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CALCULATION OF DRINKING-WATER RADIATION EXPOSURES
FROM COLUMBIA RIVER WATER SOURCES

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and
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September 16, 1958

Radiation Protection Operation
Hanford Laboratories Operation
GENERAL ELECTRIC COMPANY
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**HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON**

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CALCULATION OF DRINKING-WATER RADIATION EXPOSURES
FROM COLUMBIA RIVER WATER SOURCES

Introduction

In the continuing effort to improve the evaluation of environmental radiation exposures, it is advisable from time to time to document in detail the current methods of evaluation, the reasons for choices, the assumptions that are necessary, and the references to data used.

This document not only records the greater detail now employed in the method of evaluation of potential exposure from drinking water, but also the discontinuance of use, in exposure evaluation, of an "unaccounted fraction". This is the arithmetical difference between analyses of gross beta emission from water samples and the sum of the "beta" emissions from individual isotopes analyzed in the same samples. Its use resulted in the reporting of two estimates of potential exposure from the same samples, one with "unaccounted fraction" factored in, and the other without.

Development of the Evaluation Method

There is considerable historical development behind the present method of evaluating drinking-water exposure. Since 1945, routine measurements have been made of gross alpha and beta activity in reactor effluent and river water. Since 1944, efforts have continued to measure the individual isotopes in reactor effluent; until 1955, these were enough to indicate that potential exposures from drinking water were low.

In 1955, Healy and Rostenbach gathered all such isotopic results for not only normal effluent but also purges, ruptures, and special river samples, and produced the most comprehensive review and evaluation to date⁽¹⁾. The estimates of peak and even average concentrations foreseen for future operations were disconcertingly high in comparison to maximum permissible concentrations. Due to lack of enough analyses at that time, there were major uncertainties in evaluation and prediction. Some of the causes were:

1. Inadequate characterizations of the "normal fission products" and the "unaccounted fraction"
2. Inadequate data on purges and ruptures
3. Inadequate corrections for depletion of radioisotopes in the river, due to decay travel time, precipitation, biological uptake, etc.
4. Lack of correlations with reactor power levels, water treatment, and other process variables.

In 1956, Healy spelled out in detail⁽²⁾ the steps necessary to credible evaluation of exposure, including isotopic analyses of reactor effluent and river water, purges and rupture effluent, and correlation of isotopic content with power level, water treatment, etc., for predictions of future conditions. The reasoning and recommendations in this document are still basic, current, and necessary to further progress in the safe disposal of reactor effluent water to the Columbia River. Most of the subsequent investigations and reports in this program were due to the incentives of Healy's document.

In 1957, Clukey detailed a routine exposure evaluation method⁽³⁾ and the assumptions and choices made. At that time, evaluation was still dependent on extrapolation from reactor effluent analyses.

In 1958, Healy, Andersen, Clukey and Soldat prepared a paper for the Second Geneva Conference⁽⁴⁾ adding up exposures from all sources in the Hanford environs in 1957, in which exposures from reactor effluent discharged to the Columbia River were tabulated on the basis of actual analyses of river and sanitary water for most of the year.

Whenever gross activity measurements of air or water indicate concentrations in excess of the maximum permissible concentration (MPC) of any isotope that may be present, then isotopic analyses are necessary to evaluate potential exposures. Gross activity measurements have continuing value for showing trends and geographical distributions, but provide at best, only crude estimates of exposure.

Recently, several major changes pertinent to exposure evaluation have occurred. Isotopic analyses of river and sanitary water at points of use have been made routine; maximum permissible limits for many isotopes have been reduced; and reactor power levels and effluent activity have continued to increase. The depletion of radioisotopes in the river due to natural processes other than dilution and decay has been measured by Nielsen and Perkins⁽⁵⁾ and initial depletion data for sanitary water treatment processes have become available.

Recent revisions in the recommendations⁽⁶⁾ of the National Committee on Radiation Protection and Measurements (NCRP) pertinent to environmental exposure limits are being incorporated into the HAPRO Radiation Protection Standards⁽⁷⁾ as follows:

"The radiation or radioactive material outside the HAPRO controlled area, attributable to normal operations within the controlled area, shall be such that it is improbable that any individual will receive a dose of more than 0.5 rem in any one year from external radiation, or receive an average body burden of radionuclides more than one-tenth of the maximum permissible body burden for radiation workers. . . . The non-controlled dose and body burden may be integrated over time intervals up to one year."
(RPS 3.1)

"The concentrations of radioisotopes from Hanford in air and water in areas not controlled by HAPRO shall be limited to one-twentieth of the maximum permissible concentrations in air and water for radiation workers." (RPS 7.2)

The NCRP suggests control of concentrations in air and water (and foods) in non-controlled areas to levels not exceeding one-tenth of the respective MPC's for radiation workers. RPS 7.2 specifies control to one-twentieth MPC to allow for economic limitations of sampling and analyses, for delays between release and control-feedback from the measurements and evaluation, and for variable combinations of exposure from numerous sources to people at different locations in the environs.

The occupancy and use factors recommended by the NCRP⁽⁶⁾ are used in scientific estimates of actual environmental exposures from all sources, and not directly in the control of radioactive effluents from Hanford plants.

In environmental exposure evaluation, it is important to make measurements as close as possible to the actual exposure to minimize extrapolations from remote measurements. The degree of effort depends on such factors as the accuracy desired and economically attainable, the complexity of the ecological systems involved, and the levels of radioactivity at various points of exposure. As isotopic concentrations and analytical ability at Hanford have permitted, the points of sampling have been extended from reactor effluent water to the river, to sanitary water, to irrigation water, and to foods. At the present time, a human body monitor facility is being built which will provide information on isotopic body burdens of employees who reside in the environs. A number of such measurements have already been made with the prototype instrument.

It is necessary to produce evaluations directly comparable to the recommended maximum permissible limits for exposure^(6,7). The method of evaluating in dimensionless fractions of maximum permissible exposure, used in Table 1 and reference 4, permits the addition of exposures from the several sources, such as drinking water, food, air, external radiation, etc. By incorporating the corresponding exposures and limits, whether body burden, dose, or concentration in air, water or food, no further comparison is necessary.

Environmental exposures may be averaged over periods up to one year. Enough isotopic data are now available from past years that monthly reports in the near future may include the running 12-month average estimated exposure.

Method of Calculating Drinking-Water Exposure

The mathematical method is essentially the same as previously documented⁽³⁾. The concentration (C) of each isotope is divided by its maximum permissible concentration for the most critical organs. For each organ, the list of C/MPC is added; the largest sum is the fraction of maximum permissible exposure, and indicates the most critical organ. The method is employed in Table 1. Accepted MPC values for HAPs are listed in Appendix A of the Manual of Radiation Protection Standards⁽⁷⁾.

It is not economical to analyze every sample for isotopes contributing less than one or a few per cent to exposure, but it is necessary to know that no appreciable contributors are being overlooked, after accounting for the principal contributors in the routine program. One method is to monitor the gross activity, usually both beta and alpha emissions. However, such measurements have inaccuracies difficult to overcome, as discussed later, so that inclusion of such gross activity measurements in exposure evaluation actually can lead to greater errors.

A supplementary method is to measure the less important isotopes with less frequent routines, then make conversions for dilution, decay, precipitation, ecological up-take, etc., based on special investigations of such factors, to include them in exposure evaluations using current analyses of the more important isotopes. Table 1 includes 14 isotopes converted by such factors from numerous earlier analyses. The methods of conversion using pertinent factors are shown in Tables 2 and 3. The concentration of isotopes at mid-reactor river location in Table 3, were averaged from analyses of reactor effluent made since 1954, but mostly in 1956-7, and normalized for reactor power level and water flow.

Table 1. Calculated Exposure from Drinking Columbia River Water at Pasco, July, 1958

Isotope	Conc. (C) Units of 10^{-6} $\mu\text{c/cc}$	MPC# GI	C MPCGI	MPC# Bone	C MPCBone	MPC# Thyroid	C MPCThy.	MPC# Kidney	C MPCKid.
Na-24*	.895	8×10^{-4}	.00112	8×10^{-4}	.00112	16×10^{-4}	.00056	8×10^{-4}	.00112
Si-31	.039	9×10^{-4}	.00004	3×10^{-2}	.00000	6×10^{-2}	.00000	3×10^{-2}	.00000
P-32*	.033	8×10^{-5}	.00041	2×10^{-5}	.00165				
Ca-45*	.012	2×10^{-3}	.00001	1×10^{-5}	.00120				
Sc-46*	.018	9×10^{-5}	.00020						
Cr-51*	2.37 (.045)**	2×10^{-3}	.00118					7×10^{-2}	.00003
Mn-56	.510	3×10^{-4}	.00170					1.5×10^{-2}	.00003
Fe-59*	<.001	3×10^{-4}	.00000						
Co-60	.0012	4×10^{-5}	.00003						
Cu-64*	1.28 (.742)**	5×10^{-4}	.00256						
Zn-65*	.096 (.0027)**	2×10^{-4}	.00048	6×10^{-3}	.00002				
Zn-69	.256	7×10^{-5}	.00366	.002	.00013				
Ga-72	.043	5×10^{-5}	.00086	3×10^{-1}	.00000				
As-76*	.45	2×10^{-5}	.02250					2×10^{-2}	.00002
Sr-89*	.0031	7×10^{-5}	.00004	7×10^{-6}	.00044				
Sr-90*	.001	1×10^{-4}	.00001	8×10^{-8}	.01250				
Sr-91	.036	6×10^{-5}	.00060	8×10^{-5}	.00045				
Mo-99	.017	3×10^{-4}	.00006	5×10^{-1}	.00000				
Ag-111	.0041	5×10^{-5}	.00008						
Cd-115	.0065	2.5×10^{-5}	.00026						
I-131*	.003	.003	.00000			6×10^{-6}	.00050		
I-133	.036	6×10^{-4}	.00006			1×10^{-5}	.00360		
I-135	.013	4×10^{-4}	.00003			4×10^{-5}	.00032		
Ba-140*	.0037	3×10^{-5}	.00012	5×10^{-5}	.00007				
La-140	.0031	3×10^{-5}	.00010	3×10^{-2}	.00000				
Ce-143	.0012	4×10^{-5}	.00003	2×10^{-1}	.00000				
Sm-153	.0033	8×10^{-5}	.00004	1.8	.00000				
Np-239*	.87 (1.67)**	2×10^{-4}	.00435	1×10^{-1}	.00001				
(RE+Y)* - (La, Ce, Sm)	.223	5×10^{-5}	.00446	9×10^{-2}	.00000				
Total isotopic	.228		.04499		.01759		.00498		.00120
Total β	5.072**								
Gross β	6.45								
Gross α	<.0015								
Per cent MPE (max. perm. exposure)			4.5%		1.7%		0.5%		0.1%

* Routine analyses in Pasco river samples, July, 1958

** Activity density seen by the beta proportional counters

MPC - Maximum permissible concentration for continuous public consumption (1/10 HW-25457 REV1)

Table 2. Radioisotopes Measured Specially, in Columbia River Water at Pasco, Converted to July, 1958

Isotope	Measured Concentration	Date Sampled	River Flow* on Date Sampled	River Flow Ratio	Concentration Converted to July
	$\mu\text{c/cc}$		gps		$\mu\text{c/cc}$
La-140	9.6×10^{-9}	1-21-58	4.79×10^5	.326	3.1×10^{-9}
Ce-143	3.6×10^{-9}	1-21-58	4.79×10^5	.326	1.2×10^{-9}
Sm-153	1.0×10^{-8}	1-21-58	4.79×10^5	.326	3.3×10^{-9}

* Average river flow for July, 1958, was 14.7×10^5 gallons per second.

Table 3. Radioisotopes Measured at Other Locations and Times, and Converted to Columbia River at Pasco for July, 1958

Isotope	Radioactive Half-life (Tr), hrs	Concentration Upstream* Units of $10^{-6} \mu\text{c/cc}$	Decay Correction $e^{-.693(13.5) \frac{\text{Tr}}{\text{Tr}}}$	Concentration at Pasco in July, Units of $10^{-6} \mu\text{c/cc}$
Si-31	2.62	1.39	.0280	.039
Mn-56	2.58	18.9	.0270	.510
Co-60	45,600	.0012	.999	.0012
Zn-69	14	.498	.514	.256
Ga-72	14.1	.084	.515	.043
Sr-91	9.7	.094	.385	.036
Mo-99	67	.020	.869	.017
Ag-111	180	.0043	.949	.0041
Cd-115	54	.0077	.842	.0065
I-133	21	.054#	.640	.036
I-135	6.7	.051#	.249	.013

* In Columbia River at mid-reactor location, 4 hours after leaving reactors

** Average travel time to Pasco at July river flow = 15.5 hours ~2 hours effluent already in river

From I-131 in July at Pasco converted for decay to 4 hours and by I-131: I-133: I-135 fission ratios of 24:410:389

For calculation of decay to Pasco, travel time was read from Figure 1, which was prepared by J. K. Soldat, Regional Monitoring Operation, from numerous measurements of times for floats to travel from the various reactor areas to Pasco. The average travel time from all reactors at a wide range of river flows is within about 1% of the travel time from 100-K Area, which is also the largest contributor of radioisotopes to the river; this is called the "mid-reactor location" in this report. Decay calculated from the mid-reactor location is considered not significantly different from decay calculated from each reactor, based on previous studies (e.g., Ref. 3, Appendix C).

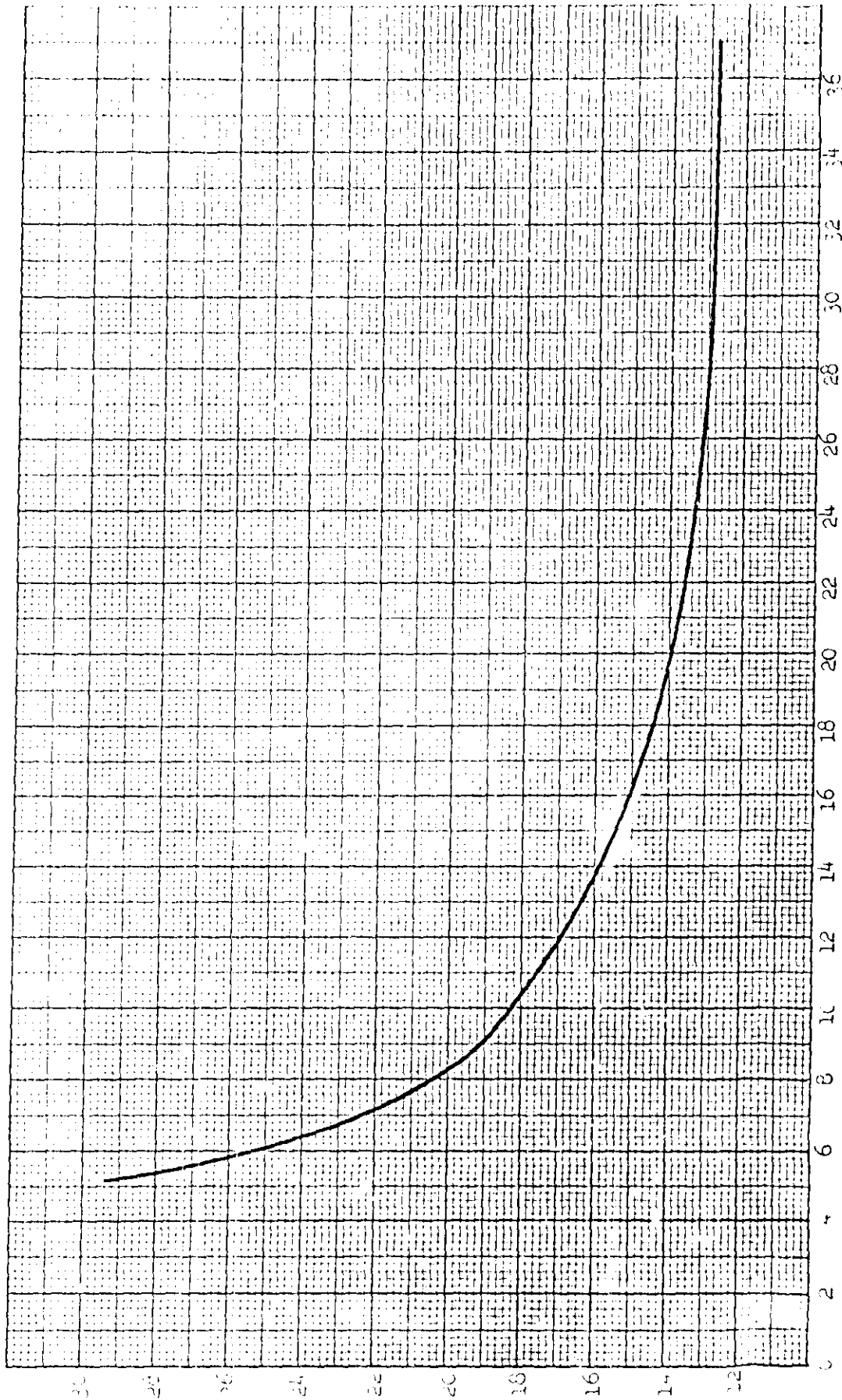
From the data in Table 1, the total measured isotopic activity density is 7.23×10^{-6} $\mu\text{c/cc}$. The total beta activity density is 5.07×10^{-6} $\mu\text{c/cc}$ and the measured gross beta activity density is 6.45×10^{-6} $\mu\text{c/cc}$, leaving an "unaccounted fraction" of 1.38×10^{-6} $\mu\text{c/cc}$. As usual with such a mixture of radioisotopes in drinking water, the gastrointestinal tract was the most critical organ, and the calculated potential exposure for July was 4.5% of the maximum permissible for persons outside controlled areas such as the Hanford project.

Depletion, other than decay, is not factored into the data in Table 3, because depletion factors are available for only one or two of the isotopes. However, depletion factors reported in reference 5 for 10 isotopes in six samples ranged from 17 to 56% with an average of 35%. If it is assumed that this average factor is valid for the isotopes in Table 3, then recalculation of GI tract exposure in Table 1 using the depleted values gives ~~3.3%~~ 4.5% MPE, instead of 4.5% MPE.

Determining Isotopes to be Measured

The recent adaptation of gamma spectrometry to radioanalyses of water samples, in combination with more sensitive procedures for specific beta- and alpha- emitting radioisotopes, has made possible the evaluation of drinking-water exposure based on more frequent and rapid routine measurements of the isotopes contributing most to exposure. These are not necessarily the isotopes of highest concentration nor even the highest beta-emitters, since the relative exposures depend on factoring in the respective MPC's which are independent of concentrations in the water. In Table 4, the isotopes listed in Table 1 are re-arranged in order of the 15 highest in concentration, the 15 highest seen by the beta proportional counters, and the 15 contributing most to exposure.

On the basis of similar previous evaluations⁽⁹⁾, the radioanalysis laboratory has been asked to add analyses for Zn-69, Ga-72, Y-93, La-140, Eu-152 (9.3 hour) and Sm-153. Future evaluations will undoubtedly indicate that the list should be changed, adding high-ranking isotopes and discontinuing others. For example, Mn-56 is usually high in the spring months, and probably should be analyzed routinely during such periods, because of its appreciable contribution to upstream GI tract exposure and uncertain corrections without it. However, its short half-life (2.58 hours) ordinarily makes it insignificant in Pasco-Kennewick sanitary water exposures, so that it need not be analyzed in such samples.



RIVER FLOW - Units of 10^5 gallons per second

Figure 1 - Travel Time from Mid-Reactor Location to Pasco vs Columbia River Flow

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Table 4. Rankings of the Fifteen Isotopes Contributing Most to Total Activity,
"Beta" Count and GI Tract Exposure

<u>Total Activity</u>		<u>"Beta" Count</u>		<u>GI Exposure</u>	
<u>Isotope</u>	<u>Units of</u> <u>10⁻⁶ µc/cc</u>	<u>Isotope</u>	<u>Units of</u> <u>10⁻⁶ µc/cc</u>	<u>Isotope</u>	<u>G/MPC</u>
Cr-51	2.37	Np-239	1.67	As-76	.02250*
Cu-64	1.28	Na-24	.895	RE+Y-(La,Ce,Sm)	.00446*
Na-24	.895	Cu-64	.742	Np-239	.00435*
Np-239	.87	Mn-56	.510	Zn-69	<u>.00366 751</u>
Mn-56	.510	As-76	.45	Cu-64	.00256*
As-76	.45	Zn-69	.256	Mn-56	.00170
Zn-69	.256	RE+Y-(La,Ce,Sm)	.223	Cr-51	.00118*
RE+Y-(La,Ce,Sm)	.223	Cr-51	.045	Na-24	<u>.00112*903</u>
Zn-65	.096	Ga-72	.043	Ga-72	.00086
Ga-72	.043	Si-31	.039	Sr-91	<u>.00060 951</u>
Si-31	.039	Sr-91	.036	Zn-65	.00048*
Sr-91	.036	P-32	.033	P-32	.00041*
P-32	.033	Sc-46	.018	Cd-115	<u>.00026 984</u>
Sc-46	.018	Mo-99	.017	Sc-46	.00020*
Mo-99	<u>.017</u>	I-133	<u>.015</u>	Ba-140	<u>.00012*994</u>
Total 15 isotopes	7.136		4.992		.04446
Total all isotopes	7.228		5.072		.04499

* Routine weekly analyses of river water for isotopes in last column, plus Ca-45, Fe-59, Sr-89, Sr-90, I-131, gross alpha, gross beta, and the rare earth + yttrium (RE+Y) group. When individual rare earth isotope results are available, they are subtracted from the RE+Y group concentration and evaluated separately, and the MPC for the RE+Y group is altered for the deletions⁽⁸⁾.

Ranking the isotopes in order of their contributions to exposure, not only of the GI tract but also bone, thyroid, kidneys, total body, gonads, etc., provides the key to analyses needed for optimum evaluations. Optimizing requires consideration of the relative exposure contributed by this source to total exposures from all radiation sources in the environment, the capacity and quality of analytical facilities and procedures, the ability to keep up with evaluations, the costs of sampling, analysis, and evaluations, and the money allocated to such a program. Since evaluation requires more than just gross beta and gross alpha determinations, the goal of such a program should be isotopic evaluation of at least 75% of the potential exposure with auxiliary measurements indicating what the percentage probably is.

As illustrated in Table 4, the program being developed for evaluation of Hanford environmental exposures from public drinking water sources will account for 95-99% of the potential exposure. A minor correction to 100% could be included in the exposure calculation, but is not justified for such a small error considering the inaccuracies of sampling, analyses, and MPC's and other factors affecting total exposure from all sources.

As data continue to accumulate from this program, it should be possible to decrease the frequency of analyses for several of the isotopes. As Healy pointed out and started earlier⁽²⁾, correlations are needed of isotopic concentrations with river flow, turbidity, temperature as it affects aquatic metabolism, reactor power levels and water treatment, and sanitary water treatment. Isotopes showing low fluctuations or good correlations might be measured only once or twice a year. Others may require quarterly or monthly analyses, while some will still merit weekly determinations. Those with long radioactive half-lives are better sampled continuously, with composite aliquots analyzed at intervals dependent on the half-lives.

The analytical sensitivities desired are discussed in reference 2.

It is very important to continue preventive procedures against the oversight of any unmeasured isotope which contributes significantly to exposure. With the hundreds of isotopes in reactor effluent water, and frequent operating changes which affect their output, particular isotopes may become significant. Routine measurement and comparison of gross activity density is one such procedure; major sustained increases in gross activity values should signal intensified search for isotopes not routinely measured which account for the increases.

The second preventive procedure which guards against oversight and helps explain unusual gross activity increases is the isotopic analysis of reactor effluent water where the concentrations are greater and decay time is shortened. The present program of analyses of one or two samples per month from each reactor for 21 isotopes and groups is being expanded by the Irradiation Processing Department to include analyses for about 35 isotopes on weekly, monthly and annual frequencies, plus continual search for other important isotopes.

Elimination of the "Unaccounted Fraction"

The arithmetical difference between the gross beta activity density of a mixture of radioisotopes and the sum of the "beta" radiation emitted by those isotopes which have been individually determined has been called the "unaccounted fraction". With knowledge of all isotopes that could be present in the mixture, a maximum permissible concentration for the "unaccounted fraction" can be chosen from the list of MPC's for the unmeasured isotopes.

Ranking the isotopes in order of their contributions to exposure, not only of the GI tract but also bone, thyroid, kidneys, total body, gonads, etc., provides the key to analyses needed for optimum evaluations. Optimizing requires consideration of the relative exposure contributed by this source to total exposures from all radiation sources in the environment, the capacity and quality of analytical facilities and procedures, the ability to keep up with evaluations, the costs of sampling, analysis, and evaluations, and the money allocated to such a program. Since evaluation requires more than just gross beta and gross alpha determinations, the goal of such a program should be isotopic evaluation of at least 75% of the potential exposure with auxiliary measurements indicating what the percentage probably is.

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As the Hanford environmental water monitoring program was developing, it was advisable to use the "unaccounted fraction" and its chosen MPC in exposure evaluations involving insufficient isotopic analyses or extrapolations from remote measurements^(1,2,3). There is a large number of isotopes known to be present initially from analyses of the reactor effluent water. To date, analyses have been made for 99 isotopes, positive measurements were obtained for 61, and there are at least a hundred more fission products which must have been in the reactor effluent although insignificant to exposure because of low fission yield or short half-life. A few years ago, only a few of these, of highest concentrations, were analyzed in the river or sanitary waters. More recently, not only have concentrations increased but there have been major improvements in analytical instruments and procedures. At the present time, more than 20 isotopes and groups are routinely measured in river and sanitary waters, and methods are available to perhaps double or triple this number routinely if justified for exposure evaluation purposes.

Since most of the isotopes in river and sanitary waters emit gamma radiation, use of the gamma spectrometer to scan a wide energy range insures that most of the isotopes contributing significantly to exposure are measured. Even though others of low energy or concentration are present, the additional effort to measure them routinely would increase the total exposure accounted for only a few per cent.

The beta proportional counter used for river and sanitary water analyses counts not only some alpha and more "soft" beta and X-radiation than the GM counters used for reactor effluent analyses, because of a thinner window, but also more gamma radiation because of higher internal gas pressure⁽¹⁰⁾. Therefore, the gross beta measurement is only a relative value if adequately standardized. Furthermore, some isotopes (e.g., Cr-51) which emit no beta but only gamma and internal conversion X-rays are counted and must be included in the summation of total beta for comparison to gross beta. Some isotopes (e.g., Co-64 and Zn-65) produce only a fractional number of "beta" counts per disintegration, so that the value in the total beta column is only a fraction of the microcurie value. The decay of Np-239 actually produces 1.92 times as many "beta" counts as disintegrations, so the actual microcurie value is about one-half the value included in total beta.

The inaccuracies in gross beta measurements in a routine program are of no consequence if the procedure is standardized, and if the values are not relied upon and used in evaluations of exposure.

These inaccuracies result mainly from the necessity, in the routine program involving hundreds of samples per month, of using average factors for converting from the observed counting rate to microcurie values, based on past isotopic and energy measurements, rather than conversion factors for the specific sample. The derivation of conversion factors and the accuracy of gross beta determinations are more fully discussed in reference 10. It would be possible, though costly, to have the calculations for every sample performed by a data processing machine such as the IBM-702. Although the "unaccounted fraction" would then be more accurate, the exposure evaluation would still depend on a conservatively chosen MPC for the "unaccounted fraction".

If statistical variations were determined routinely for the gross beta measurement, and for the sum of the individual isotopic measurements, it is quite likely that there would be an overlap of the 1 increments. Reference 3 contains data which supports this inference, and subsequent examinations of such data also indicate that the differences are not statistically significant. The present gross beta microcurie values are estimated⁽¹⁰⁾ to have an accuracy for a single sample of $\pm 100\%$, $\pm 50\%$. Such accuracy is generally sufficient to indicate trends, and investigate geographical distributions, and should continue to be improved where possible.

to insure the highest consistency among measurements of environmental sources. For the four or five values averaged per month for each kind of sample, the accuracy is estimated at about $\pm 10\%$ (10), which still results in a large uncertainty in the exposure evaluation if the "unaccounted fraction" and its conservative MPC are included. The inaccuracy would be still smaller when the running 12-month average exposure evaluation is instituted, using 50 to 60 measurements of isotopes and gross beta. This would not justify inclusion of the "unaccounted fraction", just because the inaccuracy was smaller, in the face of improved accounting for the principal isotopic contributors.

In the reactor effluent analysis program which began more than ten years ago, there are within the last year about as many negative "unaccounted fractions" as positive differences between gross beta and total beta values. Negative differences for river and sanitary water samples to date are few, despite correction factors derived for each type of sample. Further river and sanitary water results may provide refinements of the correction factors, such that eventually an appreciable number of negative "fractions" would be observed.

For this study, the best possible gross beta microcurie values for a sample each of river water at Pasco and 100-F Area were calculated by W. Y. Matsumoto, Radiological Chemical Analysis Laboratory. Special factors were included for transfer losses and the beta energy corrections were based on the respective isotopic analyses. Corrections for decay between sampling and analysis were based on the specific decay curves for each sample. The recalculated gross beta values, refined by every available correction, are compared with the original routine values in Table 5.

Table 5. Comparison of Routine and Refined Gross Beta Concentrations

<u>River Sample</u>	<u>Original Gross Beta</u>	<u>Recalculated Gross Beta</u>
100-F Area (7-21-58)	$2.70 \times 10^{-5} \mu\text{c/cc}$	$2.19 \times 10^{-5} \mu\text{c/cc}$
Pasco (7-22-58)	$7.40 \times 10^{-6} \mu\text{c/cc}$	$6.76 \times 10^{-6} \mu\text{c/cc}$

The total beta activity density of the individual isotopes measured in the Pasco river sample of 7-22-58 was $4.11 \times 10^{-6} \mu\text{c/cc}$, leaving an "unaccounted fraction" of $2.65 \times 10^{-6} \mu\text{c/cc}$. Using the methods of Tables 2 and 3, the beta concentrations of the next 15 radioisotopes contributing the most exposure were then calculated, and added to the measured isotopes. The total of all these isotopes is $5.40 \times 10^{-6} \mu\text{c/cc}$, which when subtracted from the recalculated gross beta above still leaves an "unaccounted fraction" of $1.36 \times 10^{-6} \mu\text{c/cc}$. For this sample and for practically all river and sanitary water samples it is impossible to analyze at this time for enough isotopes to eliminate the "unaccounted fraction". Even additional conversions from reactor effluent analyses would be insufficient and there would still be little improvement in the exposure evaluation.

Because of the improvements which permit isotopic analyses of the actual drinking water to account for a large percentage of the potential exposure, and because of the inaccuracy of gross beta determinations, the use of "unaccounted fraction" and its chosen MPC in evaluation of exposure from drinking water taken from the Columbia River is being discontinued. Gross beta measurements will be continued and improved to indicate trends, and the "unaccounted fraction" will be observed to help insure accounting for significant exposures. The special searches of reactor effluent, river, and sanitary waters for significant isotopes not only will be continued but

expanded to assure that no isotope contributing significantly to exposure is overlooked. Routine analyses of those isotopes which will indicate more than 75% and preferably 90-99% of the potential exposure from the various drinking water supplies will be made.

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