other tasks and priorities were identified. In order of priority to be addressed in Phase II and future phases these are:

1. Continue search for important missing basic information, especially I-131 for the Phase I period. This is a large job of high priority that will continue throughout the Project.

2. Identify special projects at Hanford that may have released isotopes not currently being considered. Example, Tritium production facilities. This is a small Phase II job of high priority that needs to be done to identify if any significant effort needs to be assigned to special project releases.

3. Determine actual speciation of radioiodine emitted from stacks during Phase I. This is needed to factor into the issue of enhanced deposition and special weather conditions such as thunderstorms. This is a small job of high priority that can be done in Phase II.

4. Identify river releases and sort them by significance. This item includes evaluating significance of routine purging and river borne element activation. This item is a high priority job. The size of the job is unclear. An initial scoping of this should be done in Phase II. It may turn out to be a fairly small job that can be done with ORIGEN and some simple modeling of the operations at the reactors.

5. Prepare a definitive report on the importance of I-129 and other isotopes of iodine besides I-131. This is a project that the TSP can do as a small white paper. It can be done in the Phase II period but is of second priority insofar as the main thrust of the dosimetry is concerned.

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6. Identify particulate releases and resulting hot spots. This is a small Phase II item of second priority.

7. Identify discrete events in which atmospheric or water releases occurred. Example is the burning on the site of contaminated tumbleweed or other environmental materials. This is a small Phase II item of secondary priority.

8. Summarize the accidental release data, and evaluate the completeness of the data. The committee feels that accidental releases are of less importance than the routine releases. This is a item of lesser priority for later Phases of the Project.

9. Ground water is probably a smaller contributor to the doses than other pathways. It will have to be addressed, but probably in a later Phase. This issue will include the contribution to the ground water from the low-level activity in trenches, ponds, reverse wells, etc on a routine basis. It will also include accidental events such as major washouts from floods, etc. This is a lower priority activity for a later phase.

SOURCE TERM
SUBCOMMITTEE REPORT

October 12, 1990

The Subcommittee convened and almost immediately most of the members with other responsibilities had to adjourn and go into this larger group. I remained behind to discuss with members of the Battelle Source Term Task what some of the proposed Phase II priorities and tasks were, and then we also adjourned to that larger group. From that, after that group broke up, I reconvened with the Battelle people and we worked out a set of recommendations, which I am proposing as source term recommendations. Those will be addressed later, I believe, in terms of tasks, budget and deliverables.

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SOURCE TERM
SUBCOMMITTEE REPORT

January 11, 1991

The Source Term Subcommittee met yesterday. Cal Heeb of PNL discussed the current Phase II task plan, which includes revising Phase I documents and reconciling the Technical Steering Panel's comments and other comments. There are five documents from the source term tasks, three by Heeb, one by Ballinger and Hall on Hanford facilities, which has about 43 comments to resolve, which I plan to get wrapped up be the end of January for issuance in March. (There is) one by Lee Burger on iodine, with about 48 comments to resolve, with the same time frame.

The comments fall into three categories, and these are comments which relate to Phases II and III, editorial comments and technical comments for the authors to resolve immediately, and which they are working on now.

The three papers by Heeb include the ORIGEN uncertainties Report, which have 42 comments, a radioactive decay scheme paper, which has no comments, and the Phase I iodine numbers, which Heeb wrote with Larry Morgan.

The ongoing task for the source term, is the closure of the iodine-131 release for the years 1944 to 1947. Firstly, the closure is scheduled for April delivery, the work to be completed in January and February.

Secondly, the power levels for D, B and F reactors have been found by day, and we have those values actually from 1948. These data have been put onto a computer and Heeb has written a program to calculate the iodine-131 content at discharge for the 1944 to 1947 time frame for Phase I. The discharges will be adjusted for the effect of constant production, as we have discussed at previous TSP meetings. This correction factor is about a factor of 1.27 larger than the average power in the reactor, which results from the discharge practice of discharging at constant plutonium inventory rather than average time.

The cooling times to be used for the connection between the discharge values and the dissolving values will be based on fuel shipping records. Where those are inadequate, standard cooling times for the particular time period will be used.

It was observed that Van Ramsdell needs iodine discharge by day and hour to correlate with the weather, which is a time hourly variable, and for which is needed the best available

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information. To develop those data we will account for the residual metal from previous dissolving and a current dissolving, that is the so-called heel, and to track them accurately using the time of dissolving, the time it takes to dissolve, and so forth.

It was suggested that the dissolving pattern could be obtained from the xenon record from the Green Run report, that is, how the dissolving proceeds in a given dissolving run from start to end. It was discussed that the iodine release may not follow the xenon and may actually be split into two parts. In fact, there is some indication in the Green Run report that iodine is released first as a fairly large burst from the initial dissolving, and then a smaller burst from the second step in the processing.

If a time distribution can be established then a Monte Carlo sampling will be used to provide sample release values for correlation with the transport calculation.

Dr. Shleien raised the question of $90,000.00 for the source term in Phase I. It was not clear how that money was divided. It was actually divided into three parts, iodine closure gets about $29,000.00, the remainder divided between the evaluation of airborne radionuclides, not iodine, released during the Phase I time period and waterborne radionuclides released during 1944 to 1957 time period.

I misstated, the airborne is 1944 to 1957, not iodine, excuse me.

The transport task: Van Ramsdell would like 100 realizations or samples of release generated from the iodine data by the Monte Carlo sampling for the 1944 to 1947 period.

During the evaluation of the releases, no isotopes are going to be thrown away or discarded. We will collect all data, including many isotopes which are of public interest, even if they are not on the dominant radionuclide lists.

The deliverable will be a bibliography of nuclide data for these other two pathways, taking into account that the handling of river water through the reactor changed over time from raw water to treated water, with changes in the chemicals, and so forth.

Some of these data may be classified, and will have to be declassified, although Shirley Gydesen noted that there are considerable data in the public domain of river measurements, and those of course are available.

The main problem in doing the waterborne evaluation is evaluating the hold-up time of the elements in the reactor. Nuclides can be classified into those that plate out onto the film and those which pass through without plating out. Those that passed through can be evaluated relatively straightforwardly. The ones which plate out present a problem, in that it is not easy to estimate how long a given activable radionuclide sits in the film exposed to the neutron flux before it is purged or beaks loose and comes out into the river.

It was suggested that an evaluation for this might be obtained by measuring influent and effluent concentrations. Influent concentrations of parent isotopes and effluents of radioisotopes that are produced by irradiation of those parent nuclides. Dr. Davis suggested that for straight through flow bromine and boron might be monitored.

The solution of the purge problem is not clear at this point, and will have to be evaluated in terms of how well it can be done if at all.

The work is proposed to be done by April, with a draft document by July.

Dr. Klingeman raised the question of river borne hot particles from air source, how would one go about calculating river hot particles, how would one identify a hot particle. There was some discussion of the problems relating to the hot particles in the river.
Dr. Davis raised the question on the availability of archived early samples from the river, and discussion was held. It was suggested that there may be samples archived at other locations, for example at the University of Washington at the Laboratory for Radiation Ecology and this point will be checked.

Dr. Shleien introduced a new issue related to dose cutoff, and after some considerable discussion, a suggestion was \made at perhaps a 500 millirem total thyroid dose for the 1944 to 1952 period for infants, with a 50 millirem limit for any single year established, based on the observation from animal data that infants are perhaps twice as sensitive regarding thyroid function effects due to iodine exposure. The 90 millirem per year adult dose might imply 50 millirem approximately for the infant.

This discussion indicates that this is really a TSP question, and the Source Term Subcommittee agreed to at least bring it up at this open meeting for consideration either now or at some later time.

There is some need for a summary paper perhaps by Dr. Shleien and Dr. Kopecky to address this question about what are the issues and requirements for cutoff and dose information. There are different needs for the purposes of the thyroid disease study and for the purposes of HEDR and to address this question, perhaps we need some input from both perspectives.

SOURCE TERM
SUBCOMMITTEE REPORT

April 4, 1991

The Source Term Subcommittee met on Thursday morning. The first item was review and discussion of the draft of the Source Term Subcommittee annual report. Dr. Caldwell raised the question of including the details of the Subcommittee recommendations to the TSP It was decided that since all members of the Panel contributed to the final budget recommendations, including source term work, that the final set of recommendations were more relevant. Dr. Davis raised question of including detailed list of Subcommittee members who made public presentations in 1990. It was pointed out that the complete list of public presentations will be made in the report of the Communications Subcommittee and that it would be redundant to repeat the subset of Subcommittee members in the Subcommittee annual report. It was moved and seconded that the draft annual report of the Subcommittee be accepted as presented. Motion passed unanimously.

Dr. Davis raised the issue of completeness; that is, of not overlooking any release however small for the purpose of establishing that nothing has been overlooked insofar as a release went. Of course, many releases are insignificant with respect to dose, but these should be identified as part of the complete record. Robkin and Heeb are preparing a proposal to address this issue.

Dr. Davis introduced the idea of using environmental levels of I-129 as an indicator of the bounds of the I-131 deposition. It was agreed that the idea was worth pursuing. It was noted that the correction for world wide fallout must be made. It was also pointed out that considerable data on fallout levels exists and can be used.

Mr. Cal Heeb of PNL reported on PNL Source Term work. His presentation was divided into ongoing work and supplemental work.

<u>Ongoing Work</u>

1. The definition of the point of consideration for the source term is the point of loss of engineering control, that is the point at which the radioactivity is released into the accessible environment. There is some ambiguity about this definition for such cases as the surface storage facilities for low level wastes. These are the ponds, trenches, cribs, and drains that received low-level liquid wastes. These facilities were intended to store activity, but to allow it to slowly percolated into the sorptive desert soil. These

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facilities represent a source to both surface and ground water and need to be considered on that basis. Although there is administrative control over these surface facilities, they have an immediate interface with the environment.

2. Iodine closure. The initial iodine release study covers the period from 1944-1947. Closure will include establishing the documentation basis for the data used for the I-131 releases during this early period. The data now available include the so-called Anderson report of annual averages of I-131 releases. This is based on notebooks originally produced by McDonald. Mr. McDonald is still alive, and contact with him has been made by Cal Heeb. The extent to which he can recall his methodology for calculating the cooling times for the fuel is still open.

   There are documents from which correlation can be made between fuel discharges and chemical processing plant dissolver runs. This will give an independent check of the cooling times and improved the quality of the I-131 release data.

   The date for closure is June, 1991.

3. Airborne releases other than I-131. For these other releases, the time period to be considered is 1944-1957. The period is selected based on the publishing of the Annual Environmental Reports which began in 1958. The current effort for the remaining airborne releases is to establish the basis for the documentation for these other releases.

4. Waterborne releases. Time period is also 1944-1957 for the same reason as the airborne release period. For both the airborne and waterborne releases, the task will provide a reference data base for the 1944-1957 releases. Data quality will be evaluated. The feasibility of reconstructing the source term will be evaluated. If no data exist, calculation will be done for specific isotopes.

   The earliest data are gross beta measurements. These do not give isotopic distributions and are of limited value for dosimetry. For this period, we will have to rely on calculations based on analysis of the facilities for the releases of individual isotopes. The gross beta data can serve as a corroboration.

   For information about element holdup times in the reactor, such data as results from the KR reactor test loop will be used. Work on examining the bases for documentation of the airborne and waterborne releases for 1944-1947 began in March. Results will be reported in August in two HEDR draft documents.

Supplemental Task

1. Develop a general source term release model to incorporate uncertainties in source term input to revised dose model. A major output from the work will be a set of I-131 release files that give the amount released by day and hour. These files will be generated by selection from statistical distribution for the various parameters that go into the release. There will be about 100 such files produced to provide input to the environmental dispersion model. The source file model will be applicable to any nuclide and release medium. The input files are scheduled to be ready for use by the revised dose model during September, 1991.

2. Dick Gilbert raised the question of the search strategy for documents and data retrieval. He pointed out that any proposed search strategy needs TSP approval. He discussed surveying researchers regarding the success of their search strategies. He agreed to report to the Source Term Subcommittee at the July meeting on Search Strategies and also on establishing defensible probability distributions for source term data files.

SOURCE TERM
SUBCOMMITTEE REPORT

October 10, 1991

The subcommittee met on Thursday, October 10.

Cal Heeb reviewed the funding and task plan for FY-92 and FY-93. The current schedule for reports is:

> For the I-131 releases from 1944-1947, the report is scheduled for November, 1991. Letter reports of references for airborne releases for the period 1944-1947 and for waterborne releases are due in November, 1991.

The Source Term Release model coding is complete.

Data are now in hand for the early I releases. The data cover 1943 individual dissolver cuts at B and T plants. The mass of discharged fuel and dissolved fuel have been compared by two independent references. Comparison of the HEDR estimates with data in the reports FTS-71 on the metal history and HWN-1991, the monthly summaries of dissolver runs agree to within 1%. Data show 1213 tons of metal dissolved in B plant and 927 tons in T plant. Data are available from daily reactor operations reports for the period 1944-1947 and from the 200 area weekly reports.

A reference to an experimental determination of the iodine release fraction based on sampling the iodine left in the dissolver solution at the end of dissolution has been found. Although the details of the experiment have not been given, including such things as the assumed yield, power lever, analysis method, etc., these data are being taken as the most reliable estimates of the release fraction. Certainly, measuring the residual iodine should give the most accurate estimate of the release fraction so long as the initial inventory is well known.

The assumptions used for the iodine inventory for the HEDR project are:

Energy per fission:	202 MeV
Fission yield:	0.0289
Half-life of I-131:	8.04 d.

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The cooling times used come from records of time of pile shutdown and date of fuel dissolution.

The historical record of the release factors have ranged from around 0.5 or less to 0.9. The two reports mentioning the measurement of residual iodine in the dissolver solution indicates 86% in one paper and 85% in the other. Other reports indicate that the release during the balance of processing ranges between 5% and 10%. Other reports also indicate that stack drainage recovers up to 5% of the iodine evolved from the dissolver. This iodine is dissolved in stack condensate and goes to liquid waste.

Cal proposes to use a histogram of probabilities of iodine release with a uniform probability between 82 and 88 % containing a 2 sigma band, and with a uniform one sigma band on each side of width 3% (79-82% and 88-91%). These bands contain approximately 95% of the total probability. The remaining 5% is uniformly distributed below 79%. For the balance of processing release, a central value of 7.5% is selected and for stack drainage a central value of 2.5% is selected.

Heeb described a new approach for finding the detailed time behavior of the releases to correlate with the atmospheric model. This approach is based on noting that initially the reactors start with fresh fuel. The highest power tubes reach the target exposure first and are discharged. This fuel is replaced with new fuel. The next discharge comes from fuel exposed to target burnup at lower power. This fuel has less iodine inventory due to its lower power. This fuel is also replaced with fresh unirradiated fuel. This process continues, with the power level of the discharged fuel continually decreasing, hence the iodine inventory. Then the first replaced fuel comes due. This fuel was again irradiated at the highest power and has the highest iodine content, so inventory jumps. The interaction of discharge of old, low power fuel and new high power fuel causes the inventory to oscillate from one discharge to another causing a distinct sawtooth in time inventory effect. The new reactors do not reach an equilibrium pattern of iodine inventory at discharge for over 3 years, or clear through the end of the Phase I time period. Cal is coding the irradiation histories to provide a time correlated discharge inventory for input to the atmospheric transport model. Since the exact time variation of the meteorological conditions is so important, the details of the variation of the iodine inventory is crucial. His efforts will result in an I-131 release which will be the best possible we can expect from the available data and will result in the iodine source term having a minimal contribution to the overall dose uncertainty.

It is appropriate at this time to commend Cal Heeb for an impressive piece of work in this instance and for outstanding work in general and to express our appreciation to him and to PNL for his being the Source Term Task Leader.

SOURCE TERM
SUBCOMMITTEE REPORT

April 23, 1992

Presentations were made by Cal Heeb and Dick Perkins.

Cal Heeb summarized the status of the Phase I iodine-131 work. He described the data sets that have been found. The source Term task now has logs of dissolving time-of-day for 314 runs from a record set of 1981 dissolving cuts. There have been 17 measured profiles of the evolution pattern of iodine with time after the start of dissolving. These data have been used to generate 100 realizations of iodine release for input to the atmospheric dispersion model. Data quality has been evaluated based on 2 to 3 independent sources of information; for example comparison of burnup from the power histories to the discharge record. The draft report is due in mid-may to PNL internal review. Projected release to the TSP for review is the end of June.

The team of Dick Perkins, Chuck Thomas, and Bob Hall were introduced. These are PNL staff, both active and retired-part-timers who will be working on the water activity evaluation. Dick Perkins has been doing radiochemistry on water since 1952, Bob Hall is an old-timer on the cooling systems. He is now retired and working part time on the project.

Perkins gave a complete review of the history of the measurements of water activity from the beginning of the project through the mid 1960's. From mid 47 to mid 57, total beta activity data in the water are available. Isotopics were determined by radiochemical separations and identification by chemistry and half-life. After 1957 specific nuclides were measured by radiochemistry and by gamma spectrometry as those instruments become available.

Based on the flow patterns of the cooling water from the reactor exit to the retention basins to the river, the average holdup time was estimated to be 4 hours. Radioactivity originated in irradiation of elements carried through the reactor in the cooling water, in elements adsorbed onto the corrosion film of the flow channels and from fuel ruptures. Most of the activity appears to be due to hold up in the film. Effective irradiation time for many elements is many times longer than the less than one second time for the direct flow through the cooling channels of the reactors. There are some data available on film composition from a study done on a process tube in KE reactor.

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The research plan is to use measurement data to determine the ratio of various nuclides to total beta in the 47-57 time period extrapolated back to 4 hours post discharge from the reactor and use these ratios to extrapolate back to the pre-1947 period when there were only total beta data available. Corrections will have to be made for isotopes which do not decay by beta emission but by electron capture and so did not contribute to total beta measurements.

The isotope to total beta ratios that are known show considerable variation through the years and seasonally within a year. The cause of the variability will be investigated. Some may be due to variation in input to the Columbia from tributaries such as the Spokane river. Other possible reference activity for the ratio will be examined.

In addition to the activity carried in water, gasses such as tritium C-14, and Ar-41 will be looked at. Tramp uranium generated activity such as neptunium-239 will be accounted for.

The water activity study is off to a good start with very senior experienced PNL personnel carrying out the work.

SOURCE TERM
SUBCOMMITTEE REPORT

July 16, 1992

The Source Term Subcommittee was called to order at 10:10 a.m. Committee members present were Pat McGavran, Ken Kopecky, and Glyn Caldwell. Other TSP members present included Pete Klingeman, Stan Davis, Norma Jean Germond, Mary Lou Blazek, and John Till.

Cal Heeb reviewed the forthcoming final report on the iodine releases for the Phase I time period. He showed the proposed Table of Contents and discussed the items. The chapters will include:

1. Method of calculating I-131

2. Description of the handling of the spent fuel

3. Description of the dissolving schedule at the T and B processing plants

4. Description of how the iodine content of the dissolver runs were determined.

5. Description of haw the release fractions were determined.

6. Description of the reactor model used to estimate the power levels that each of the batches of discharged fuel were operated at. Description includes the validation method in which the projected burnup of discharged fuel batches were compared to recorded values.

7. Description of source term release model giving the hourly releases of I-131 and how the 100 realizations used for input to the atmospheric model were obtained. Central value results are also given for comparison to previously estimated release values such as are in the Anderson report and the preliminary Phase I report.

8. Appendices will contain details of:
   reactor model
   statistical model
   power history of reactors
   data source information (references)

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table of dissolving amounts
description of B and T plant operations
ranges of stochastic results

Heeb pointed out that of the 1980 dissolver cuts considered, only 300 have enough information recorded to allow defining the exact release time. The remainder can only be defined to within a shift in a day or to a day.

The schedule for the final report was given as:

Concepts and Analysis Department review 7/27-7/31
After configuration management release of the code, report will go the internal peer review--8/14-9/2
Then to document clearance: estimated by 9/18
Then to external peer review: 9/18-?
Estimate of release to TSP by 10/30.

The question was raised about speeding up the process to release the document to the TSP in time for the October meeting. Cal Heeb, Mark Freshley, and Dill Shipler will evaluate question and respond to M. Robkin by Friday morning, 7/17.

Dick Perkins gave a report on the progress of the estimation of the radionuclide released to the Columbia River. The task objective was defined as: Develop a basis for estimating the releases of radionuclides to the Columbia river from the Hanford reactors. He described the water treatment plant where low concentrations of sulfuric acid, alum, sodium dichromate and separan were added to condition the water and remove suspended particles. After exposure in the reactor , the water was discharged into a holding basin for decay of short-lived activity. After a holdup of up to about 4 hours, the water was discharged into the river.

The once through reactors operated from late 1944 to mid 1971 when the last one, KE, was shut down. The issues that are being considered are:

Activation of natural and added trace elements in process water
Corrosion and dissolution of aluminum surfaces
Sorption of trace elements in process water onto the corrosion film where they are held up and activated considerably more than the direct flow through would imply.

Considerations include:

Neutron fluxes
Composition of materials
Concentration of parent elements

Efficiency of the water treatment and addition of elements by treatment.
Fuel element ruptures
Purging.

The radionuclides of concern include P-32, Np-239, Zn-65, As-76 as high priority elements, Mn-56, Na-24, Sc-46, Cr-51 as medium priority elements, and other elements including I-131, Cu-64, Sr-90, Co-60, Te-132, H-3, Cs-137.

It was pointed out that from Sept. 44 to mid-47 there were no continuous measurements of beta activity in the water.

From mid-47 to August 57 there is total beta activity data
From 55 to 57 there are some individual nuclide data.
(Actually there are some individual data back to 1950.)
From 57 to end there are individual nuclide data.

The proposed plan is to determine the ratio of each nuclide to total beta at four hours post discharge from the reactor and use the ratio as a guide to extrapolate to times when no individual nuclide data are available. Allowing for variations in holdup time from reactor discharge to release to the river, estimate release rates of nuclides to river.

A description was given of the parent elements introduced from water processing compared to those native to the river. Consideration of the measured activity in the water indicates that elements are held up since rate of discharge of activity was much greater for many isotopes than can be accounted for by straight through flow with the cooling water. Hold up times estimated to range from a few seconds to up to 26000 sec (for Sc-45). The inventory of nuclides on the corrosion film ranged from a fraction of a days release of a nuclide to 110 days worth of release.

A decay curve for 100H effluent water taken on Sept. 25, 1952 was shown indicating a dominant early decay of around 2 hours. Implication is that Mn-56 is a dominant contributor to the activity. A table of experimental holdup times showed that values ranged from 1 to 5 hours. Jim Thomas commented that he has data showing that times as low as 20 minutes may have occurred in some cases.

Calculation of the ratio of average gross beta to power level of the reactors indicates that the ratio increased with power level. Regression of the trend with power indicates about a linear increase in the ratio of beta activity to power. The reasons have not yet been determined. Consideration will be given on the effect of temperature on the sorption and activation process.

It has been suggested that using Na-24 as a basis for ratioing nuclides rather than total beta may give a more consistent results for extrapolating nuclide concentrations. This is being investigated. Present results indicate that both methods of ratioing give highly variable results.

One problem with beta measurements is that not all activity decays by beta emission so that total beta does not account for some activity. The counting preparation for water samples involves heating samples, so volatiles can be lost. Volatilization is a function of element and compound form. The question of counting efficiency for each nuclide thus needs to be addressed.

SOURCE TERM
SUBCOMMITTEE REPORT

October 8, 1992

M.A. Robkin presented a report on his study of the discharges of radioactivity in the cooling water used in the Hanford Production reactors.

The findings were summarized as

1. Data for the entire operations period was examined. The data set is not complete, but does span the period. Data include both specific daily curie discharges for total beta and for specific isotopes. Monthly average curie/day releases were obtained from reports which tabulated values based on extrapolating from two to four daily release values. These reports indicate that extrapolation was done on the basis of ratioing from the operating power for the day of the measurement to the monthly average power. A simple average of the few daily measurements was made for each month.

2. The data show a great deal of variation for a given reactor and a given isotope.

3. The total beta curies/day was compared to the sum of the individual isotopes for each reporting period which included some specific daily average values as well as a large number of monthly average values. The comparison showed a great deal of fluctuation with the gross beta values sometimes larger and sometimes smaller than the sum of the betas. The average percentage discrepancy between the gross beta and the sum of the isotopes was not extreme, but the large month to month variation resulted in a large coefficient of variation. The ratios of individual isotopes to total beta and individual isotopes to Mn-56 were calculated for each monthly reporting period. Mn-56 was chosen because its parent, Mn-55, has an activation cross-section that depends the least on higher energy neutrons compared to the isotopes reported. For a given isotope and reactor, these data also show large variation. The overall average values of these ratios were reasonably in agreement from reactor to reactor, but the coefficients of variation, the ratio of standard deviation to average value was large, ranging from a modest 20-30% to several hundred percent. Cal Heeb showed data indicating that for selected isotopes, for example Na-24, the variation in the discharge per megawatt-day ranges around a factor of ten.

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4. A simple model of the activation process was presented. Assuming that the adsorption of parent isotope is rapid and the inventory on the film comes to equilibrium quickly compared to the rate of activation, the activation generated and discharged from the reactor can be represented by a simple two compartment model. The model shows that activation depends on two important parameters, the energy dependent activation cross-section for a parent isotope and on the effective holdup time in the reactor.

   The energy dependence of the reaction cross-section varies from parent isotope to parent isotope, so that even for the simplest case, it is not possible to calculate a fixed activity ratio based on only on activation by low energy neutrons. If this were the only problem, it would be reasonable straightforward to calculate activation ratios based on cross-section information and neutron spectra. However, the activation is also a strong function of the holdup time in the reactor.

   The holdup time is determined by the dynamics of deposition and release of a parent nuclide in the reactor cooling channel surface film. Earlier modeling efforts by Perkins, Silker and others showed that for a simple activation model, the effective holdup times range from 3.8 seconds for sodium to 2300 seconds for phosphorus to 26,000 seconds for Scandium. In addition, for at least one isotope, As-76, four separate and distinct holdup times were identified, implying at least 4 holdup compartments. The holdup time of most isotopes is not known and has a significant effect on activation.

   The tabulated discharge data for all 8 production reactors were put into a set of spreadsheets. A complete set has been given to the PNL source term task leader. The simple model used to identify the important parameters and a somewhat reduced spreadsheet giving data on the isotope ratios, the activity per megawatt of power, the comparison of total beta to the sum of individual isotopes, and the monthly average curies/day released has been given for comment to several reviewers. This report can be made available to those interested. The amount of data processed is preliminary. There are other relationships that can be profitable explored to see if there are any guides that can be used to generate release data for isotopes and/or dates for which no information has yet been found.

In the discussion, Bruce Napier pointed out that the maximum time interval for averaging discharge data should be one month. Wally Walters supported this comment by pointing out that river conditions can significantly vary over this sort of time interval.

A question was raised about the source of fission products in the effluent. These most likely come from dissolved uranium in the river water or tramp uranium on the fuel element surfaces.

We are in a position with respect to the release of radioisotopes in the reactor effluent rather like the position we were in with respect to I-131 at the end of Phase I. There is a lot more work to be done and activation relationships to be explored. With further work, some useful relationships may be developed that will permit interpolating the releases in the unreported time periods or extrapolating the releases of unreported isotopes of interest.

SOURCE TERM
SUBCOMMITTEE REPORT

January 7, 1993

The Source Term Subcommittee met at 10:15 on Thursday, January 7 to discuss the selection of radionuclides to be included in the air pathway and the water pathway.

Cal Heeb briefed the Committee on the most recent evaluation of the dominant radionuclides by Bruce Napier. I-131 contributes nearly all of the dose contribution to the cumulated dose from 1944. The next most important element is ruthenium which contributes a small percentage of the total.

Robkin pointed out that the dose cumulated from 1944 is not of interest to a resident who moved into the affected area later, say after 1947. The question need to be addressed as to whether their cumulative doses are above the dose decision level and whether other elements now need to be included to evaluate the cumulated dose.

Napier discussed a summary from the scoping study showing the history of the I-131 contribution to the annual effective dose equivalent for an infant born in that year who drank milk from a backyard cow. Up to 1962, I-131 is still the dominant isotope. In 1955, the contribution from I-131 fell to about 50% with the balance due to two ruthenium isotopes. However, the effective dose equivalent for the iodine from that year was only 1.5 millirem. He showed that the scoping study indicated that the annual dose to an infant falls off considerably as time goes on. Infants born in later years get a much smaller dose. By the 1950's, an individual who was born in a year and remained an infant until 1972 would accumulate only about 1 rem. the accumulated dose would be smaller for a real individual as he ages.

By 1966, the annual dose has fallen to a fraction of a rem contributed by the other 12 nuclides considered since, by then, there was not much iodine released. The variable performance of the silver reactors was addressed by assuming an effective I-131 release fraction of 1%. This value was an upper limit of values consistent with the recorded environmental data. Noble gases were assumed to have 100% release.

The overall release pattern used was

Noble gases, 100% for all times
I-131, 90% until 1949
1% after except for 10% in 1951 when the silver reactor failures occurred
Other isotopes, 1 part in 10 million.

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For the REDOX releases, the assumption was.
Initially, ruthenium release about 1 part in 10,000
After operations stabilized, release 1 part in 100,000
Other fission products release fractions much smaller

For the other reprocessing plants, the sand filters kept the releases well controlled.

Ruthenium releases included particulates, assumed to be very small size to maximize dosimetry.

No dosimetry done yet with releases from the reactors.

Davis brought up the question of Ar-41 releases either from argon contamination of the helium cover gas or dissolved in the cooling water. Heeb pointed out that argon activity due to gas activation would not have been a significant problem but that dissolved argon has yet to be addressed. Perkins pointed out that fission gasses from irradiation of tramp uranium or fuel leaks would have been released and need to be evaluated.

Heeb discussed the water borne source term. He predicted that lack of data may present a difficulty in evaluation. For the early time period only gross beta data are available. It has not been possible to extract specific isotope distributions from the gross beta data. The effort is currently directed in trying to use the detailed isotopic information from later years to backcast the details of the isotopics in the water. In addition to the difficulties in obtaining a usable algorithm to do this, there are the difficulties due the fact that the physical conditions of the cooling water in the reactors in the early years were considerable different than in the 1960's. As a result, parameters such as holdup times would be much different. The effective holdup time is one of the critical parameters of the water activation, perhaps the most important. Other parameters are flux spectrum dependent and would be somewhat different for a production reactor in its early days. The variable holdup times in the retention basins will have to be evaluated. The range of holdup times is important only for activities with relatively short half-lives. Even for isotopes with half-lives of the order of half a day or so, the effect of holdup time variation will be relatively small.

Napier commented that the list of significant water borne isotopes previously prepared has not changed. The water pathway effective dose equivalent estimate was 5-10 mrem for most local residents. For heavy fish eaters, the dose might go up to 100 mrem. For people who ate a kilogram per day of resident fish per day, the dose might go to 1 rem.

Heeb pointed out that the Source Term Task must report results to the Transport Group by June. It will be necessary to have a rapid decision on the collection of water borne

isotopes as soon as possible. Napier pointed out that there are no current scoping studies done for the water pathway. The Phase I results and calculations done last fall are available. Nuclides other than those evaluated then are not in hand.

Klingeman pointed out that those nuclides that stay in solution in the river need to be addressed since those are the ones that will be significant for biological uptake as well as sediment bound nuclides that are taken up by benthic organisms Perkins also pointed out that sediment bound nuclides in the river will become soluble when they reach salt water.

Napier will try to provide a scoping study in time for the February Budget meeting in Seattle to give the Source Term Subcommittee enough information to make a recommendation about the set of nuclides to be considered for the river pathway. There will be about 5 nuclides of importance. There will be some others of lesser importance for which other considerations may affect the selection. It was agreed to delay decision on a recommendation for the water borne nuclides until the February meeting.

Discussion returned to the question of the air releases. After some discussion, Shleien proposed a set of isotopes be recommended to the TSP as the set selected for evaluation of the contribution to the air pathway dose. These were I-131, Ru-103, Ru-106, Pu-239, I-129, C-14, H-3, Ar-41, Ce-141, Sr-90. He made a motion to recommend this set to the TSP Caldwell seconded the motion. There was no further discussion. The motion passed with one negative vote (recorded as a vote change following the vote).

The Source Term Subcommittee recommends to the TSP that the set of isotopes proposed by Dr. Shleien be selected as the set to be evaluated for the air pathway.

JOINT REPORT OF THE SOURCE TERM AND ENVIRONMENTAL TRANSPORT SUBCOMMITTEES

April 1, 1993

The Source Term and Environmental Transport Subcommittees meet jointly on Thursday afternoon, April 1, from 1:00 until 5:00 pm. Maurice Robkin chaired the afternoon session. He distributed the agenda and received requests for supplemental agenda items.

Status of Source Term Work

Cal Heeb, PNL, gave a status report on source term work. He covered several aspects of air and river source term activities.

Regarding air releases from separations plant operations, the I-131 work for hourly releases during 1944-47 has been completed and reported in PNWD 2033. Data input on I-131 for 1948-49 hourly releases has been completed and the next steps are to run the source term release model and develop 100 realizations of results. The 1950-91 monthly releases will then be completed, but this will require some evaluation of effective release factors for the silver reactors. Other radionuclides to be considered include Ru-103 and Ru-106, for which timelines for the release history of incidents and monthly estimates of releases at dissolving will be provided. For other radionuclides, calculations will be required based on best available information about releases, this effort probably resulting in "bounding" estimates.

Heeb next discussed air releases from the nuclear reactor operations. Stack gas releases from reactor building vent stacks contained Ar-41, H-3, and C-14. Estimates will be made from stack gas monitoring data.

Heeb then discussed source term releases to the Columbia River from the nuclear reactors. Bruce Napier, PNL, was asked to add information about the five radionuclides that he considers important for dose calculations. Heeb noted that he was prepared to deliver river source term information on 12 radionuclides, which included Bruce's set of five. These would be provided to Wally Walters, PNL, for river pathway transport work. (See later agenda item for additional discussion and vote.)

Heeb noted that river source-term data will become available by June 30. Monthly values for the source term are being developed for the effluent from each reactor. He has completed the assembly and verification for 13,000 activity measurements for the period 1958-71.

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Heeb said that he has compiled the record for 2017 fuel element failures, identified by reactor and year. He described ways in which such failures occurred and some of the consequences.

Heeb then reported on his schedule of deliverables. His task is on schedule and expects to meet all target dates.

Progress on River Modeling

Wally Walters, PNL, provided a detailed progress report on river pathway work that has occurred since the January TSP meeting. This covered three main topics.

First Walters discussed the ongoing work on river modeling using a simplified radionuclide transport model. A Phase I type of approach (dilution and decay) was applied downstream to Bonneville Dam for the period 1964-66. The objectives of this work were to a) show the feasibility of radionuclide "routing" downstream, b) identify the significance of several variables (dilution, radioactive decay, travel time, sediment effects, and evaluation of monitoring data), and c) assist the TSP in its decision making. Walters provided several types of illustrations, including: total reactor monthly average release rates for the period; Columbia River hydrographs at Ringold and Finley; travel times along the river to various locations at different river discharges, and calculated results of transport for As-76, Np-239, Zn-65, P-32 and Cr-51. Plume effects were evident in the Hanford reach. The relative influences of half-life (decay rate) and dilution as a function of river discharge were illustrated. Source term dilution due to tributaries was also discussed and found to be mainly important for the Yakima and Snake Rivers.

Walters next discussed the testing of the CHARIMA model, an available model that is being adapted to the Columbia River for intermediate-level modeling. Marshall Richmond, who worked on the Phase I project for Battelle and is now on the Washington State University faculty, is on contract to assist Wally and handle most of the computer modeling. Initial work has been to validate the model for river profiles along the Hanford reach downstream to McNary Dam, in preparation for effluent plume analysis. Validation will next occur downstream to Bonneville Dam.

Walters then discussed key milestones for the river modeling work and plans for the final report. The modeling and draft report will be completed by September 30 and then begin PNL reviews. Walters said that the TSP should receive a draft report in December.

Recommendation on Source Term Nuclides for River Pathway

Maurice Robkin introduced a discussion of the choice of radionuclides for source term analysis for the river pathway. He referred to the budgeted activities now underway to

provide source term data on 12 radionuclides, including one "identifier". These are: As-76, Na 24, Np-239, P-32, Zn-65, Cr-51, Ga-72, Y-90, gross beta, I-131, Mn-56 and Sc-46. He noted that the first eight have been estimated by Farris and Napier (PNL) to contribute the top 99% of the dose from the surface water pathway, whereas the last four have other special interests for the river pathway (such as the amount released). Robkin further reminded the group that Napier has determined that only the first five radionuclides (As-76, Na 24, Np-239, P-32, Zn-65) need to be carried through the dose calculations because his latest screening studies suggest that they contribute about 90-95% of the total dose from the river pathway.

Robkin asked the joint subcommittees to make a recommendation to the TSP on the radionuclides to be included in completion of the source term work for the river pathway. It was moved and passed that recommendation be given to the TSP to retain the group of 12 radionuclides for source term work on the river pathway but that the TSP accept Napier's recommendation to only carry his list of five radionuclides through the dose calculation process.

Backcasting Model for Radioactive Contamination

Maurice Robkin distributed his report to the Source Term Subcommittee titled "Test of Backcasting Model for Radioactive Contamination of Production Reactors Cooling Water Effluent". He discussed the main aspects of his study and its conclusions. The backcasting of specific radionuclides based on gross-beta measurements does not seem to be generally justified, based on his relatively simple statistical evaluations. However, Rick Bates, a PNL statistician, is examining a more extensive data set with a much more sophisticated statistical analysis. The results may be more promising than those reported by Maurice.

Native American Fishing Sites

Deward Walker reported that the Native American Working Group discussed his new report on "usual and accustomed fishing sites" at its meeting on Wednesday, March 31. This report was also presented and discussed at our joint-subcommittee meeting. A list of 115 sites between Priest Rapids and Blalock Island (Bonneville Dam) was prepared. These were then grouped into 16 clusters, such as The Dalles/Celilo fishery, to better accommodate the needs for a more restricted number of locations to use in river pathway modeling and dose calculations. Walker noted that in addition to fish consumption, important exposure pathways included water use for bathing and consumption, and the use of beaches. The joint subcommittees commended Deward on his efforts to pull this information together in a useful format. Bruce Napier indicated the need for further information, such as on the years that particular fishing sites were used and on the amounts and species of fish caught. This need was supported by Environmental Transport Subcommittee members.

Atmospheric Transport

Allan Murphy distributed and briefly reviewed the "final" summary report of the TSP/CDC review of the RATCHET model, titled "Review of the Regional Atmospheric Transport Code for Hanford Emission Tracking (RATCHET)" and authored by Dr. CJ Nappo. Murphy plans to distribute the full report (with appendices) to the TSP members later in April for their review.

Van Ramsdell, PNL, then discussed the Review-Panel comments and recommendations. He indicated the actions taken in each case to respond to suggestions and correct errors. He noted that some comments can not be adequately addressed without carrying out an additional research effort and that this is beyond the scope and budget for current air pathway work.

Maurice Robkin then raised the question of which radionuclides to consider in dealing with air dispersion, beyond I-131, Ru-103 and Ru-106. Whereas I-131 dominates in the early years, people coming to the area later may have received air-pathway doses from other radionuclides. Bruce Napier indicated that this was discussed in the scoping document but that he did not have a copy with him and would instead discuss the matter with Cal Heeb.

Adjournment

There being no other matters to bring before the joint subcommittees, Maurice Robkin declared the meeting adjourned.

JOINT REPORT OF THE SOURCE TERM AND ENVIRONMENTAL TRANSPORT SUBCOMMITTEES

July 15, 1993

The Source Term and Environmental Transport Subcommittees met jointly on Thursday afternoon, July 15, from 1:00 p.m. - 5:00 p.m.

The agenda items for the meeting were:

1. Review and status of PNL Source Term Work, presented by Cal Heeb, PNL.
2. Review of Rick Bates's (PNL) statistical analysis of water activity interpolation and backcasting, presented by Cal Heeb, PNL.
3. Planning for Source Term Work after May, 1994.
4. Coordination of TSP Source Term and Transport planning for HEDRP after May, 1994.
5. Progress on river modeling, presented by Wally Walters, PNL.
6. Discussion of Battelle's "Software Requirements Specifications", by Pete Klingeman, TSP.
7. Status report on air pathway, presented by Allan Murphy, TSP, and Van Ramsdell, PNL.

Status of Source Term Work

Cal Heeb described the active PNL source term work. He is finishing up two certified source term files, one for input into the air model and one for input into the river model. The air release source includes releases to the air from the reprocessing plants and from the operating once-through reactors. The water source includes releases from the operating reactors.

The air release source is for input into RATCHET. The time period includes 1944 through 1947 as described in PNWD 2033 HEDR, plus two additional years at the same level of detail (hourly) as the 1944-1947 data. In addition to these data, the entire period of operations of the separations plants will be covered on a monthly grid (1944-1972) for the six approved radionuclides. These are I-131, Ru-106, Sr-90, Ru-103, Pu-239, and Ce-144 from the reprocessing plants. In addition, C-14, Ar-41, and tritium from the reactors will be included.

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The STRRM model will be run through 1949, accounting for the installation of water scrubbers in May 1948 and the sand filters in October 1948. The resulting iodine release factor decreased from 90.5% to 28.5% to 25% as these filters were brought on line. The final RATCHET input runs are in progress.

The air source from the reactors came from a) the graphite gas system, a mixture of He and CO2, and b) inleaked air carrying N-14 and Ar-40. These activate by neutron capture to produce H-3 from He-3, C-14 from N-14, and Ar-41 from Ar-40. The water source of C-14 and Ar-41 comes from dissolved air in the cooling water. The tritium comes from ternary fission of natural uranium dissolved in the river and tramp uranium on the fuel element cladding surfaces and from activation of the deuterium in the cooling water. The deliverable will be an evaluation of the importance of these sources.

The monthly release data for the separations plants are based on a simple formulation of tons per month of fuel processed times the inventory in Ci per ton times a release factor. The monthly data are available for all four plants and include tons processed, burnup, and cooling time. Ci/ton will be based on ORIGEN2 cross sections for each nuclide of interest. The neutron spectrum will be for the N-reactor since there is no spectrum available for a production reactor. The uncertainties resulting from this were evaluated earlier in the project and will be used here.

The release factor for iodine after the silver reactors were installed is based on some 400 measurements of release factors made in the 1960's. Account is taken of normal operations and of periodic degraded operation. Dr. Shleien raised a question of the fate of I-129 when the silver reactors were regenerated. Some discussion followed about some possibilities for estimating the doses due to this long-lived iodine isotope. Dr. Davis requested that some words be addressed to the release of ternary fission generated tritium. A code has been written to perform these calculations.

A new model to estimate the river releases has been developed, the Source Term River Release Model, STRRM. The model is a Monte Carlo simulation. The model is essentially that the release is proportional to the product of the monthly average water activity concentration for a nuclide in pCi/ml times the reactor coolant flow rate times the effective full power operating time per month. When the daily pCi/ml data are not available, the monthly source will be set to the average release rate in Ci/day times the effective full power days per month. The calculation depends on about 13,000-15,000 measurements from the literature.

The values for any given month are obtained from a hierarchy of knowledge.

1. There are one or more measurements of pCi/ml in the month.
2. There is the monthly average Ci/d available.

3. The nuclide lends itself to a regression extrapolation to the month with no data of type 1 or 2. The regression model is based on all of the physical operating parameters of the reactor and its cooling system.
4. If there are no data or applicable regression, a global average of all months with data is used. The average is taken over all reactors and all time for which there are data and includes an uncertainty estimate.

A global overview of all of the considered nuclides and times indicates that about 48% of the data are at level 1, about 3% is at level 2, about 16% at level 3 and about 33% at level 4. Each nuclide had its own pattern of data quality. P-32 tends to have lots of measurement data whereas for Ga-72 the measurements are much sparser and much more global averaging was needed. Heeb presented a set of viewgraphs showing the distribution of data quality for each nuclide for each month in the period of interest.

The I-131 source is from both activation of native and tramp uranium and from fuel failures, with fuel failures contributing up to 89% of the water source of I-131. There were some 2000 fuel failures which were recorded only as to which tube the failed slug came from. The power of the slug, and hence its I-131 content, depended on its position in the tube. A statistical location estimator due to R.R. Bloomstrand is generated by a rupture potential function which depends in a highly non-linear fashion on the tube power, temperature and burnup. Each tube's power was taken as a chopped cosine used to distribute the tube burnup over the slugs in the tube. The temperature at a slug position is obtained from the thermodynamics given the tube inlet temperature and tube power. The burnup is proportional to the power.

The result of this estimation was to predict a strong peak in the rupture rate somewhat above the midpoint of the tube.

The uncertainty for the water source term generated by STRRM arises from uncertainties in:

107 Retention basin holdup time for shorter half-life nuclides
Temperature and flow measurements
Reactor power level
Activity measurements
ORIGEN2 cross-section values due to spectrum uncertainty
Mass of fuel released into river for each failure
I-131 and Np-239 releases from fuel failures
Failed slug position
Burnup versus position in the tube
Regression for the monthly activity
Global fit projection

Heeb showed an example of the results for 100 realizations of the model indicating mean, one sigma spread, and min and max value for Zn-65 and P-32 from D reactor. There were more measurements for P-32 and the distributions are tighter.

The status of STRRM is:

Data files reorganized to 1 file for 1 month for 1 nuclide for all 8 reactors for 100 realizations with SHUFLR code. Code produces file for input to CHARIMA. 1 file has been give to Marshall Richmond to begin CHARIMA run. STRRM testing has been done. Final production runs to be done in July.

No specific action item was identified for the current contract. The Source Term Task will provide all of the inputs to the Transport Task as needed. After some considerable discussion, the general tone of the TSP members at the meeting was that all facilities and their releases additional to the reactors and reprocessing plants should be identified and tabulated to satisfy the TSP commitment for completeness made early in the project. Planning for future source term work was tabled pending TSP decisions on future goals for the HEDR project.

Progress on River Modeling

Wally Walters, PNL, provided a detailed progress report on river pathway work that has occurred since the April TSP meeting. The main emphasis has been on a thorough, formal testing of the modeling procedures and the CHARIMA river transport model in anticipation of receiving source term data files in July.

Walters, first reviewed the "task and key milestone" chart that was presented at the joint subcommittee meeting on April 1. Work is progressing very close to that schedule. Walters noted which tasks had been completed, when completion occurred, and which tasks were still continuing. Travel time routing, the conceptual model, and effluent plume analysis are completed. CHARIMA model validation is nearly complete to Bonneville Dam, with some difficulties to resolve at the Celilo Falls-to-McNary Dam reach. The model will be validated downstream to River Mile 100, just below the mouth of the Willamette River. From there to the mouth, a simpler transport model will be used to average out the daily tidal variability and produce monthly travel time values. Exploratory runs with source term data have begun, using a preliminary source term data file. Final production runs should take place during September. With respect to report documents, a draft will be provided to the Battelle Project Office by September 30, and the TSP can expect its review draft by December 15.

Walters then described the model testing and validation process. CHARIMA was installed and a test plan was run. A data file was developed on channel geometry, using information from the U.S. Army Corps of Engineers. The model was then tested for a steady (constant) input discharge. This was done first to Richland, then to McNary Dam, then to Bonneville Dam. It will next be tested to the Portland vicinity. Because this is a steady-state test rather than the more realistic unsteady-state test for which the model was designed, Walters considers this phase of testing as "credibility verification" rather than modeling verification. Unsteady flow testing (the actual model verification) will next occur. This will first be done for several Hanford reach locations where hourly data on water surface elevation are available, using the model with hourly upstream discharge hydrograph data as inputs. (Note: the final production runs will use the unsteady flows for one-month intervals to produce monthly radioactivity concentration values at downstream locations, but hydrographs vary considerably over short periods of days, requiring this level of simulation to give proper accuracy to results.) Next, the model will be run for locations downstream of McNary Dam, using river discharge at The Dalles as validation data. Then the model will be run for the entire river from Priest Rapids downstream to the Portland area.

Walters next discussed the planned experimental runs to determine sediment effects on radioactivity transport. The source term data file will be used to route specific radionuclides downstream. The resulting concentrations at test locations will be compared with environmental monitoring data. From this comparison, empirical correction factors will be developed to account for the net uptake and release of radionuclides by suspended sediment in the water. In doing this, it will be assumed that As-76 is <u>not</u> affected by sediment, whereas Zn-65 and P-32 <u>are</u> affected, giving a means for comparing transport to monitoring data for As-76 and establishing uptake coefficients based on Zn-65 and P-32. The sediment correction factors will be used to modify the river transport output files delivered to the dose model.

Walters indicated that after final testing of the full model, the QA work on the model will be completed. Then the initial deterministic production runs will be conducted, followed by Monte Carlo runs.

Marshall Richmond, PNL contractor from Washington State University, presented results from the initial rums of the CHARIMA model. He showed the computed water surface profiles between Priest Rapids and the Portland vicinity using a <u>steady</u> discharge of 79,000 cubic feet per second at Priest Rapids. The process was repeated with one dam (Bonneville), two dams (Bonneville plus McNary), three dams (Bonneville, McNary, and The Dalles), and four dams (Bonneville, McNary, The Dalles, and John Day) to represent the changing river conditions between 1944 and 1971. Problems arose in calculations between Celilo Falls and The Dalles because river flow was over falls and cascades rather than with the smooth water surface that is assumed in the model.

Richmond showed results of an unsteady flow introduced to the model at Priest Rapids. A rectangular flood wave was shown to modify its shape in the downriver direction to Richland, both with respect to discharge and water surface elevation, as should occur in nature. This test indicates that the model is operating correctly.

Richmond explained the plume modeling approach. Based on field measurements made of the plume in the 1960s, an adjustment factor can be calculated. It will relate the average radioactivity concentration within the plume to shoreline radioactivity concentrations.

Action items for river pathway work:

- Complete ongoing tasks according to established schedule
- Better knowledge of the uptake and release of radionuclides by suspended sediment is desirable, to establish better correction factors and to help differentiate between sediment-sorbed radioactivity and hot particles from the reactor.

Discussion of Battelle's "Software Requirements Specifications"

Pete Klingeman led a question-and-answer discussion of the Battelle pre-decisional draft paper "Software Requirements Specifications for Columbia River Pathway: Source Term, River Transport, Environmental Accumulation and Dose Models: (BN-SA-3799 HEDR). The questions asked were based on TSP member reviews of the paper. Responses were offered by Bruce Napier, Bill Farris and Mike Thiede.

Clarification was given of the distinction between river cross sections and reach segments (paragraph 4.5). Klingeman recommended that this paragraph be revised to clearly identify the location of each of the 14 cross sections and to be sure that the reach from the Walla Walla River to the Umatilla River is included.

TSP concern was voiced that the period of dose calculation was not clearly stated. Paragraph 3.5 refers to source term data for September 1944 through January 1971 inclusive, for a total of 371 months. Paragraph 3.6 refers to source term data on seven radionuclides for river transport work for January 1950 through January 1971 for a total of 253 months. The River Transport Module section refers to requirement 3.5 (see paragraph 4.6). The Environmental Accumulation Data section refers to the 1950-1971 period of concern for drinking water (see paragraph 5.3). The Environmental Accumulation and Dose Module section refers to requirement 3.5 (see paragraph 6.11) for Columbia River anadromous fish and Willapa Bay shellfish. Thus, it would appear that doses will be calculated for the period September 1944 through January 1971. But, Bruce Napier stated that river pathway dose calculations will only be calculated for 1950 through January 1971. This would cause a gap in dose calculations from September 1944 through December 1949 (a 5+ year period). Napier stated that the outcome of the September 1992 meeting on river pathway modeling led to this decision. However, no one present had the

documents at hand from which to check this. THIS SITUATION MUST BE CONSIDERED BY THE TSP. WAS SUCH A GAP IN DOSE CALCULATIONS INTENDED AND IS IT ACCEPTABLE?

TSP concern was expressed that lamprey eels were not included among the biological species, yet were possibly important as a food source to Native Americans. No resolution of this was achieved.

TSP concern was expressed that the published bioconcentration factors may not be representative of the Columbia River pathway. Napier state that reliance will be placed on nearest-location environmental monitoring data for aquatic biota, rather than published values. There was additional TSP concern over how this will relate to waterfowl, since they range over such great distances. THERE REMAINS SOME TSP CONCERN OVER THE BIOACCUMULATION FACTORS. However, no resolution of this dissatisfaction was achieved.

Status Report on Air Pathway

Allan Murphy indicated that the report of the CDC/TSP peer review of the RATCHET model has been transmitted to Battelle. Chairman John Till has requested that Battelle provide the TSP by mid-August with a letter report describing the actions taken by Battelle staff in response to the issues and questions raised - and recommendation made - by the reviewers. Formal TSP approval of the RATCHET model will be sought via a telephone/telefax vote taken in late August or early September.

Murphy also indicated that a meeting will be held at Battelle on August 19-20 to discuss the technical aspects of the validation of the RATCHET model. Participants in the meeting will include D. Barth, K. Kopecky, and A. Murphy from the TSP and B. Napier, J Ramsdell, and J. Simpson from the Battelle staff. It is anticipated that this meeting will lead to the identification of a suite of methods that can be used to assess and describe the validity of the RATCHET model. These methods also may prove to be useful in other HEDRP validation studies.

Van Ramsdell provided a brief status report on the RATCHET model and related air pathway work. He indicated that the meteorological data base for the period 1944-1949 is now complete and has been subjected to various quality control checks. The data base is now stored on his SUN workstation. A report describing this data base will be forwarded to the TSP at the end of September (on schedule).

The issues and questions raised in the course of the peer review of the RATCHET model have been addressed, and various revisions and refinements have been made to the model. The model code is now under independent review, and no major problems have

been encountered. An updated version of the RATCHET users' guide, consistent with the revised/refined computer code, is in preparation.

A report exploring possible alternative representations of wind field in the RATCHET model has been completed and distributed to the TSP. This report provides justification for the particular wind field representation to be used in the operational version of the RATCHET model.

Van Ramsdell indicated that several other refinements to the RATCHET model are contemplated, including a modification in the treatment of puff radius which may reduce certain inconsistencies in the results and, at the same time, decrease model run time. He also reported that the RATCHET model will be ready to start computer runs to obtain 100 realizations of atmospheric transport calculations later this month (July).

Adjournment

There being no other matters to bring before the joint subcommittees, Maurice Robkin and Pete Klingeman declared the meeting adjourned.

JOINT REPORT OF THE SOURCE TERM AND ENVIRONMENTAL TRANSPORT SUBCOMMITTEES

October 9, 1993

A joint meeting of the Source Term and Environmental Transport Subcommittees was held Thursday in which we received some reports by Cal Heeb that were based on the status of the source term work and statistical analysis. Cal gave a detailed discussion of this on Friday, so I won't go into that.

We have no specific action items. I reported on the work I did this summer, gave a very preliminary set of data on the numbers I found. The only item which I would call an action item was to go back and split out the data and treat them separately from the data which have actual measurement values reported. That's really all we have.

A joint meeting of the Source Term and Environmental Transport Subcommittees was held on Thursday, October 7, 1993. The agenda included reports by Cal Heeb of Battelle, reporting on the status of the source term work; by Rick Bates of Battelle reporting on the statistical analysis of the source term; by Wally Walters of Battelle, reporting on the status of the river transport work; by Van Ramsdell of Battelle, reporting on the status of the atmospheric dispersion work; and by Maurice Robkin, TSP, reporting on his summer study of the releases from those Hanford facilities which did not contribute the major releases already considered by PNL. Maurice Robkin chaired the first and last parts of the meeting dealing with the source term and Allan Murphy chaired the middle part of the meeting dealing with environmental transport.

Cal Heeb described the status of the current tasks. He limited himself to an overview of the following subtasks, with a more detailed report to be given on Friday, October 8:

1. Release to the air from the chemical processing plants,
2. Release to the river from reactor operations, and
3. Releases to the air from reactor operations.

The details of this work were presented on Friday, and will not be repeated here.

Rick Bates described his task, which was to look at all of the reactor effluent data and come up with the best representation of activity in terms of known reactor physical parameters and a best estimate of the uncertainty in any projected values. The eight once-through production reactors were separated into two groups; KE and KW on the one hand

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and B, C, D, DR, F, and H on the other. The separation was made on the basis of the power history of the reactors.

The regression approach selected was to use as simple a model as possible, using such parameters as river flow, reactor flow, reactor inlet and outlet temperatures, water pH and reactor power lever. The multiple regression for seven nuclides, P-32, Sc-34, Cr-51, Zn-65, As-76, Y-90, and Np-239 against the parameter set gave correlation R squared values between about 0.3 and 0.7.

Thus, the regression predictor is not expected to by an overwhelming success but is considered to be usable. The regression fit is used to generate data of quality class 3 for those periods of time for which there are no measured data given for concentrations or discharge rates.

The discharge activity values seem to fit lognormal distributions with values concentrated toward the lowest values and with a long tail toward high values. The distribution allows for the exclusion of negative values and the capturing of high tail values.

Maurice Robkin described his preliminary results for the releases of radioactivity from facilities other than the primary air and water releases from the reactors and reprocessing plants. The values are preliminary and will be rechecked and verified before a final report. In the discussion , it was pointed out that treating "less-than" values as "equal-to" values will lead to a large overestimate of releases. A rational approach to treating "less-than" values will be worked out. The work will continue with vetting the values and addressing the detection limit question.

JOINT REPORT OF THE SOURCE TERM AND ENVIRONMENTAL TRANSPORT SUBCOMMITTEES

January 20, 1994

Preliminary discussion of latest Battelle final reports for Source Term and Environmental Transport

Source Term Reports for Atmosphere and River

Stan Davis raised the question of activation of A-41 from air dissolved in the reactor coolant that was then outgassed at the holdup basins. Cal Heeb responded that there were no data available. Some estimates can be made based on calculation.

Maurice Robkin asked whether the N reactor was considered in source term work for the river pathway. Cal indicated that because the N reactor did not depend on once-through cooling with river water (the case for the other eight reactors) the only condition that might result was leakage to the river during fuel reprocessing.<<<< ?? Cal noted that he is preparing a report on the N reactor that will be submitted to the Battelle project office in mid-February.

Green Run

J.R. Wilkinson (Confederated Tribes of the Umatilla Indian Reservation) noted Pete Klingeman asked for some guidance on interpretation of Figure 4.1, which involves ratios of observed to calculated retention times in the retention basins. Cal explained the Soldat measurements of gross beta that allowed better estimates of retention time by considering throughput of the flow without complete mixing rather than simply using the cross-sectional area of the tanks and flow rate. These were used to make better calculations of retention for individual radioisotopes (sometimes as brief as one hour rather than the previously assumed four hours) but could not be used to correct gross beta calculations because of the unknown mix of radionuclides present using this measure of radioactivity.

River Transport Report

Because this report was delivered at the last minute and some TSP members had not yet received a copy, Marshall Richmond was asked to give a brief summary. He described the model inputs (river flows, channel geometry, and channel roughness), the model calibration and verification process (using flows to predict river surface elevations for comparison with

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historical data), and model use to develop river water radionuclide concentrations for dose calculations. He and Wally Walters then gave further elaboration on the findings and responded to questions on the report. They noted that the reactor effluent plume extending from downstream past the Tri-Cities was considered analytically. Tributary inflows were treated as causing instantaneous dilution of Columbia River flow at the point of entry. Water withdrawals from the river would have concentrations based on modeled values, with no other special analytical treatment. Tidal effects in the river below Bonneville Dam were ignored for simplicity with little effect down to Portland. Sediments were not accounted for but their effects on dose was considered to be minor, except for specific pathways where they were accounted for indirectly -- e.g., with oysters using direct monitoring data. Most calculated radionuclide concentrations in river water agreed well with monitoring data. However, there were cases were there was scatter, with the greatest scatter found for ZN-65 at Richland in 1965 (shown in Figure 25 of the report). In the ensuing discussion, Jack Corley noted the procedures used for handling composite samples, such that they contained exactly what was in the water, without contamination from the container. J.R. Wilkinson (Confederated Tribes of the Umatilla Indian Reservation) noted concerns over the implications for Treaty "reserved rights", thermal loading of the river from reactor operations, and resulting impacts on salmon.

Deadlines for TSP Member Review and Response of Draft Final Reports of Source Term and Environmental Transport

a. PNL-2222-HEDR: RADIONUCLIDE RELEASES TO THE ATMOSPHERE FROM HANFORD OPERATIONS, 1944-1972.

    1. TSP comments due to Kathy CharLee: Feb. 15, 1994
    2. TSP summary and comments due to PNL: Feb. 25, 1994

b. PNL-2223-HEDR: RADIONUCLIDE RELEASES TO THE COLUMBIA RIVER FROM HANFORD OPERATIONS, 1944-1971.

    1. TSP comments due to Kathy CharLee: Feb. 1, 1994
    2. TSP summary and comments due to PNL: Feb 22, 1994

c. PNL-2225-HEDR: RECONSTRUCTION OF RADIONUCLIDE CONCENTRATIONS IN THE COLUMBIA RIVER FROM HANFORD, WASHINGTON TO PORTLAND, OREGON: JANUARY 1950-JANUARY 1971.

    1. TSP comments due to Kathy CharLee: Feb. 7, 1994
    2. TSP summary and comments due to PNL: Feb 23, 1994

Remaining Task Work for PNL

Source Term: Cal Heeb reported a review of releases from facilities other than those from which the major releases came will be made. These include

C Plant Semi-works
Z plant- Plutonium Finishing Plant
U plant- Uranium Recovery
108B Tritium Recovery Building
Plutonium Recycle Test Reactor (300 area)
Low Power Reactors: High Temperature Lattice Test Reactor
Physical Constants Test Reactor
TRIGA research reactor
PUREX campaigns: N Fuel reprocessing
Thorium reprocessing
SEFOR
Plutonium Recycle Test Reactor

Criticality accidents: Possible releases from 2 criticalities:

1951: Pu nitrate solution criticality: Farmhouse near White Bluffs. Estimate of 1E17 fissions.

1962: 234-5 Building: RECUPLEX Pu finishing plant. Pu nitrate tank criticality. Estimate of 1E17 fissions. Criticality lasted for 37 hours before access permitted shutting down reaction.

Fast Flux Test Facility:

All releases were atmospheric except for PRTR which might have had some release to the river.

River Pathway Task: Wally Walters reported.

1. Receipt and incorporation of TSP comments and publication of final report for subtask 0404C (milestone 0404C).
2. Prepare for CDC audit on Feb. 20, 1994.
3. QA files completion.

Atmospheric Pathway Task: Van Ramsdell reported.

1. Task work is finished.
2. Summarized model domain and mass balance for I-131. He pointed out that the balance indicates 10% radioactively decayed (calculated); 56% deposited within the domain (calculated); the balance (34%) is assumed to have left the domain. Coefficients of variation were estimated as 5% for the source term; 10% for the iodine leaving the domain; 16% for the decay; and 12% for deposition.

   The non-deposited iodine is ascribed to iodine as organic compounds. Organic iodine serves as a reservoir for inorganic and particulate iodine. It is expected that little iodine left the domain towards the west and south. It was suggested that the directions of the escape from the domain probably were distributed similarly to the deposition footprint.

Mechanisms for Future Interaction Between TSP and Hanford Contractors

The subcommittees discussed the need for a mechanism to permit interaction between TSP and Hanford contractors to deal with requirements for information as future TSP work evolves. There exits currently a Technical Service Agreement that permits PNL workers to deal with external interactions on an ad hoc basis. These agreements seem to be set up with various agencies such as universities, etc, to facilitate interaction. An example is the probable need for document access, review and retrieval as necessary.

JOINT REPORT OF THE SOURCE TERM AND ENVIRONMENTAL TRANSPORT SUBCOMMITTEES

April 21, 1994

The Source Term and Environmental Transport subcommittees held a joint meeting from 1:30 to 3:30 pm in the Silver Room at the Pasco Red Lion Inn. Maurice Robkin chaired the session and Pete Klingeman took notes.

The first order of business was to develop the meeting agenda, based on suggestions previously discussed and input from subcommittee members. It was decided to cover two main topics: 1) a review of Battelle response to the river report TSP comments from the January subcommittee meeting; and 2) discussion of possible further work needed on source term and environmental transport to develop doses to special populations.

Update on Battelle Modifications to Report PNWD 2225

Wally Walters (PNL) described some of the revisions being made in response to TSP comments on report PNL-2225-HEDR: Reconstruction of Radionuclide Concentrations in the Columbia River from Hanford, Washington to Portland, Oregon: January 1950-January 1971. He said that two new subsections would be added to the report: one on scatter of monitoring data and one on the modeling treatment of the river near Portland. He then discussed each of these.

Most calculated radionuclide concentrations in river water agreed well with monitoring data. However, there were cases were there was scatter, with the greatest scatter found in the reaches nearest the Hanford Reservation. A distinct seasonal relation to scatter was noticed which has been re-evaluated since comments were made at the January joint-subcommittees meeting. Wally showed results of earlier validation work with Cr-51 for the river pathway and the WSU-CHARIMA model. He showed that the May-July (wet season) environmental monitoring data fit tightly about the simulated concentrations of radioactivity in water, whereas there was more scatter during other months (particularly winter months) of low river flows. During the low-flow periods, hydropower peaking operations at Priest Rapids caused large daily and day-to-day variations over a weekly cycle in water discharge that resulted in variable dilution of radioactive effluent from the reactors. Hence, day-to-day concentrations of radionuclides in river water were much more variable for all months except for the May - July high flow period. The CHARIMA model runs, using monthly-average source term values and daily river discharges tended to smooth out the variability, whereas grab sampling would not have done so and even composite samples might reflect

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some of the actual variability in river concentrations. Tributary inflows caused dilution of Columbia River flow at the point of entry that reduced the relative variability. Travel-time dispersion further reduced the variability.

The result is that Wally considers some of the scatter in the model validation work to be attributable to variable river discharge that could have affected and added scatter to the environmental monitoring data. Hence, some of the uncertainty in dose estimates from river modeling may be due to actual river phenomena that affected the monitoring data.

The modeling treatment of the Columbia River at Portland (river mile 100) was then discussed. A wide range of river levels occurs near Portland due to tides, tributary inflows from the Willamette River, and upriver flows in the Columbia River. For modeling purposes a fixed river water surface elevation was selected at 10 feet, mean sea level. This is about 8.5 feet above the minimum low water plane and about 6.5 feet below the flood stage plane at RM 100. The consequence of this is that the model gives a somewhat steeper slope and faster travel time (with less opportunity for radioactive decay before the water reaches Portland) at very large river discharges and a somewhat flatter slope and slower travel time (with more opportunity for radioactive decay) at very small river discharges. At and near average conditions, the differences are small.

Wally also showed the TSP members some Cr-51 data for the Columbia River at The Dalles where the results were plotted on a semi-log plot rather than an arithmetic plot. This shift in the plotting scale for radioactivity gave the data a more consistent appearance over the year.

Possible Remaining Source Term and Environmental Transport Work to Develop Doses to Special Populations

Discussion addressed the on-site close-in modeling for doses to workers, construction workers, military, etc. Van Ramsdell suggested using a polar grid out to 20 km with a straight line Gaussian plume to do a simple calculation with 100 realizations. He suggested using monthly source term data and spreading it uniformly to interact with the hourly meteorological data. The joint frequency distribution for the 100 area is different than for the 200 area because of the effect of the bluffs. A separate meteorological data base for the 100 area is needed. Winds tend to blow up and down river. More recent met data are available that can be applied to the earlier period. There are met data for the 100 area from the operating station there for the last 10 years.

Inhalation will probably dominate doses in the 100 area. External exposure may also be important for noble gas plumes. Cal Heeb pointed out that for the releases of about half a million curies of Ar-41 total from all 8 reactor stacks, Bruce Napier did an estimate of the dose to someone living on the Wahluke slope of about 1.2 millirem.

Van Ramsdell suggested using annual reports of doses from the 100 and 200 areas based on calculated values of doses using unit releases of nuclides. These unit dose values can be multiplied by the actual amounts of any source term we develop to give the resulting dose. He also suggested using the I-131 concentration calculation and obtaining the concentration of Kr-85 by multiplying the ratio of the inventory of Kr-85 to that of I-131. To implement this, it is necessary to know the release fraction of I-131 appropriate to the given concentration value. Cal Heeb pointed out that in the annual reports of doses due to releases from the reactors, Ar-41 dominates the dose by large amount.

For the water pathways, the short-lived nuclides may be important for the cases of site workers, construction workers, and military personnel using water from riparian wells or directly from the river. Mark Freshly pointed out that the Hanford Environmental Health Foundation provided a summary of the drinking water system based on on-site monitoring data for both water quality and radionuclides for potable water coming from the Columbia River and from ground water. References should be in the groundwater report.

Jack Corley commented that military personnel stationed at the NIKE site used water trucked in but he didn't know the exact source. The water could have come either from the Richland water supply or from Site facility water. For the early construction period of WPPPS reactor and FFTF workers used well water. Occupational exposure for these workers is addressed in the groundwater report.

These suggestions indicate that a considerable amount of the information needed to evaluate the doses to on-site persons may already be in hand.

JOINT REPORT OF THE SOURCE TERM AND ENVIRONMENTAL TRANSPORT SUBCOMMITTEES

July 14, 1994

The Source Term and Environmental Transport subcommittees held a joint meeting from 8:00 to 11:30 am in the Workshop Room at the Pasco Red Lion Inn. Maurice Robkin and Pete Klingeman co-chaired the session.

The meeting agenda consisted of discussion of final work by the subcommittees and possible future work needs that might be considered by the reduced TSP in the areas of source term and environmental transport.

Hot Particle Problem

The meeting began with a discussion of the Hot Particle problem. Maurice Robkin proposed that the meeting address the scientific issues involved in evaluating the importance and impact of hot particles. These include the distribution of size and shape of emitted particles, their composition, what radionuclides may be associated with them, how they are transported and what sort of modeling would be required to handle the problem.

Del Barth mentioned the Comprehensive Report on the Hot Particles outlining some of their properties and radioactivity. Jim Thomas reminded the Committee of a series of Hot Particle Documents including a report by Till and Miller, a TSP staff memo by Joe Stohr, an April 1992 HEAL memo by Jim Thomas, and a Senate Government Affairs Committee report by Bob Alverez addressing plutonium particles in the period from the mid-1940s to about 1951 and the ruthenium particles from 1952 to 1954. These documents contain extensive references on the subject. Main area of concern was worker exposure.

Thomas pointed out that particle concentrations found on Mount Rainier, Washington and Mullen Pass, Idaho, were in about the same as the concentrations found in Richland. These were particles containing plutonium, cerium and strontium. The particles were identified as from the rusting of the black iron ductwork at the reprocessing plants. Shleien mentioned a report on autoradiography of particles found on masks, filters and wipes. Davis pointed out the problem in the early days before vehicles were monitored of the redistribution of larger hot particles deposited on site near their source. These particles were picked up and transported by vehicles.

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Murphy raised the question of what is the existing knowledge about transport of particles. It was pointed out that for small particles, in the range of smokes and fumes, there is quite a bit of information in the air pollution literature. Experts such as Professor Tim Larson at the U.W. can serve as consultants about the dispersion of this size range. Caldwell pointed out that at the Comprehensive Information Center at the Nevada Test Site there is a lot of information on measurements of air filters. The size and composition distribution may be different, but for the small respirable particles, their observed distributions may be relevant.
Shleien suggested approaching problem from a worst-case situation to get an upper bound of dose. If doses on worst-case are not important, the problem can be set aside. Some of the monitoring information from 1947-1948 can be used. Templeton mentioned the Geneva Conference report by H. Parker on the ruthenium particles.

Robkin pointed out the possibility that the mist emission will carry soluble nuclides from the reprocessing solutions. If these adsorb onto transportable non-respirable particles and are breathed and swallowed, they can cross the gut and be transferred through the body. If the mist droplets evaporate, they may become solid particles small enough to be respirable. If they remain soluble, they can cross through the lung tissue to the blood and be redistributed in the body.

Shleien mentioned other sources of reports: Suitland, a federal storage facility in the Washington, D.C. area, at DOE headquarters; and at the Federal Archivist. Shleien will be surveying some of these documents.

Robkin will talk to Fred Cross, PNL, on the dosimetry of soluble plutonium. Roessler will talk to Karl Morgan about hot particle experience at Oak Ridge.

Davis cautioned the Committee about the risk of using a worst case scenario for the impact of Hot Particles. It has some use for scoping and to give the upper bound to determine if there is justification for more detailed investigation. The very low probability of the worst case assumptions being the actual cases can lead to misinterpretation and misuse of the result by individuals who are not experienced in these sorts of calculations. Murphy offered to identify an appropriate expert in transport of particles.

Barth recommended developing the equivalent of what is called in the context of the Clean Air Act a "criterion document" that describes what is and is not known about hot particles. Templeton suggested review and collation of the Hanford data. There was a consensus that the proper first step is to collect, collate and interpret all of the exiting documentation on the Hot Particles starting with Hanford data and then perhaps expanding to other sites where the problem also occurred. Shleien pointed out that there are already other searches going on at Rocky Flats, INEL, Savannah River Site, and Fernald. We may be able to make use of information uncovered in these searches.

Glyn Caldwell moved that the TSP begin the consideration of the Hot Particle Problem by doing an extensive document review to determine the "Who, What, Where, When and How" of the problem, mostly at Hanford, as well as elsewhere, to interview the available experts, make an assessment as to whether a Hot Particle dose assessment can be made from those data. Based on that procedure, the TSP would then recommend any additional scientific work that would be needed to carry out such a dose assessment. Any TSP member who has time, funds and interest is encouraged to begin on the various information collection tasks. Robkin will serve as a collection point to collate all of the information discovered and to evaluate it for recommendation for the next step in the analysis.

Davis discussed the problem of activation of the trace elements in the river water and described a procedure to make a rough estimate of these based on comparison with a reference nuclide such as sodium. This may not represent an important dose contribution, but needs to be addressed for completeness.

Davis will write up his work on the activation of trace elements in the once-through production reactors by the end of this fiscal year. Argon-41 activation in the reactors from dissolved argon may be a problem in the vicinity of the settling basins. Also Ar-41 generated from occasional air leakage into the reactors may have produced some dose in the reactor workers. Davis will also look at the tritium production from He-3 in the reactor helium cover gas. Noble gas releases from the reprocessing plants should be examined for significance to dose to workers in or near the plants. Davis will write up his recommendations for future work on these problems.

Shleien suggested that the seasonal bias in the model calculations compared to environmental measurements should be addressed. Particularly the winter underestimate should be addressed.

<u>Proposed "Closure Report" for Environmental Transport Subcommittee</u>

Pete Klingeman distributed for discussion a draft outline of a closure report that he intends to write for the Environmental Transport Subcommittee. As background to discussion of the outline, this Subcommittee has worked with a wide range of task activities since it was established. These activities have now come to a close, although there will be elements of environmental transport to be considered in future TSP work. The three main transport pathways have been discussed in many Battelle reports and at all TSP meetings. The future TSP is not likely to have a subcommittee specifically charged with environmental transport, even though it is likely to have a technical subcommittee. Therefore, it appears to be appropriate at this time for the chair of the ET Subcommittee to summarize in a brief report the workings of that group and the documents produced.

The Subcommittee discussed the pros and cons of a closure report. Everyone agreed that if written, the chair should do the work! Klingeman agreed to develop a draft report in the next month and distribute copies to the subcommittee members for review by mid-August. They will respond by mid-September. He will then revise the draft by the end of September and submit it to the TSP for their October meeting.

Subcommittee members and others present at the meeting recommended that the report be generally non-technical, but contain references to the important actions taken and the technical information sources. It would be about 10 pages long and could be thought of as a large "Fact Sheet" on the overall Environmental Transport effort of the TSP. In effect, the report would be a "road map" of the Environmental Transport Subcommittee process and activities that would be useful to someone who is interested in what was done by the TSP on environmental transport. But the report would not be filled with the results – which would be indicated by reference to other reports.

The suggestion was made that other subcommittees that will "expire" may also want to consider closure reports for their activities. The Environmental Transport Subcommittee proposes to the TSP that this be considered. The availability of such reports in the near future for these groups may assist the future TSP in later writing the final dose reconstruction project documents.

Discussion of Future Environmental Transport Work

A short discussion followed concerning recommendations made earlier this year about transport-type tasks in the long-range workplan. There was general agreement that validation of the river and air pathway dose calculations was a very high priority. There is good confidence in the work on the river pathway. Some questions about the bioconcentration factors were for anadromous fish may not be resolvable due to the limited environmental monitoring data; therefore, use of resident fish as surrogates provides a suitable alternative approach for dose estimates. Validation of the atmospheric pathway doses and uncertainties is more critical. The importance of supplemental work was stressed and it was considered that many of the proposed tasks in the long-term work plan developed last Spring deserve high priority.

APPENDIX E

TECHNICAL STEERING COMMITTEE REPORTS

OF THE HEDRP FOR THE SOURCE TERM

TECHNICAL STEERING COMMITTEE REPORTS OF THE HEDRP FOR THE SOURCE TERM

Hanford Facilities and Their Releases Presented to the Health Professional Workshop, Richland, Washington, June 6, 1990. M. Robkin, June, 1990.

Radionuclides Discharged from Hanford Production Reactors - Report to the Technical Steering Panel, October 8, 1992, M. Robkin, October, 1992.

Releases from Other than the Main Release Facilities: Report to the TSP, M. Robkin.. Selection of Significant Nuclides Released to the Air, M. Robkin, June, 1993.

Summary of Final Source Term Report, Iodine-131 Releases from Hanford 1944-1947, M. Robkin.

TSP Fact Sheet # 10: The Source Term Reveals the Location and Amount of Hanford Radiation Releases, TSP, July, 1990.

TSP Fact Sheet # 12: The Green Run, TSP, March, 1992.