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SUBJECT RADIOISOTOPES RELEASED IN DUN LIQUID EFFLUENTS

INTRODUCTION

Per your request, we have summarized for both the single-pass reactors and N Reactor the amount of radioisotopes released in DUN reactor liquid effluents. Wherever possible, the sources of the radioisotope and the important parameters which influence the release of the radioisotope have been identified. Because of the great dissimilarities between the two types of reactors, the data are discussed on an individual basis.

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I. SINGLE PASS REACTORS

A. Quantities of Radionuclides Released

The most radiologically significant isotopes in the single-pass reactor effluent are monitored on a routine basis. Samples for analysis of ^{76}As , ^{32}P , ^{51}Cr , ^{239}Np , and ^{46}Sc are taken five times per week and a weekly composite for analysis of ^{131}I and ^{65}Zn are obtained from the reactor outlet risers. Occasionally other isotopes are measured. Data for the latest year are given in Table I.^(1,2) The daily averages in terms of picocuries per milliliter are the average concentrations of the radioisotopes in the undiluted effluent corrected to time of sampling. The daily averages in terms of curies per day and curies per month have been corrected for reactor flow and time-operated efficiency.

Performance And Cost Effectiveness

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An analysis of the K Reactor trench inlet taken on December 12, 1969 (just before the "hot duck" incident) is given in Table II. The results show that the water in the trench is primarily reactor coolant effluent water. Flows to the trench have since been changed so that now a major part of the flow to the trench ^{is from the storage basins which is} has not been passed through the reactor. Note that the ^{trench} analysis is only for the major gamma emitters - a phosphorus analysis was not conducted because of the special separation required. ^{32}P concentration in this sample should be similar to that in reactor effluent, however.

As an indication of the relative magnitudes of trace radioisotope discharged to the Columbia River, the data in Table III are presented. This data, obtained in 1957 and reported on March 1, 1958, ⁽³⁾ is one of the most complete sets of data available. Several less complete analyses have been obtained since then, however, and agreement among the various analyses is fairly good.

B. Source of the Radionuclides

The radioisotopes in the single-pass reactors can be traced to primarily three sources: parent materials in the river water, parent materials added as water treatment chemicals (and impurities), and corrosion of in-reactor components.

The major fraction of the reactor effluent water radioisotopes is produced by (n, γ) reactions on parent materials which enter the reactor in the coolant water. ^(4,5,6) Most of these parent materials are present in the raw river water and are not removed by the water treatment process.

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TABLE I

QUANTITIES OF RADIONUCLIDES RELEASED WITH THE SINGLE-PASS
REACTOR COOLING WATER EFFLUENT, 1969^{1,2}

		Daily Average (pCi/ml)			Daily Average (Curies/Day)			Month's Total (Curies)
		C	KE	KW	C	KE	KW	
As-76	January	95.9	72.8	65.6	29.7	64.1	65.9	4950
	February	107.0	70.7	103.3	41.7	68.6	85.7	5488
	March	154.4	97.0	133.4	69.5	96.0	116.0	8726
	April	199.6	102.0	148.9	147.8	30.6	174.2	10578
	May	--	140.1	182.0	--	135.9	109.2	7598
	June	--	101.0	113.0	--	108.1	101.7	6294
P-32	January	10.7	6.4	8.6	3.3	5.1	8.5	524
	February	15.4	9.4	11.7	6.0	9.1	9.7	694
	March	15.3	10.1	15.0	6.9	10.0	13.1	528
	April	21.5	9.3	14.1	10.5	2.8	16.5	811
	May	--	4.3	15.2	--	4.2	9.1	412
	June	--	4.1	10.7	--	4.4	9.6	420
Cr-51	January	209.0	89.0	136.0	64.8	78.3	134.6	8609
	February	272.0	81.0	114.0	106.1	78.6	94.6	7820
	March	226.0	89.0	129.0	79.7	88.1	112.2	8680
	April	261.0	85.0	159.0	127.9	25.5	186.0	10182
	May	--	120.0	201.0	--	4.2	9.1	412
	June	--	132.0	163.0	--	141.2	146.7	8637
Np-239	January	99.0	62.0	94.0	30.7	54.6	93.1	5530
	February	130.0	74.0	99.0	50.7	71.8	82.2	5732
	March	149.0	96.0	124.0	67.1	95.0	107.9	8370
	April	295.0	61.0	163.0	144.6	18.3	188.7	10548
	May	--	92.0	182.0	--	89.2	109.2	6150
	June	--	101.0	111.0	--	108.1	99.9	6240
Zn-65	January	14.6	10.6	6.9	4.8	9.3	6.8	648
	February	19.9	11.9	10.1	7.8	11.6	8.5	781
	March	18.4	13.3	10.0	8.5	11.8	8.8	902
	April	27.2	11.6	11.8	13.4	3.5	13.9	924
	May	--	10.5	15.8	--	10.2	9.5	611
	June	--	7.5	10.5	--	8.0	9.5	525
I-131	January	0.64	0.79	0.67	0.31	0.70	0.66	52
	February	0.94	0.55	0.44	0.37	0.53	0.37	36
	March	0.94	0.52	0.57	0.42	0.52	0.50	45
	April	1.06	0.44	0.65	0.54	0.13	0.76	43
	May	--	0.48	0.47	--	0.47	0.28	23
	June	--	0.45	0.51	--	0.48	0.46	28
Sc-46	March	7.4	7.7	8.6	3.3	7.6	7.5	570
	April	10.8	6.8	10.1	5.3	2.0	11.8	573
	May	--	7.4	16.2	--	7.2	9.7	524
	June	--	10.1	11.1	--	10.8	10.0	624
Mn-56	April	76,000	17,000	69,000				
	May	--	29,000	53,000				
	June	--	12,400	19,300				

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TABLE I (Cont'd)

		Daily Average (pCi/ml)		Daily Average (Curies/day)		Month's Total (Curies)	AEC Limits
		KE	KW	KE	KW		
As-76	July	121.9±21.0	105.4±18.0	55.2	88.6	4458	Avg - 4049
	August	73.5±19.0	86.9±36.3	52.876	79.1	4089	
	September	97.7±23.7	84.6±56.5	84.1	39.5	3708	
	October	90.6±25.8	124.3±17.9	53.73	84.7	4290	
	November	88.4±36.4	77.4±14.7	46.2	67.0	3396	
	December	82.1±11.3	72.8±8.7	59.6	80.9	4355	
P-32	July	9.4±1.7	10.2±2.7	4.27	8.6	399	306
	August	7.5±1.8	11.7±1.3	5.4	10.6	496	
	September	6.1±2.4	10.5±2.4	5.3	4.3	288	
	October	6.0±1.6	11.3±1.0	3.6	4.1	239	
	November	6.6±1.8	3.5±2.3	3.5	3.0	195	
	December	4.9±1.5	3.1±1.1	3.6	3.5	220	
Cr-51	July	108.3±17.9	140.8±26.0	49.0	118.4	5189	4706
	August	91.5±21.1	137.5±16.0	65.9	125.0	5918	
	September	95.0±22.4	117.3±37.5	81.8	66.4	4446	
	October	83.2±23.3	121.8±19.3	49.3	56.7	3286	
	November	93.8±7.3	101.3±15.3	49.0	87.7	4101	
	December	91.1±6.8	94.2±6.2	66.1	104.7	5295	
Np-239	July	82.8±31.8	86.0±23.9	37.5	72.3	3404	2801
	August	71.0±41.1	81.8±3.2	51.0	74.4	3887	
	September	66.0±8.2	77.1±16.9	56.8	53.9	3321	
	October	41.4±10.3	55.2±9.0	24.5	37.6	1925	
	November	40.6±6.0	44.9±6.2	21.2	38.8	1800	
	December	42.0±9.8	44.4±2.8	30.5	49.3	2474	
Sc-46 (Daily)	July	8.1±3.3	10.9±3.9	3.6	9.2	397	
	August	7.3±3.3	6.9±1.9	5.2	6.3	357	
	September	5.1±1.6	7.5±3.4	4.3	5.3	288	
	October	7.6±2.1	8.13±3.7	4.5	5.5	310	
	November	7.4±2.2	10.5±3.8	3.9	9.1	390	
	December	12.0±9.1	9.9±2.6	8.7	11.0		
I-131	July	1.09	0.46	0.49	0.39	27.3	19.8
	August	0.19	0.52	0.17	0.47	19.8	
	September	0.35	0.36	0.30	0.25	16.5	
	October	0.33	0.35	0.19	0.24	13.3	
	November	0.32	0.34	0.17	0.29	13.8	
	December	0.56	0.45	0.41	0.50	28.2	
Zn-65	July	11.4	6.6	5.2	5.5	332	
	August	10.9	9.6	7.8	8.7	512	
	September	4.2	8.3	3.6	6.7	309	
	October	9.1	9.4	5.4	6.4	366	
	November	11.4	6.8	5.9	5.9	354	
	December	9.9	8.6	7.2	9.6	520	
Sc-46 (Composite)	July	4.8	6.0	2.2	5.0	223	
	August	6.8	5.4	4.9	4.9	304	
	September	11.0	4.6	9.5	3.2	381	
	October	3.8	2.5	2.3	1.7	394	
	November	4.4	7.4	2.3	6.4	261	

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TABLE II. K AREA TRENCH WATER ANALYSES

<u>Radioisotope</u>	<u>Concentration, pCi/ml</u>
^{51}Cr	70.0
^{46}Sc	6.0
^{131}I	0.4
^{76}As	70.0
^{54}Mn	0.5
^{239}Np	30.0
^{65}Zn	7.0
^{24}Na	400.0

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TABLE III. COMPLETE RADIOISOTOPE ANALYSES

Isotope	Half-Life	Concentration	
		Reactor (4 Hours) pCi/cc	River at 100-F Area* pCi/ml
Mn-56	2.6 h	1000	1.4
Cu-64	12.9 h	700	0.92
Na-24	15.0 h	500	6.0
Cr-51	27.0 d	300	10.6
Np-239	2.0 d	300	3.7
As-76	1.0 d	280	2.3
Si-31	2.6 h	170	0.1
Zn-69	14.0 h	70	0.92
Ga-72	14.0 h	35	0.50
Sr-92	2.7 h	30	0.061
U-239	23.0 m	30	4.0
I-130 & 133	12.5 d 21 h	25	0.31
Y-92	3.5 h	20	0.08
Nb-97	72.0 m	20	0.05
Sr-91	9.7 h	17	0.16
Zn-65	365.0 d	15	0.34
P-32	14.0 d	10	0.2362
Y-90	64.0 h	10	
I-135		10	0.14
Y-93	10.1 h	10	0.13
Eu-152	96.0 m	8	0.166
Sm-153	47.0 h	8	0.16
W-187	24.0 h	7	0.06
La-141	3.9 h	7	0.028
Nd-149	1.8 h	4.5	0.0022
K-42	12.4 h	<4.5	
La-140	40.2 h	4.0	0.080
I-132		4.0	0.01
Eu-157	15.2 h	3.0	0.044
Ba-140	12.8 d	2.0	0.045
Mo-99	67.0 h	2.0	0.032
Sm-156	9.4 h	2.0	0.011
Th-232		<1.6	
Sc-46	83.8 d	1.5	0.055
Cd-115	43.0 d	1.3	0.024
Ce-143	33.0 h	1.0	0.029
I-131	8.0 d	1.0	0.0175
Ce-141	32.5 d	1.0	0.016
Pr-142	19.2 h	1.0	0.012
C-14	5730.0 y	0.7	
Nd-147	11.1 d	0.6	0.011
Total Ca(Ca-45)	163.0 d	0.6	0.013
Ag-111	7.5 d	0.4	0.022
Y-91	64.2 h	0.4	0.007
Total Fe(Fe-59)	45.0 d	0.3	0.0198
Sr-89	50.6 d	0.3	0.0126
Mn-54	312.0 d	0.3	0.0057
Zr-95	65.0 d	0.3	0.0054
H-3	12.0 d	10.0	

* Calculated.

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TABLE III. (Cont'd)

Isotope	Half-Life	Concentration	
		Reactor (4 Hours) p/Ci/cc	River at 100-F Area* pCi/ml
Pm-149	53.0 h	0.3	0.0049
Eu-156	15.2 d	0.25	0.005
Pr-145	5.9 h	0.2	0.018
Pm-151	28.0 h	0.2	0.0029
Co-60	5.2 y	0.2	0.002
Na-22		<0.2	
Rb-86		<0.2	
Cs-136		<0.2	
Ge-71		<0.14	
Pr-143	13.7 d	0.1	0.002
Cs-136	13.0 d	0.1	<0.002
Tl-204	3.75 y	0.1	<0.001
Ce-144		<0.1	<0.002
Ni-65		<0.07	
Ge-71	11.0 d	0.05	<0.001
Ru-103	40.0 d	0.05	
Sc-47	3.4 d	0.04	
Fe-55	2.4 y	<0.04	<0.0006
Sr-90(& Y-90)	28.8 y	0.02	0.00037
U-238	4.5x10 ⁹ y	0.015	0.00015
Cs-137	30.0 y	0.01	0.0009
P-33	25.0 d	0.01	<0.012
Sn-113		<0.01	0.001
Sn-125		<0.01	
Sr-85	65.0 d	0.007	
At-211		<0.0014	0.0000004
Ac-227	21.2 y	0.0007	0.000007
Pu-239		<0.00008	
Cm-242		<0.00008	
Am-241		<0.00008	
Ni-63		<0.00003	

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A fraction of the parent materials are absorbed on the aluminum process tubing and ~~slug~~^{fuel} jacket corrosion films and are slowly released subsequently to the water. The relatively high concentration of radioisotopes in the effluent water results from this holdup of parent material and subsequent release of radioactive daughters to the effluent stream. Analyses of corrosion films on fuel elements shows that these films have an inventory of radionuclides equivalent to that discharged during several months of reactor operation.

The radioisotopes produced in large amounts as a result of the addition of water treatment chemicals include ^{72}Ga and ^{153}Sm (about 60 and 50 percent, respectively, due to alum* addition), ^{24}Na and ^{51}Cr (about 25 and 100 percent, respectively, due to $\text{Na}_2\text{Cr}_2\text{O}_7$), and ^{32}P (about one-third of the ^{32}P is produced from sulfur due to the $\text{SO}_4^{=}$). (4)

Aluminum process tubing and slug jacket corrosion may account for major amounts of a few radioisotopes, assuming the corrosion products leave the corrosion film. If all the corrosion products actually entered the water, they would contribute as follows: ^{60}Co (50 to 100 percent), ^{59}Fe (70 to 100 percent), ^{72}Ga (30 to 70 percent), ^{65}Zn and ^{64}Cu (1 to 30 percent), and ^{24}Na (about 4 percent). Since aluminum samples actually increase in weight when exposed to process water, the amount of a given element leaving the corrosion film is certainly in question. On the other hand, since aluminum is the major element in the corrosion film and ^{24}Na is produced by the reaction $^{27}\text{Al} (n, \alpha) ^{24}\text{Na}$, it is possible that ^{24}Na is eluted continually from the film matrix. Thus, aluminum corrosion could release

* Note: Alum (aluminum sulfate) has been replaced with bauxite plus sulfuric acid as a process treatment chemical; however, these values are felt to still hold true.

much more ^{24}Na than would be calculated from the corrosion rate of aluminum (with its equilibrium ^{24}Na content).

A more detailed discussion of the source of the major isotopes of interest is given below.

^{32}P

Approximately one-third of the ^{32}P in the single-pass reactor effluent is formed by the $^{32}\text{S}(n,p)^{32}\text{P}$ reaction and two-thirds by the $^{31}\text{P}(n,\gamma)^{32}\text{P}$ reaction. Very little ^{32}P is formed by the $^{35}\text{Cl}(n,\alpha)^{32}\text{P}$ reaction or from secondary sources of parents such as $^{30}\text{Si}(n,\gamma)^{31}\text{Si} \xrightarrow[2.6 \text{ hr.}]{\beta} ^{31}\text{P}(n,\gamma)^{32}\text{P}$. Phosphorous-32 in the effluent water is in the form of the orthophosphate in an equilibrium mixture of $\text{HPO}_4^{=}$ and H_2PO_4^- .

Addition of phosphates and sulfates to reactor cooling water has verified the relative contributions. ⁽⁵⁾ Irradiations of reactor salts in closed quartz ampoules showed that ^{32}S would produce over 50 times as much ^{32}P as ^{31}P if only once-through irradiation is considered. ⁽⁵⁾ Because this ratio is not true in reactor effluent, this indicates that a significant portion of the ^{32}P originates from parent phosphorus that has absorbed onto the in-reactor films and that the phosphate is more efficiently absorbed than the sulfate. The absorption of phosphates on aluminum is a well-known phenomena which appears to be in operation here. It has been calculated that only 0.3 percent of the influent phosphorus atoms engage in a reaction with the corrosion films. ⁽⁷⁾ Since raw Columbia River water contains only 1-10 ppb P, and 1-5 ppm S, it can be seen that sulfate

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in water

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addition, per se, in our water treatment process has only a minor effect on ³²P formation (alum addition does have beneficial effects as discussed later under parameters).

It should be noted here, for comparison with the N Reactor data discussed later, that the use of deionized water in a single-tube K Reactor test did not reduce the amount of ³²P in the effluent. Apparently the trace amounts of ³¹P in the deionized influent are not efficiently removed.

⁷⁶As

Radioactive arsenic is present in the effluent as a mixture of arsenous acid, H_3AsO_4 (sometimes this is absent), and the arsenates, Na_2HASO_4 and NaH_2AsO_4 . Additional Tests have shown that effluent arsenic is directly proportional to the ⁷⁵As parent in the feed water, and that an in-reactor holdup time of 20 hours occurs. Activation analyses of feed water chemicals showed none which would contribute a significant fraction of the ⁷⁵As; therefore, all the ⁷⁵As originates in the Columbia River, primarily in the Spokane River drainage. For this isotope, also, the use of deionized water had no effect in reducing ⁷⁶As concentrations.

⁶⁵Zn

Zinc is present in the effluent primarily as the cation. It is an impurity in aluminum process tubes and fuel jackets; thus, corrosion of these components plus zinc in the influent water all contribute to radiozinc.

^{51}Cr

The radioactive ^{51}Cr in the effluent exists as a small amount of Cr^{+++} and *^{51}Cr exists almost entirely as Cr^{+3} . All the* an equilibrium mixture of $\text{CrO}_4^{=}$ and $\text{Cr}_2\text{O}_7^{=}$. In the river, ^{51}Cr in the effluent is derived from the sodium dichromate additions which are made to inhibit aluminum corrosion. Reduction of the amount of dichromate in the inlet water has not resulted in a proportional decrease in ^{51}Cr due to absorption on the in-reactor films.

 ^{56}Mn

Manganese-56 exists in the effluent as Mn^{+3} and Mn^{+2} , probably as a complex of the two. Some ^{56}Mn originates from irradiation of impurities in the aluminum tubes and fuel jackets but most originates in the raw water. Parent manganese has been traced to the Spokane River drainage system.

 ^{24}Na

Sodium-24 exists in the effluent as the cation. Several sources of ^{24}Na exist: about four percent occurs from corrosion of the fuel⁽⁴⁾ (somewhat more could occur due to the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reaction), 25 percent is added directly as water treatment chemicals, and the remainder from the water.

 ^{72}Ga and ^{153}Sm

About 60 percent of the ^{72}Ga and 50 percent of the ^{153}Sm are produced from parents introduced as impurities with the water treatment chemicals.

Gallium-72 also originates from irradiation of impurities in aluminum.

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^{65}Ni and ^{60}Co

The majority of these nuclides originate from corrosion of fuel jackets. About one percent nickel is added as an alloying agent in fuel jackets and the cobalt ~~con~~gener is not readily separated from the nickel.

^{239}Np and Fission Products (^{85}Kr , $^{89,90,91}\text{Sr}$, $^{91,92,93}\text{Y}$, ^{99}Mo , ^{131}I , ^{133}Xe ,
 $^{137,138}\text{Cs}$, ^{140}Ba , $^{141,143,144}\text{Ce}$)

Neptunium-239 exists in the effluent primarily as the cation. The ^{239}Np and the fission products originate from uranium in the Columbia River (0.5 ppb) and as trace impurities on fuel jackets. Measurements of ^{239}U concentrations accounted for only 1-6 percent of the ^{239}Np measured; therefore, additional work on in-reactor holdup times is required to clarify mechanisms. (5) It has been shown that once a fission has occurred in uranium absorbed into the films that there is sufficient kinetic energy so that most of the fission products are immediately released to the effluent water; i.e., there is no holdup in the films of the individual fission product isotopes. (5)

C. Parameters Which Affect Effluent Activity

The majority of the parent isotopes contributing to the more significant nuclides in single-pass reactor effluent originate in the Columbia River, are absorbed in the in-reactor films, activated, and are slowly released to the effluent. Other parent isotopes are added with the water treatment chemicals and a few originate as impurities in in-reactor components and are released by corrosion. Thus, the most significant parameters in controlling effluent activity involve either water treatment conditions to reduce

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parent isotopes in the cooling water or to reduce parent isotopes contained as impurities in the chemicals. Other parameters would include water treatment conditions to control the absorption and release of radioactive nuclides from the in-reactor films and to control in-reactor corrosion.

Practically all of the research and development has been devoted to reducing parent nuclides in the influent water or to prevent their absorption into in-reactor films. This R&D effort has involved such things as determining optimum water treatment chemical concentrations, optimum Zeta potential, alternate water treatment processes and chemicals, and additives. In addition, numerous studies were first required to determine the source and mechanisms involved leading to the activation and release of the effluent radionuclides. Results of these tests have been well documented. (5,8,9)

Optimization of the water treatment chemical concentrations has significantly reduced effluent activity. For example, contrary to what might be expected, high (18 ppm) alum feed was found to reduce ^{32}P , ^{76}As , and ^{239}Np , by 60, 75, and 60 percent, respectively, as compared to operation at 6 ppm alum.

The Columbia River water contains about 10 ppm sulfate, 18 ppm alum feed adds 9 ppm sulfate and pH adjustment to 6.6 (normal river water is about pH 8) for fuel jacket corrosion control adds ^{another} 15 ppm sulfate. Thus, about 70 percent of the total sulfate in the influent water is added during water treatment. The higher alum feed was found to more efficiently remove the ^{31}P parent, which is a more important source of ^{32}P due to in-reactor absorption, than at lower alum feed and this more than offsets the increased ^{32}P production from the $^{32}\text{S}(\text{n},\text{p})^{32}\text{P}$ reaction. Similar reductions in effluent activity have been obtained by controlling the higher alum feed by Zeta potential rather than by using a fixed feed rate.

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Alternate water treatment chemicals tested in an effort to reduce impurities added have shown somewhat greater reductions in effluent activity but their cost is prohibitive. For example, flocculation with aluminum nitrate provided a reduction of ^{32P, 70As, and 234mPa by} 75, 75, and 65 percent, respectively, over 6 ppm alum but operating costs would have increased \$240,000/year/reactor, over high alum feed.

Some of the additives tested have shown excellent reductions in effluent activity but, again, their cost would be prohibitive. One of the better additives, sodium silicate, caused increased fuel jacket corrosion and the cost of the half-plant test alone was almost a quarter of a million dollars (\$135,000 direct chemical costs).

Reductions in sodium dichromate concentrations do not produce proportional reductions in effluent ⁵¹Cr activity. This is because the activity is more dependent on the small amount of parent material absorbed in the in-reactor films than on the bulk amount of chemical in the coolant water.

Probably the most important parameter in effluent activity is the seasonal variation of parent isotopes in the Columbia River. Because the parent isotopes originate in various drainages of the Columbia River, the seasonal patterns vary somewhat. Sodium, uranium, sulfur, and phosphorus tend to decrease (by dilution) when the main river flow increases indicating a constant source; copper, manganese, and arsenic increase early in the year when the Spokane River system floods and decrease when the main stream flow increases; and zinc and cobalt concentrations follow the main Columbia River flow curve, indicating origin in Canada. The manganese-56

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activity was particularly bothersome in 1969 when the Grand Coulee dam reservoir was drawn down; effluent ^{56}Mn increased during the period almost a factor of ten higher than normal.

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II. N REACTOR RADIOACTIVE LIQUID EFFLUENTS

There are two major sources of radioactivity which are discharged from the N Reactor:

- (1) Dissolved material in the river water used for cooling which may become radioactive by neutron interaction, and
- (2) Corrosion products from the coolant system piping which may become radioactive by neutron interaction.

River Water

It is apparent that the demineralizer system used for processing river water prior to its use in the reactor coolant system is not completely effective in retaining dissolved materials. This has been shown by an analysis which indicated that radioactive iodine present in the influent to the demineralizer as a result of K Reactor operation was also present in significant quantities in the demineralizer effluent.

Since the river water contains dissolved elements primarily in the form of dissolved salts, both cations and anions are present. The majority of the anions exist in the form of sulfates, silicates, and bicarbonates.

Table IV gives an indication of the elemental concentration in the Columbia River during an eight-month period in 1962. There is a possibility the sulfur and phosphorus concentration in the form of sulfates and phosphates may have increased since 1962 because of increased farming activity and increased use of fertilizers.

Quantities and Sources of Radioactive Material Released to the Environment

The radioactive material produced by neutron interaction is seen in the discharge streams of the following reactor systems:

- 1) Reactor Coolant (primary loop)
- 2) Control Rod Coolant
- 3) Graphite Coolant
- 4) Fuel Storage Basin

At the present time, items 1), 2), and 4) discharge to the crib and subsequently discharge to the river after percolation through the soil. The discharge from item 3) is mixed with the ^{Effluent} ~~outfall from the outfall~~ from the dump condensers.

To determine the radioactive material concentration in the discharge of these systems, a number of selected radionuclides were chosen for analysis in grab samples from these systems in December 1969 and January 1970. The radionuclides selected had half lives greater than one day and included radioisotopes of particular interest when considering the public health and safety, such as ^{90}Sr , ^{137}Cs , ^{65}Zn , ^{131}I , and ^{32}P . These radioisotopes which tend to reconcentrate in the food chain as well as others which could be generated by reactor operation were analyzed.

Table V presents the average radionuclide concentrations from the four reactor systems listed above. Grab samples for these systems were obtained on four different days and were taken during normal routine power operation. The results of these tests were averaged and the concentrations of radioactive material reported is a result of this averaging. The "raw water," "filtered water," and "demineralized water" were sampled on three similar occasions and the results averaged to give the concentrations reported. Table VI summarizes the radionuclides released to the 1301-N crib.

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It should be noted that uranium in a natural form is found in the river water and has been shown to exist in sufficient quantities to produce measurable quantities of fission products from neutron interaction during a single pass through the K Reactors.

Table IV also indicates that the concentration of the elements found in the river water is not constant and some large seasonal variations may be observed.

There is evidence then to indicate that one source of radioactive material being discharged from N Reactor arises from neutron interaction with dissolved material in the river water which has escaped retention in the demineralizer system and has been injected into the coolant stream. River water subject to neutron interaction is also used in the control rod cooling system and graphite cooling system.

While material in the water of the storage basin is not subject to neutron interaction, the water does contain radioactive material. The radioactive material present probably arises from leakage of damaged fuel elements and targets stored in the basin and activated corrosion products which are discharged with the fuel elements during charge-discharge operations.

Coolant System Corrosion Products

A large portion of the coolant system is constructed of ferrous and corrosion resistant metal alloys. However, corrosion is not negligible and very small quantities of corrosion products can be subjected to neutron interaction.

The quantity of corrosion products present at any time for interaction with neutrons produced in the core is a function of coolant chemistry and possibly coolant temperatures which could cause variations in corrosion rate and mass flow.

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The seal well data are from the analysis of a single sample.

Table VII indicates possible sources of the radionuclides whose concentrations have been presented in Tables V and VI.

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TABLE IV
ELEMENTAL CONCENTRATIONS IN COLUMBIA RIVER WATER COLLECTED AT 100B AREA DURING 1962

Date Collected	Filterable Solids(ppm)	Element concentration (ppb)											P	S
		Na	Cu	Mn	As	La	U	Zn	Fe	Co	Sc			
January 5	0.3	1,230	--	1.69	1.34	--	0.27	--	63.1	0.013	0.001	--	4,300	
	0.6	830	2.21	0.45	--	--	--	--	--	--	--	10	4,000	
February 2	0.5	890	27.5	0.45	0.96	0.027	0.50	5.38	8.3	0.011	0.001	9	3,100	
February 9	3.6	940	0.72	--	0.87	0.022	0.13	4.09	27.2	0.032	0.002	1	1,800	
February 16	12.1	970	15.0	3.66	1.05	0.994	0.59	5.18	9.7	0.011	0.002	50	5,600	
February 23	1.3	1,170	1.68	0.73	0.99	0.028	0.81	7.91	20.6	0.019	--	16	4,500	
March 2	1.3	1,210	3.60	0.27	0.97	0.047	0.64	12.5	12.2	0.013	--	14	4,300	
March 9	1.0	1,290	2.59	0.28	0.94	0.052	0.65	8.63	6.6	0.011	0.001	16	4,500	
March 16	1.2	1,340	1.77	0.64	--	--	--	--	--	--	--	12	3,600	
March 23	3.3	1,270	7.09	0.78	--	--	--	--	--	--	--	24	5,500	
March 30	3.2	850	1.44	0.84	1.14	0.378	0.72	6.06	6.7	0.011	0.002	21	5,000	
April 6	1.2	1,440	6.38	0.49	--	--	--	--	--	--	--	--	5,300	
April 13	29.3	1,220	16.8	4.68	1.43	0.190	0.73	14.2	17.4	0.021	0.006	27	5,000	
April 20	3.2	1,320	2.82	86.9	2.49	0.253	0.33	23.2	--	0.032	0.002	20	4,800	
April 27	1.5	1,280	1.40	11.1	4.87	1.762	0.17	37.6	--	0.030	0.001	--	5,500	
May 4	8.0	1,340	1.68	4.58	2.24	0.138	0.33	24.6	--	0.025	0.002	17	4,500	
May 11	5.7	1,210	1.97	2.10	1.80	0.074	0.32	25.5	--	0.025	0.002	24	4,500	
May 18	4.0	1,190	4.35	0.74	2.71	0.921	0.29	--	--	0.021	0.004	20	4,100	
June 12	7.2	800	2.49	0.89	2.15	0.154	0.24	35.4	--	0.021	0.004	11	3,300	
June 15	4.6	800	2.24	0.75	1.96	0.127	0.22	23.8	--	0.028	0.003	7	3,300	
June 22	4.6	800	1.76	0.81	0.87	0.010	0.17	21.9	83.9	0.047	0.002	2	3,100	
June 29	4.7	380	--	0.60	0.90	0.001	0.17	20.3	11.6	0.019	0.002	5	3,300	
July 6	3.2	800	1.85	2.07	1.04	0.044	0.18	26.9	101.0	0.065	0.003	4	3,100	
July 13	3.4	760	1.20	0.62	0.93	0.009	0.16	28.9	20.4	0.028	0.005	2	3,300	
July 20	1.4	750	1.44	0.27	--	--	--	--	--	--	--	2	3,600	
July 27	2.4	790	1.64	0.25	1.09	0.142	0.16	16.8	13.0	0.017	0.002	5	2,900	
August 3	1.2	710	1.62	0.21	3.46	0.017	0.17	13.9	7.8	0.001	0.002	1	2,500	
August 10	1.3	730	1.47	0.57	--	--	--	--	--	--	--	1	2,900	
August 17	0.7	790	2.40	0.68	1.24	0.001	0.18	19.7	62.7	0.039	0.004	1	3,000	

TABLE V

N REACTOR LIQUID STREAM RADIONUCLIDE CONCENTRATIONS IN pCi/liter

Isotope	Half-Life	10CFR20 Limit	Raw Water Inlet	Filtered Water ⁶	Demin. Water	Rod Outlet	Graphite Outlet	Primary Loop	North Basin	South Basin	Crib Inlet ⁷	Seal Well 1	Seal Well 2
H	3 ¹ 12 y	2 3 E6	*<1.0 E3	—	—	—	—	—	2.2 E5	2.6 E5	5.8 E5	*<1.0 E3	*<1.0 E3
P	32 ³ 14d	2 E4	<1.0 E2	*<1.5 1	*<1.5 E1	<4.0 E3	7.4 E4	8.8 E4	2.8 E4	5.0 E4	3.3 E4	8.2 E1	1.6 E2
Sc	46 85d	4 E4	<9.1 E1	3.3 1	<4.0 E1	2.6 E2	1.8 E3	3.4 E3	2.1 E3	1.1 E3	4.6 E3	8.0 E1	8.5 E1
Cr	51 26d	2 E6	<8.5 E2	1.0 3	8.7 E1	5.4 E4	5.9 E3	6.7 E4	9.4 E4	<5.1 E4	5.5 E4	6.8 E2	<1.3 E3
Mn	54 310d	1 E5	1.6 E1	*<2.0 2	*<2.0 E2	5.1 E2	6.3 E3	4.6 E4	3.0 E5	3.4 E5	1.0 E5	*<2.0 E2	*<2.0 E2
Fe	59 46d	5 E4	*<6.0 E1	*<6.0 1	*<6.0 E1	9.6 E2	7.7 E4	1.8 E5	5.3 E4	7.0 E4	8.6 E4	*<6.0 E1	*<6.0 E1
Co	58 72d	9 E4	⁵ ND	ND	ND	1.8 E2	5.0 E2	2.4 E4	2.1 E3	1.9 E3	4.6 E3	ND	ND
Co	60 5.2 y	3 E4	*<1.0 E2	*<1.0 2	*<1.0 E2	1.5 E2	4.2 E4	6.4 E4	8.1 E4	1.0 E5	9.4 E4	*<1.0 E2	*<1.0 E2
Zn	65 250d	1 E5	<5.4 E1	3.4 1	<4.5 E1	2.7 E3	4.7 E3	<4.0 E3	6.0 E3	7.9 E3	6.1 E3	9.8 E1	1.3 E2
Sr	90 25y	3 E2	*<2.0 E1	*<2.0 1	*<2.0 E1	<1.8 E2	<4.7 E1	*<2.0 E1	1.7 E4	1.7 E4	2.8 E3	*<2.0 E1	*<2.0 E1
Nb/Zr	95 35/65d	1E5/6E4	1.4 E1	*<6.0 1	*<6.0 E1	3.8 E2	<1.8 E3	5.9 E4	2.4 E4	2.7 E4	2.6 E4	*<6.0 E1	*<6.0 E1
Mo/Tc*99	67h/2E5Y	4E4/3E6	ND	ND	ND	*<2.0 E3	5.8 E4	3.5 E5	*<2.0 E3	*<2.0 E3	<1.0 E5	ND	ND
Ru	106 1y	1 E4	6.3 E0	*<1.0 1	*<1.0 E1	<1.7 E2	<3.3 E2	<1.2 E3	2.0 E4	6.9 E3	2.1 E3	*<1.0 E1	*<1.0 E1
Sb	122 2.8d	3 E4	<4.2 E1	9.2 1	<5.1 E1	<1.6 E3	<8.3 E3	5.4 E3	6.0 E3	8.9 E3	5.8 E3	1.0 E2	1.5 E2
Sb	124 60d	2 E4	ND	ND	ND	2.0 E2	6.9 E3	6.6 E3	1.6 E3	2.1 E3	2.5 E3	ND	ND
I	131 8d	3 E2	<2.5 E1	<4.0 1	<1.5 E1	1.5 E2	9.0 E3	1.5 E5	2.8 E3	3.4 E4	2.9 E4	*<6.0 E0	<1.2 E1
I	133 ⁴ 22h	1 E3	—	—	—	2.4 E3	—	2.4 E6	4.5 E4	5.5 E5	4.7 E5	—	—
Xe	133 5.3d	None for water	ND	ND	ND	<2.2 E3	2.6 E4	<4.9 E5	6.8 E4	<1.1 E5	<9.8 E4	ND	ND
Cs	134 2.1y	9 E3	ND	ND	ND	2.7 E2	7.8 E2	1.5 E3	1.2 E4	1.3 E4	2.6 E3	ND	ND
Cs	137 33y	2 E4	2.5 E1	*<2.5 1	*<2.5 E1	1.3 E2	7.4 E2	1.2 E3	6.6 E4	3.8 E4	1.1 E4	*<2.5 E1	*<2.5 E1
Ba/La140	13d/40h	2E4/2E4	*<1.2 E2	*<1.2 2	*<1.2 E2	1.0 E3	5.0 E3	3.5 E5	5.4 E4	7.5 E4	6.5 E4	1.7 E2	<2.5 E2
Ce	141 32d	9 E4	ND	ND	ND	<5.6 E2	*<2.5 E2	*<2.5 E2	1.2 E4	1.4 E4	<1.6 E4	ND	ND
Ce	144 290d	1 E4	ND	ND	ND	<1.5 E2	*<1.0 E3	*<1.0 E3	1.6 E4	8.2 E3	<2.9 E3	ND	ND
Eu	154 5.4d	2 E4	ND	ND	ND	<4.6 E3	1.8 E4	<1.7 E4	1.1 E4	1.0 E4	1.7 E4	ND	ND
Eu	156 15d	None	ND	ND	ND	<1.2 E3	*<5.0 E3	<3.1 E4	<1.2 E4	<1.3 E4	<3.9 E3	ND	ND
Ta	182 115d	4 E4	ND	ND	ND	<3.1 E2	3.0 E3	3.1 E3	1.4 E3	<5.0 E3	2.9 E3	ND	ND
Np	239 2.3d	1 E5	<6.9 E2	7.4 2	3.2 E1	2.0 E3	7.3 E3	<4.2 E4	1.6 E3	<3.4 E3	7.7 E4	6.5 E2	1.0 E3

NOTES: ¹ Tritium data obtained from averaging BNW data for crib and basins over December 1969 and January 1970.
² Seal well sample taken January 1970.

³ Floating point notation used for concentrations; e.g., 2.5E3 $\equiv$ 2.5×10^3 .

⁴ All isotopes except H-3 and I-133 averaged from special samples obtained November 19, 1969, November 24, 1969, December 3, 1969, and January 3, 1970 and analysed by U.S. Testing Company.

⁵ I-133 concentrations deduced from I-133/I-131 ratio of previous crib data. Average Ratio = 16.1 found from four effluent samples.

⁶ ND: Not Detected

⁷ Filtered water taken from rod inlet

⁸ Crib inlet taken at "horse Trough"

* Below analytical limit as given; e.g., *<1.0E3 means concentration detected but below 1000 pCi/l.

— Not analyzed for.

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TABLE VI

RADIONUCLIDES GOING TO 1301-N CRIB (For Typical Stream Flows)

Isotope		Half-Life	10CFR20 Limit		Primary Loop		Rods		Basins		Total Into Crib
			pCi/l		pCi/l	Ci/Day	pCi/l	Ci/Day	pCi/l	Ci/Day	Ci/Day
H	3	12y	3	E6					3.0E5	0.33	0.33
P	32	14d	2	E4	8.8E4	0.38	4.0E3	0.024	5.0E4	0.054	0.46
Sc	46	85d	4	E4	3.4E3	0.015	2.6E2	0.001	2.0E3	0.002	0.02
Cr	51	26d	2	E6	6.7E4	0.29	5.4E4	0.32	9.4E4	0.102	0.71
Mn	54	310d	1	E5	4.6E4	0.20	5.1E2	0.003	3.4E5	0.37	0.57
Fe	59	46d	5	E4	1.8E5	0.78	9.6E2	0.006	7.0E4	0.076	0.86
Co	58	72d	9	E4	2.4E4	0.105	1.8E2	0.001	2.0E3	0.002	0.11
Co	60	5.2d	3	E4	6.4E4	0.28	1.5E2	0.0009	1.0E5	0.109	0.40
Zn	65	250d	1	E5	4.0E3	0.017	2.7E3	0.016	8.0E3	0.009	0.04
Sr	90	25y	3	E2	<2.0E1	--	1.8E2	0.001	1.7E4	0.018	0.02
Nb/Zr	95	35/65d	1E5/6E4		5.9E4	0.26	3.8E2	0.002	2.7E4	0.029	0.29
Mo/Tc*	99	61h/62D	4E4/3E6		3.5E5	1.53	2.0E3	0.012	2.0E3	0.002	1.54
Ru	106	1y	1	E4	1.2E3	0.005	1.7E2	0.001	2.0E4	0.022	0.03
Sb	122	2.8d	3	E4	5.4E3	0.024	1.6E3	0.010	9.0E3	0.010	0.05
Sb	124	60d	2	E4	6.6E3	0.029	2.0E2	0.0012	2.0E3	0.002	0.03
I	131	8d	3	E2	1.5E5	0.65	1.5E2	0.009	6.4E4	0.037	0.70
I	133	22h	1	E3	2.4E6	10.46	2.4E3	0.014	5.5E5	0.60	11.07
Xe	133		None		4.9E5	2.14	2.3E3	0.014	1.0E5	0.109	2.26
Cs	134	2.36	9	E3	1.5E3	0.006	2.7E2	0.002	1.3E4	0.014	0.02
Cs	137	33y	2	E4	1.2E3	0.005	1.3E2	0.0008	6.6E4	0.072	0.08
Ba/La	140	13d/40h	2E4/2E4		3.5E5	1.53	1.0E3	0.006	7.0E4	0.076	1.61
Ce	141	32d	9	E4	<2.5E2	--	5.6E2	0.0034	1.4E4	0.015	0.02
Ce	144	290d	1	E4	<1.0E3	0.004	1.5E2	0.0009	1.6E4	0.017	0.02
Eu	154	5.4y	2	E4	1.7E4	0.074	4.6E3	0.028	1.1E4	0.012	0.11
Eu	156	15d	None		3.1E4	0.14	1.2E3	0.007	1.3E4	0.014	0.16
Ta	182	115d	4	E4	3.1E3	0.014	3.1E2	0.002	5.0E3	0.005	0.02
Np	239	2.3d	1	E5	4.2E4	0.18	2.0E5	1.20	3.4E3	0.004	1.38
TOTALS						19.1	1.7		2.1	22.9	

NOTES: All isotope concentrations except H-3 and I-133 found from averaging special samples found during winter 1969-1970.

H-3 concentrations averaged from BNW data from December 1969 and January 1970.

I-133 concentrations found from ratioing other data: $\frac{I-133}{I-131} = 16.1$.

Average Flows: Primary Loop 800 gpm: $\left(\frac{800 \text{ Gal}}{\text{Min}} \right) \left(\frac{3.758 \text{ l}}{\text{Gal}} \right) \left(\frac{1440 \text{ Min}}{\text{Day}} \right) = 4.36E6 \text{ l/Day}$

Rods 1100 gpm: $(1100) (3.758) (1440) = 6.00E6 \text{ l/Day}$

Basins 200 gpm: $(200) (3.785) (1440) = 1.09E6 \text{ l/Day}$

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TABLE VII

<u>Radiosotope</u>	<u>Reaction</u>	<u>Cross Section Barns</u>	<u>Natural Abundance of Target Isotope</u>	<u>Possible Source and Remarks</u>
^{32}P	$^{31}\text{P} \text{ n, } \gamma \text{ } ^{32}\text{P}$	0.19	100%	Thermal neutron interaction with phosphorus from dissolved phosphates in the cooling water; interaction with trace quantities of phosphorus from ferrous alloys making up the coolant system piping; interaction with phosphorus impurities in NH_4OH used for coolant system pH control.
	$^{32}\text{S} \text{ n, p } ^{32}\text{P}$	0.3	95%	Fast neutron reaction with sulfur from dissolved sulfates in the cooling water; interaction with trace quantities of sulfur from ferrous alloys making up the coolant system piping; interaction with sulfur impurities in the NH_4OH used for coolant systems pH control. Cross section listed in average for all energies above the threshold reaction level of 3 Mev.
^{46}Sc	$^{45}\text{Sc} \text{ n, } \gamma \text{ } ^{46}\text{Sc}$	23	100%	Thermal activation of Sc in cooling water.
	$^{46}\text{Ti} \text{ n, p } ^{46}\text{Sc}$		7.93%	Fast neutron reaction with Ti corrosion product of control rod.
^{51}Cr	$^{50}\text{Cr} \text{ n, } \gamma \text{ } ^{51}\text{Cr}$	17	4.31%	Thermal activation of coolant system piping corrosion product
^{54}Mn	$^{54}\text{Fe} \text{ n, p } ^{54}\text{Mn}$	.011	5.82%	Fast neutron reaction with coolant system piping corrosion product.

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TABLE VII (Cont'd)

<u>Radioisotope</u>	<u>Reaction</u>	<u>Cross-Section (Barns)</u>	<u>Natural Abundance of Target Isotope</u>	<u>Possible Source and Remarks</u>
^{59}Fe	$^{58}\text{Fe } n, \gamma \text{ } ^{59}\text{Fe}$	1.2	0.33%	Thermal activation of coolant system piping corrosion product.
	$^{59}\text{Co } n, p \text{ } ^{59}\text{Fe}$	0.00025	100%	Fast neutron interaction with coolant system piping corrosion product. Threshold energy ~ 0.8 MeV.
^{58}Co	$\text{Ni } n, p \text{ } ^{58}\text{Co}$	0.04	67.9%	Fast neutron interaction with coolant system piping corrosion product.
^{60}Co	$^{59}\text{Co } n, \gamma \text{ } ^{60}\text{Co}$	37.0	100%	Thermal activation of coolant system piping corrosion product.
^{65}Zn	$^{64}\text{Zn } n, \gamma \text{ } ^{65}\text{Zn}$	0.46	48.9%	Thermal activation of Zn in cooling water; activation of trace impurity from coolant system piping corrosion product.
^{90}Sr	$^{235}\text{U } n, \text{f.p.}$	~ 580	0.72%	Thermal fission of U in cooling water.
Nb/ ^{95}Zr	$\downarrow$	$\downarrow$	$\downarrow$	$\downarrow$
Mo/ ^{99}Tc	$\downarrow$	$\downarrow$	$\downarrow$	$\downarrow$
^{106}Ru	$\downarrow$	$\downarrow$	$\downarrow$	$\downarrow$
^{122}Sb	$^{121}\text{Sb } n, \gamma \text{ } ^{122}\text{Sb}$	6.8	57.3%	Thermal activation of Sb in cooling water; activation of corrosion product impurity.
^{124}Sb	$^{123}\text{Sb } n, \gamma \text{ } ^{124}\text{Sb}$	3.4	42.8%	Thermal activation of Sb in cooling water; activation of corrosion product impurity.

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TABLE VII (Cont'd)

<u>Radioisotope</u>	<u>Reaction</u>	<u>Cross-Section (Barns)</u>	<u>Natural Abundance of Target Isotope</u>	<u>Possible Source and Remarks</u>
$^{131}_{\text{I}}$	$^{235}_{\text{U}} \text{ n, f.p.}$	~ 580	0.72%	Thermal fission of U in cooling water.
$^{133}_{\text{I}}$				
$^{133}_{\text{Xe}}$				
$^{134}_{\text{Cs}}$				
$^{137}_{\text{Cs}}$				
$^{140}_{\text{Ba/La}}$				
$^{141}_{\text{Ce}}$				
$^{144}_{\text{Ce}}$				
$^{154}_{\text{Eu}}$				
$^{156}_{\text{Eu}}$				
$^{238}_{\text{Np}}$	$^{238}_{\text{U}} \text{ n, } \gamma \rightarrow ^{239}_{\text{U}} \xrightarrow{\beta^-} ^{239}_{\text{Np}}$	2.7	99.3%	Thermal neutron absorption by U in cooling water.

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