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RADIOLOGICAL SIGNIFICANCE
OF 100-N SANITARY WATER

MARCH 1971

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PACIFIC NORTHWEST LABORATORIES DIVISION
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RADIOLOGICAL SIGNIFICANCE
OF 100-N SANITARY WATER

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March 1971

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ABSTRACT

Various methods have been compared for estimating GI Tract dose from radionuclides in drinking water. Estimates based on "total beta" measurements appear to be more accurate than those based on the measured concentration of a single radionuclide. A recommended method of making "total beta" dose estimates is described.

NOTE

This report was prepared before the KE production reactor shutdown was announced. Although the shutdown virtually eliminated unnatural radioactivity in 100-N sanitary water (and therefore the need for GI Tract dose estimation), this report remains unchanged in order to describe what existed and what was planned prior to the shutdown.

RADIOLOGICAL SIGNIFICANCE OF 100-N SANITARY WATER

INTRODUCTION

Since the startup of 'N' reactor in 1963, sanitary water supplied by the 100-N water treatment plant has been used by 100-N and WPPSS personnel for drinking and other purposes. Because the raw water is taken from the Columbia River not far downstream from an operating production reactor, it contains some radioactivity. While processing the raw to sanitary water removes much of the radioactivity, some remains to contribute slightly to the radiation exposure of those drinking it.

Columbia River water is pumped to the 183-N filter plant, where chemical treatment, flocculation, and filtration make it suitable for process applications. From the filters, water flows by gravity to a 4000,000 gallon clearwell and is then pumped to the filtered water storage tank near the 183-N pump building. A portion of this water is further chlorine-treated for sanitary water usage at the 100-N and WPPSS facilities.

Use of Columbia River water removed not far downstream from operating reactors has not been unique to 100-N. In the past, each reactor area processed its own sanitary water, and 100-D Area for many years was a major supplier of export water used for sanitary water and other purposes in the 200 Areas. Even with all reactors operating, however, the GI Tract dose received from drinking 100 Area sanitary water was only about 2% of the standard (1500 mrem/year) for individuals in uncontrolled areas. The dose received by Hanford plant workers from drinking sanitary water at all 100 Areas (except for 100-BC, the most upstream reactor area), was estimated from routine analysis of 100-D sanitary water.

Prior to 1964, the GI Tract dose rate was obtained by comparing the concentration of various radionuclides in sanitary water with appropriate maximum permissible concentrations. This method was abandoned in 1964 when it was found that frequent measurement of total beta activity was a better indicator of Richland and Pasco GI Tract Dose than infrequent measurement of the individual

radionuclides. (Reference) In 1966, detailed radionuclide measurements were combined with total beta measurements to form the presently used procedure in which frequent total beta measurements are used to normalize dose calculated monthly from measured radionuclide concentrations.

Following construction of WPPSS facilities, an extensive sanitary water sampling and analysis program was initiated to measure more accurately the exposure of WPPSS personnel, who were assumed to be members of the general public. Beginning in January 1968, monthly sanitary water grab samples were analyzed for 16 radionuclides and weekly integrated samples were analyzed for 4 radionuclides. Grab samples taken as frequently as three per week were analyzed for total beta activity. Dose was calculated using appropriate dose conversion factors, assuming an average water ingestion rate of 0.93 liters per day. It was evident here, as at other reactor areas, that GI Tract dose was more significant (i.e., a larger fraction of the appropriate limit) than dose to the whole body or to any other organ. Therefore, only dose to the GI Tract has been routinely calculated. Now that nearly three years of radiation exposure information has been accumulated for those drinking 100-N sanitary water, and now that only one reactor (KE) remains operating upstream of 100-N, it seems appropriate to determine if such an extensive program is still warranted. That is the purpose of this study.

SUMMARY AND CONCLUSIONS

The shutdown of all but one (KE) reactor upstream from 100-N has considerably reduced the GI Tract dose from drinking 100-N sanitary water. It is apparent that the remaining exposure results from only a few radionuclides and that dose can be estimated in various ways.

The most accurate alternative to the present method is to estimate dose by using the total beta activity measured in periodic grab samples. Since GI Tract dose is of interest and total beta activity measurable (by normal methods) only when KE is operating, the grab samples should be obtained with KE reactor operating. Suitable corrections should then be made for the reactor's operating history during each month. It is recommended that GI Tract dose be calculated

as follows:

$$\text{GI Tract Dose (mrem/month)} = \frac{\text{Average Total Beta Activity (c/m/ml)}}{82 \text{ c/m/ml per mrem/l}}$$

$$\times 19.4 \text{ l/month} \times \frac{T_k}{T_m}$$

Here: T_k is days of KE operation during month.

T_m is length of month in days.

Since this method tends to underestimate the dose from Mn-56, that radionuclide should continue to be measured and suitable dose corrections made during those months when its concentration is high.

ROUTINE DOSE ESTIMATES - 1968 TO 1970

The average GI Tract dose from drinking 100-N sanitary water is estimated to have been 53, 29, and 9.5 mrem during 1968, 1969, and 1970, respectively. Using 1969 as an example, Table 1 demonstrates, as stated earlier, that the GI Tract is the critical organ for ingested 100-N sanitary water. It also demonstrates that exposure of the various organs results almost entirely from a few radionuclides. This fact is more apparent in Table 2, in which all the measured radionuclides are listed in decreasing order of GI Tract dose contribution. The table also shows that, with an occasional exception, the relative dose contribution of these radionuclides remains reasonably constant. The greatest exception is Mn-56, which sometimes becomes quite significant during the spring flood. Another exception to this occurs when the reactors are shut down; however, at such times the GI Tract dose rate is low and the change in relative dose contribution is therefore insignificant.

DOSE ESTIMATION BY OTHER METHODS

The GI Tract dose contribution of eight radionuclides is plotted in Figures 1 and 2. Of these eight radionuclides, Na-24 and Np-239 were investigated as possibly reliable indicators of dose. Sodium-24 is a rather interesting prospect for this purpose, since it contributes on the average about 25% of the

TABLE 1

ORGAN DOSE COMPARISON - 1969
(mrem/yr)

<u>Major Contributors</u>	<u>GI Tract</u>	<u>Bone</u>	<u>Total Body</u>	<u>Thyroid (Adult)</u>
²⁴ Na	5.4	1.3	1.3	1.3
³² P	-	<0.4	-	-
⁵⁶ Mn	15.8	-	-	-
⁶⁴ Cu	1.3	-	-	-
⁷² Ga	0.9	-	-	-
⁷⁶ As	0.6	-	-	-
¹²² Sb	1.2	-	-	-
¹³¹ I	-	-	-	<1.3
¹³³ I	-	-	-	5.4
²³⁹ Np	2.9	-	-	-
RE+Y*	0.5	-	-	-
Other Radio-nuclides	-	0.1	0.1	0.2
<hr/>				
Total Dose**	28.6	1.8	1.4	8.2
Percent of Limit***	1.9	0.12	0.28	0.55

* Rare earths plus yttrium are considered to be a single radionuclide for GI Tract dose estimation purposes.

** Total dose from these major contributors. The contribution by other measured radionuclides was very small.

*** Limits Used:

Total Body - 0.5 rem/yr
Other Organs - 1.5 rem/yr

TABLE 2

CONTRIBUTION OF VARIOUS RADIONUCLIDES TO 100-N GI TRACT DOSE

(Listed in decreasing order of contribution.) - From 'N'
Sanitary Grab analyses except where noted by *.

Sample Date →	January 8, 1969			February 5, 1969			March 5, 1969			April 9, 1969		
Order	Nuclide	mrem/l	%	Nuclide	mrem/l	%	Nuclide	mrem/l	%	Nuclide	mrem/l	%
1	²⁴ Na	0.047	27.8	²⁴ Na	0.012	21.8	²⁴ Na	0.020	16.3	⁵⁶ Mn	0.425	94.0
2	²³⁹ Np	0.025	14.8	²³⁹ Np	0.0080	14.5	⁵⁶ Mn	0.068	55.3	²⁴ Na	0.0076	1.7
3	⁵⁶ Mn	0.060	35.5	⁵⁶ Mn	0.021	38.2	²³⁹ Np	0.012	9.8	²³⁹ Np	0.0056	1.2
4	¹²² Sb	0.012	7.1	⁷² Ga	<0.0037	6.7	¹²² Sb	0.0044	3.6	⁶⁴ Cu	0.0039	0.9
5	⁶⁴ Cu	0.0097	5.7	¹²² Sb	0.0029	5.3	⁶⁴ Cu	0.0042	3.4	⁷² Ga	<0.0037	0.8
6	⁷² Ga	0.0044	2.6	⁶⁴ Cu	0.0023	4.2	⁷² Ga	<0.0037	3.0	¹²² Sb	0.0021	0.5
7	RE+Y	0.0041	2.4	RE+Y	0.0010	1.8	RE+Y	0.0035	2.8	⁴⁶ Sc	<0.0010	0.2
8	⁵¹ Cr*	0.0027	1.6	⁷⁵ As	0.0010	1.8	⁵¹ Cr*	0.0025	2.0	RE+Y	0.0008	0.2
9	⁷⁶ As	0.0020	1.2	⁴⁵ Sc	<0.0010	1.8	⁴⁶ Sc	0.0022	1.8	⁷⁶ As	0.0008	0.2
10	⁴⁶ Sc	0.0012	0.7	⁵¹ Cr*	0.0008	1.4	⁷⁶ As	0.0012	1.0	⁵¹ Cr*	0.0005	0.1
11	⁶⁵ Zn*	0.0008	0.5	⁶⁵ Zn*	0.0005	0.9	⁶⁵ Zn*	0.0012	1.0	^{69m} Zn	<0.0003	0.07
12	^{69m} Zn	0.0005	0.3	^{69m} Zn	0.0004	0.7	^{69m} Zn	0.0005	0.4	⁶⁵ Zn*	<0.0002	0.04
13	³² P	0.0004	0.2	⁹⁵ Zr-Nb	<0.0002	0.4	⁹⁵ Zr-Nb	<0.0002	0.2	⁹⁵ Zr-Nb	<0.0002	0.04
14	¹³³ I	0.0003	0.2	³² P	0.0001	0.2	³² P	<0.0001	0.08	³² P	<0.0001	0.02
15	⁹⁵ Zr-Nb	<0.0002	0.1	¹³³ I	0.00007	0.1	¹³³ I	0.0001	0.08	¹³³ I	0.00004	0.01
16	¹³¹ I	0.00002	0.005	¹³¹ I	0.000006	0.01	¹³¹ I	0.000003	0.002	¹³¹ I	<0.000003	0.001
TOTAL		<0.169	~100		<0.055	~100		<0.123	~100		<0.452	~100

* Integrated Sample

TABLE 2 (Continued)

Sample Date →	May 7, 1969			June 4, 1969			July 9, 1969			August 6, 1969		
Order	Nuclide	mrem/l	%	Nuclide	mrem/l	%	Nuclide	mrem/l	%	Nuclide	mrem/l	%
1	⁵⁶ Mn	0.161	88.0	⁵⁶ Mn	0.066	77.6	⁷² Ga	<0.0037	16.8	²⁴ Na	0.017	28.8
2	²⁴ Na	0.0065	3.6	²⁴ Na	0.0052	6.1	⁵⁶ Mn	0.010	45.4	²³⁹ Np	0.0098	16.6
3	²³⁹ Np	0.0043	2.3	²³⁹ Np	0.0038	4.5	²⁴ Na	0.0021	9.5	¹²² Sb	0.0050	8.5
4	⁷² Ga	<0.0037	2.0	⁷² Ga	<0.0037	4.4	²³⁹ Np	0.0014	6.4	⁵⁶ Mn	0.016	27.1
5	¹²² Sb	0.0037	1.7	¹²² Sb	0.0024	2.8	⁴⁶ Sc	<0.0010	4.5	⁷² Ga	0.0037	6.3
6	⁴⁶ Sc	<0.0010	0.5	⁶⁴ Cu	0.0016	1.9	¹²² Sb	0.0008	3.6	⁶⁴ Cu	0.0026	4.4
7	⁶⁴ Cu	0.0010	0.5	⁴⁶ Sc	<0.0010	1.2	⁶⁴ Cu	0.0006	2.7	⁴⁶ Sc	0.0011	1.9
8	⁷⁶ As	0.0009	0.5	RE+Y	0.0005	0.6	⁷⁶ As	0.0004	1.8	RE+Y	0.0009	1.5
9	RE+Y	0.0008	0.4	⁷⁶ As	<0.0004	0.5	RE+Y	0.0002	0.9	⁵¹ Cr	0.0009	1.5
10	^{69m} Zn	<0.0003	0.2	^{69m} Zn	<0.0003	0.4	⁶⁵ Zn*	<0.0002	0.9	⁷⁶ As	0.0006	1.0
11	⁶⁵ Zn*	<0.0002	0.1	⁵¹ Cr*	0.0002	0.2	⁹⁵ Zr-Nb	0.0002	0.9	⁶⁵ Zn*	0.0002	0.3
12	⁵¹ Cr*	0.0002	0.1	⁶⁵ Zr*	<0.0002	0.2	⁵¹ Cr	0.0002	0.9	⁹⁵ Zr-Nb	<0.0002	0.3
13	⁹⁵ Zr-Nb	<0.0002	0.1	⁹⁵ Zr-Nb	<0.0002	0.2	³² P*	<0.0001	0.4	³² P*	0.0001	0.2
14	³² P	<0.0001	0.05	³² P	<0.0001	0.1	¹³³ I	0.00001	0.04	¹³³ I	0.0001	0.2
15	¹³³ I	0.00005	0.03	¹³³ I	0.00002	0.02	¹³¹ I	<0.00003	0.01	¹³¹ I	<0.000003	0.005
16	¹³¹ I	<0.000003	0.002	¹³¹ I	<0.000003	0.004						
TOTAL		<0.183	~100		<0.085	~100		<0.022	~100		<0.059	~100

TABLE 2 (Continued)

Sample Date →	September 10, 1969			October 8, 1969			November 5, 1969			December 3, 1969		
Order	Nuclide	mrem/l	%	Nuclide	mrem/l	%	Nuclide	mrem/l	%	Nuclide	mrem/l	%
1	²⁴ Na	0.042	29.0	²⁴ Na	0.025	33.3	²⁴ Na	0.022	25.6	²⁴ Na	0.033	42.3
2	²³⁹ Np	0.016	11.0	²³⁹ Np	0.014	18.7	²³⁹ Np	0.011	12.8	²³⁹ Np	0.016	20.5
3	⁵⁶ Mn	0.062	42.8	¹²² Sb	0.0053	7.1	⁵⁶ Mn	0.038	44.2	⁶⁴ Cu	0.0066	8.5
4	¹²² Sb	0.0067	4.6	⁶⁴ Cu	0.0049	6.5	¹²² Sb	0.0044	5.1	¹²² Sb	0.0055	7.0
5	⁶⁴ Cu	0.0065	4.5	⁵⁶ Mn	0.017	22.7	⁶⁴ Cu	0.0042	4.9	⁷² Ga	0.0037	4.7
6	⁷² Ga	<0.0032	2.2	⁷⁶ As	0.0028	3.7	RE+Y	0.0016	1.9	RE+Y	0.0023	2.9
7	⁴⁶ Sc	0.0024	1.6	⁷² Ga	<0.0017	2.3	⁵¹ Cr	0.0011	1.3	⁵¹ Cr	0.0016	2.0
8	RE+Y	0.0022	1.5	RE+Y	0.0015	2.0	⁷⁶ As	0.0010	1.2	⁷⁶ As	0.0015	1.9
9	⁷⁶ As	0.0014	1.0	⁴⁶ Sc	0.0015	2.0	⁴⁶ Sc	0.0009	1.0	⁵⁶ Mn	0.006	7.7
10	⁵¹ Cr	0.0014	1.0	⁵¹ Cr	0.0013	1.7	⁷² Ga	0.0006	0.7	⁴⁶ Sc	0.0012	1.5
11	⁶⁵ Zn*	0.0004	0.3	⁶⁵ Zn*	0.0006	0.8	⁶⁵ Zn*	0.0005	0.6	⁶⁵ Zn*	0.0005	0.8
12	³² P*	0.0002	0.1	³² P*	0.0002	0.3	⁹⁵ Zr-Nb	0.0002	0.2	¹³³ I	0.0002	0.2
13	¹³³ I	0.0002	0.1	⁹⁵ Zr-Nb	<0.0002	0.3	³² P*	0.0001	0.1	⁹⁵ Zr-Nb	0.0002	0.2
14	⁹⁵ Zr-Nb	<0.0002	0.1	¹³³ I	0.0001	0.1	¹³³ I	0.0001	0.1	³² P*	0.0001	0.1
15	¹³¹ I	0.000003	0.002	¹³¹ I	<0.000002	0.003	¹³¹ I	0.0000004	0.0005	¹³¹ I	0.00001	0.01
TOTAL		<0.145	~100		<0.075	~100		0.086	~100		0.078	~100

GI Tract dose. Neptunium-239 also looks reasonably good for this purpose, although its average contribution is less than 15% of the total dose. Due to its great fluctuation, Mn-56 would not be at all suitable. The remainder of the eight radionuclides (Sb-122, Ga-72, Cu-64, As-76, and rare earths + yttrium) would not be reliable indicators of dose, since either they contribute so little of the dose or they are difficult to measure. Gallium-72 is especially difficult to measure, and its concentration is typically reported as a "less than" value. This erroneously causes the Ga-72 dose contribution (Figure 2) to appear inversely proportional to total dose.

Table 3 summarizes the average and maximum contributions of these radionuclides to both the radioactivity concentration in 100-N sanitary water and to GI Tract dose. Dose factors used to convert radionuclide intake to GI Tract dose also are shown.

The effective dose factor, obtained by dividing total mrem/l by total pCi/l for each sample, was calculated and plotted as shown in Figure 3. Except for periods during which the upstream reactors were not operating and for periods of high Mn-56 concentration, the effective dose factor remained reasonably constant. With the reactors shut down, as during parts of the winter and spring of 1970, the concentrations and relative contribution of various radionuclides in 100-N sanitary water changes. Short-lived radionuclides such as Mn-56 disappear and the relative dose contribution of such things as Sb-122 and As-76, both of which have large dose factors, increases. During an extended shutdown, these radionuclides also disappear, and the dose (by then insignificant) results from Zn-65 and other longer-lived radionuclides.

Since a major objective of this study was to select a simpler, more economical method of estimating GI Tract dose, dose for the years 1968, 1969, and most of 1970 was calculated using measured Na-24 concentrations and the assumption that Na-24 contributes an average 24.2% of the GI Tract dose. For example, the dose for January 1969 was calculated as follows:

$$\text{GI Tract Dose} = \frac{0.047 \text{ mrem/l} \times 19.4 \text{ l/month}}{24.2\%} = 3.7 \text{ mrem}$$

* From Table 2.

TABLE 3

CONTRIBUTION OF MAJOR RADIONUCLIDES IN 100-N
SANITARY WATER FROM JANUARY 1968 TO OCTOBER 1970*

Radio-nuclide	Percent Contribution To				Dose Factor (mrem/pCi)
	Radioactivity Concentration		GI Tract Dose		
	Average	Maximum	Average	Maximum	
⁵⁶ Mn	28.	89.	40.	94.	18. x10 ⁻⁶ **
²⁴ Na	38.	65.	24.	42.	6.2x10 ⁻⁶
²³⁹ Np	7.9	23.	14.	24.	19. x10 ⁻⁶
¹²² Sb	1.2	2.9	7.6	16.	66. x10 ⁻⁶
⁷² Ga	≤1.0	<5.2	<4.9	<18.	53. x10 ⁻⁶
⁶⁴ Cu	9.6	22.	4.2	8.5	4.6x10 ⁻⁶
⁷⁶ As	0.4	3.0	2.2	7.1	77. x10 ⁻⁶
RE+Y	<u>0.4</u>	1.0	<u>1.8</u>	5.2	47. x10 ⁻⁶
TOTAL	<87.		<99.		

* Several months excluded for lack of data, because no reactors were operating when the sample was obtained, or because Mn-56 analyses were not performed.

** During this study the Mn-56 dose factor in use since 1968 was discovered to be in error. The error originated in the ICRP Committee II report, which improperly listed LLI as the critical portion of the GI Tract. The LLI dose factor used was 4.4x10⁻⁶ mrem/pCi. However, the ULI dose factor is 18.x10⁻⁶ mrem/pCi as shown in this table.

The results are shown in Figure 4 along with doses calculated in the normal manner from the measured concentration of each radionuclide. Obviously, this method is not very accurate when compared with the normal method of dose calculation. (The normal method may also contain inaccuracies, however. For example, it is nearly certain that the high GI Tract dose in October 1968 is erroneous.)

The possibility of using Np-239 as an indicator was then examined in the same manner as for Na-24. The results are shown in Figure 5. Like Na-24, it also does not provide a reliable method.

From the beginning of this study it seemed likely, as R. B. Hall concluded in 1965 concerning Richland and Pasco sanitary water, that total beta activity might well be the best indicator of GI Tract dose. From 1968 to 1970, each 100-N sanitary water sample analyzed for various radionuclides was also analyzed for total beta activity. Figure 6 shows total beta activity of each sample plotted against GI Tract dose (millirem per liter) calculated from the measured radionuclide concentrations in each sample. By the linear least squares method, the equation $y = 1.2 + 68.9X$ was found to fit best the data, if those marked * and + were excluded. The slope of this curve (i.e., c/m/ml/mrem/l) divided into the monthly average total beta activity (c/m/ml) in 100-N sanitary water produces an estimate of monthly GI Tract dose. A simpler curve of slope 82 c/m/ml/mrem/l yields a better estimate of the dose, when compared with the normal method of calculation. Figure 7 shows monthly GI Tract dose calculated in this (latter) manner compared with dose calculated in the normal manner from the measured radionuclide concentrations. Although specific radionuclide concentrations were not measured prior to 1968, total beta activity was measured, and the GI Tract dose for 1966 and 1967 calculated by the above method is also shown in Figure 7. This method more accurately estimates GI Tract dose, as calculated in the normal manner, than either the Na-24 or Np-239 methods mentioned earlier.

The dose calculation methods discussed above are compared on an annual basis in Table 4.

TABLE 4

COMPARISON OF ANNUAL AVERAGE 100-N
GI TRACT DOSE (mrem) CALCULATED BY SEVERAL METHODS

<u>Year</u>	<u>Method</u>			
	<u>Normal*</u>	<u>Total Beta</u>	<u>²⁴Na</u>	<u>²³⁹Np</u>
1968	53.**	26.	27.	31.
1969	29.	21.	19.	17.
1970	9.5	11.	5.8	4.7

* Calculated from measured radionuclide concentrations

** Includes 19 mrem, believed erroneously high, for the month of October, 1969.

FIGURE 1. CONTRIBUTION TO TOTAL DOSE RATE BY THE MAJOR RADIONUCLIDES

FIGURE 2. CONTRIBUTION TO TOTAL DOSE RATE BY THE MAJOR RADIONUCLIDES

FIGURE 3. EFFECTIVE DOSE FACTORS

FIGURE 4. GI TRACT DOSE FROM DRINKING 100-N SANITARY WATER

FIGURE 5. GI TRACT DOSE FROM DRINKING 100-N SANITARY WATER

FIGURE 6. 100-N DRINKING WATER - TOTAL BETA ACTIVITY VS GI TRACT DOSE RATE**

FIGURE 7. GI TRACT DOSE FROM DRINKING 100-N SANITARY WATER

REFERENCE

R. B. Hall. Use Of Total Beta Counts To Estimate GI Tract Dose Rates,
Battelle-Northwest, Richland, Washington. October, 1965. (Unpublished
data.)

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