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August 17, 1959

TITLE

Significance of Rupture Debris in the Columbia River

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SIGNIFICANCE OF RUPTURE DEBRIS IN THE COLUMBIA RIVER

By

J.D. McCormack

And

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Equipment & Instrumentation
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August 17, 1959

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PRELIMINARY REPORT

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SIGNIFICANCE OF RUPTURE DEBRIS IN THE COLUMBIA RIVER

INTRODUCTION

Failure of fuel elements during reactor operations permits fission isotopes
to enter reactor cooling water and subsequently, the Columbia River. Rupture
debris may contribute appreciably to the overall contamination of the river,
hence it becomes necessary to evaluate the significance of this source of
radioactive material, establish realistic limits, and indicate controls to
be exercised.

The mechanisms of fuel element failures have been given extensive study to in-
crease the integrity of the element and thereby reduce the operational dif-
ficulties associated with ruptures. The radiological impact of rupture debris
entering the Columbia River, in contrast, has received little study or analy-
sis during the same period. Routine monitoring and biophysics research have
shown the monthly average concentration of isotopes in the Columbia at Pasco
to be less than 5% of the MPC for continuous occupational exposure. The spec-
ific contribution of ruptures to the fraction of MPC has not been clearly
identified.

Estimates made for the years 1952-1954[1] placed the contribution from ruptures at
a low value relative to other isotopes appearing in normal effluent. Estim-
ates for the following years showed the quantity of rupture debris increasing
materially as a result of greater frequency of the more severe failures. Dur-
ing this interval the gamma rupture detectors were under development and in
1955 the first full reactor installation was made at 100-H. Since that time
gamma monitors have been installed on all reactors. It was anticipated that
these instruments could be utilized as fission product monitors. After cal-
ibrating as such, they could then conceivably, record actual quantities of
fission products in the stream, and signal a warning when this rupture debris
exceeded a concentration of radiological concern.

Since reference (1) there has been no work reported which evaluated the
significance of rupture debris and presented experience with the rupture mon-
itors in regard to estimating release of fission product to the Columbia
River.

OBJECTIVE

It is the objective of this report to review the recent past reactor operat-
ing experience with regard to ruptures and to discuss rupture significance
with respect to the contamination of the Columbia River. Release of fission
product through serious incidents, not experienced to date, is considered.

SUMMARY

The contribution of ruptures to the fraction of MPC in the Columbia River at Pasco was estimated on the basis of the radioactive strontium content of the Columbia River at Pasco, from 107 Basin samples, and from rupture monitor charts during ruptures using available data on rupture monitor calibration. Data are not highly accurate, although results are reasonably consistent.

It is estimated that ruptures contribute 20% of the present Sr^{89-90} content of the Columbia River at Pasco and about 4% of the gross fission product activity. The average rupture studied released an estimated 30 curies of fission products to the river, measured at Pasco, or 10% of the previously used weighted average. Adequate data are lacking to determine the basin and river clean-up of rupture debris, but because of the apparent solubility of many of the fission products, clean-up by particle deposition is thought to be minor. Additional work to characterize rupture debris is needed to permit clean-up to be estimated.

Estimates were made of plausible but low probability releases of fission product from ruptures with the attendant impact on the Columbia River. It is estimated that a release of 22 grams of uranium/minute with nominal exposure of 500 MWD/ton would raise and maintain the Columbia River to the MPC at 24 hour decay. A very unlikely coincidence of ruptures could raise the river to three times the occupational limit (MPC).

ESTIMATES OF FISSION PRODUCTS RELEASED FROM RUPTURES

There are few data available which permit good estimates to be made of fission product released during ruptures. The available data have been reviewed and four methods of estimating the fission product released from ruptures are discussed with the reservations and limitations indicated.

1. Release Estimate Based Upon Categorically Assigning a Given Weight Loss to Two Principal Rupture Types.

    Experience shows a rather wide variety of ruptures occurring. Few quantitative data are taken upon examination of a ruptured fuel element relating to weight loss of the element. In a very general way, however, ruptures were characterized by being either: (1) severe, usually a side rupture, or fragmented element, or (2) others, such as split or end cap failure.

    Through visual estimates of volume of uranium lost and some few measurements, a release of 150 grams for the severe ruptures and 9 grams loss for all others were assigned these two classes[1]. It was on this basis that rupture debris released to the Columbia River was estimated in reference (1). The limitations in accuracy of this method of estimating rupture debris are obvious, and the estimated release of fission product using these data were subject to a large uncertainty. It was

believed that the use of these values would likely over-estimate the quantity of fission product reaching the Columbia River, but trends could be shown using this interpretation.

In the conversion of weight of uranium (either 150 or 9 g) to fission product curies, the known power level and irradiation time were used. A decay of 24 hours between the average point of release and Pasco was assumed.

Rupture frequency, severity, and consequent curies of fission products at 24 hours decay, estimated by the above method, are shown in Fig. 1. Rupture frequency and characterization were taken from summary reports of rupture experience[2].

2.   Release Estimates Based Upon Rupture Monitor Response

The application of the rupture monitors to estimating release of rupture debris requires that the signal be known as a function of fission product concentration in the cross header sampled by the monitor. Some fifteen ruptures have been used to estimate the calibration factor in terms of curies per minute for one major division on the rupture monitor chart[3].

Results of these tests placed the average rupture monitor sensitivity at 0.016 curies per minute per major division on the strip recording chart. This factor was calculated assuming 100 day irradiated metal, and a cross header flow of 1200 gpm. As indicated in HW-61326 the data show a dispersion which results in low precision for a particular determination of the calibration factor. The average of these individual determinations should be more nearly the true value. It is doubtful, however, that the average calibration factor will give an average release more accurate than a factor of two.

Using the average calibration factor and the integrated area under the strip chart trace of the monitor signal, the average curies released per rupture were calculated for some 12 ruptures. The average release calculated in this way was 16 curies per rupture, which is about 5% of the 1956-1958 average release calculated assuming 150 and 9 g uranium loss ruptures.

3.   Release Estimates Based Upon Analyses of Columbia River Water for Fission Products.

The quantity of Sr^{89-90} present in the Columbia River for the period of April to October, 1958, was determined from Sr^{89-90} analyses of integrated Pasco raw water samples, and the average river flow during the sampling period (usually one week). In Figure 2 the Sr^{89-90} thus determined is plotted against rupture frequency. Although the least squares fit to the data is a straight line with positive slope of 0.5 curies per rupture, a low correlation coefficient (0.26) is obtained for the data because of the dispersion. For the number of points
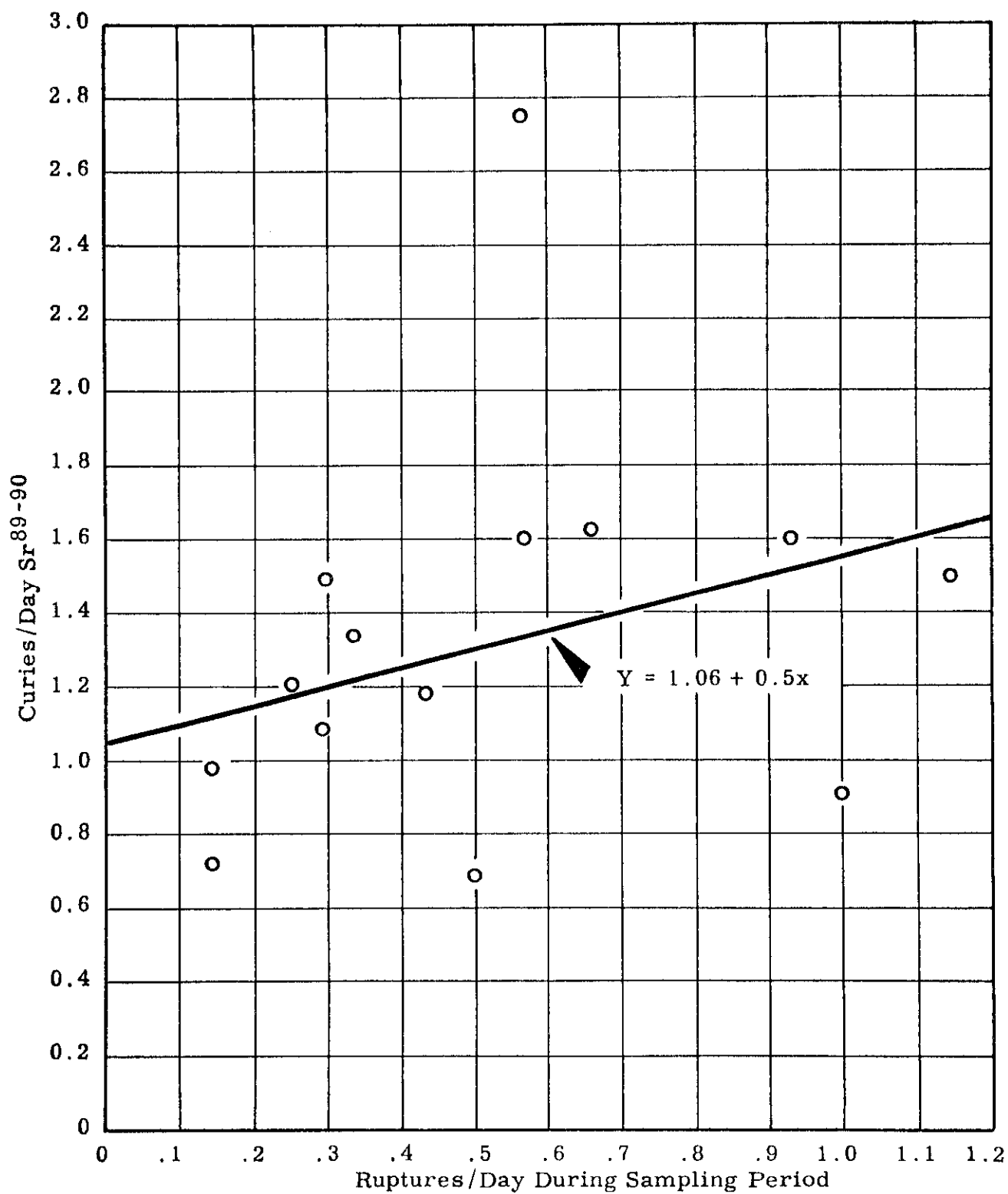
FIGURE 1

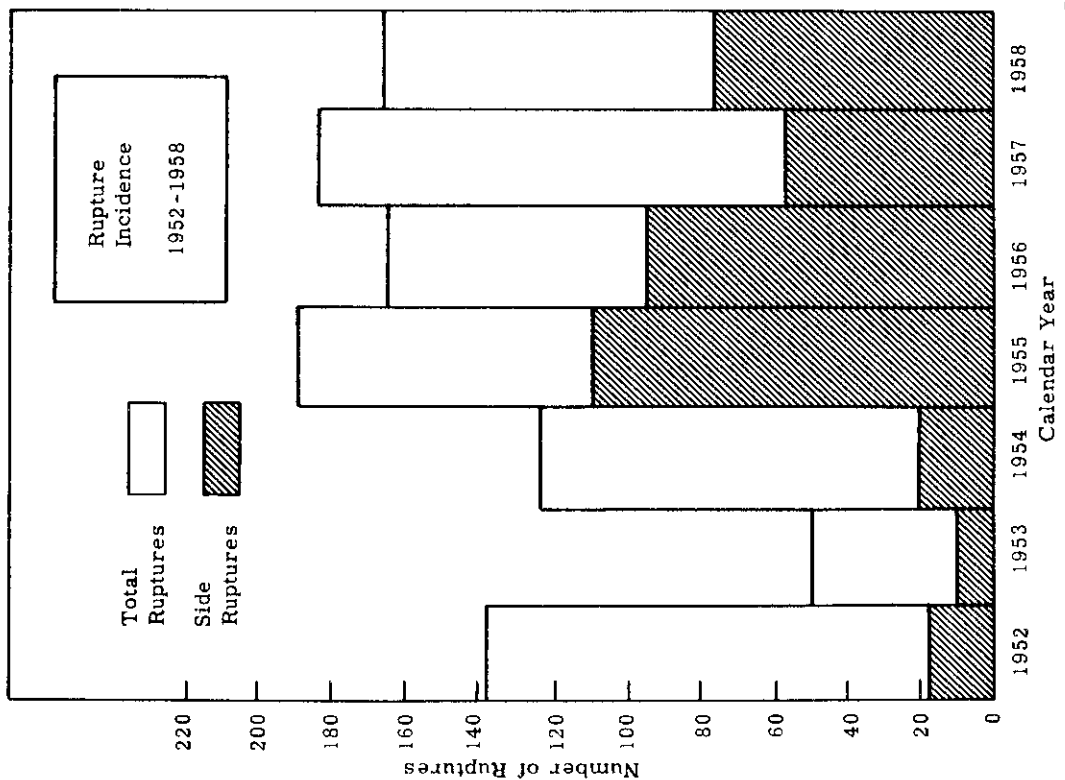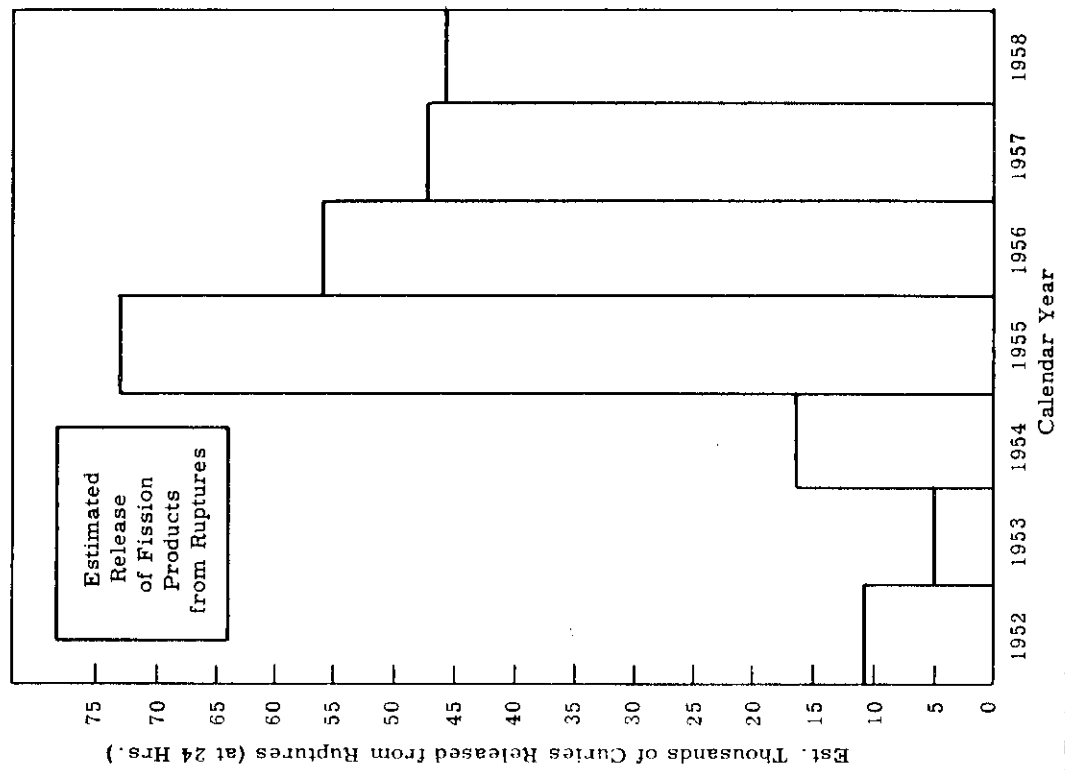FIGURE 2

Sr^{89-90} at Pasco vs. Rupture Frequency
Apr. - Sept. 1958

plotted and the variation shown the correlation coefficient is significant at the 60% level.

Routine grab samples at Pasco during the same period, which were always higher in Sr content than the integrated sample, averaged 2.2 curies per day of Sr^{89} and 0.43 curies per day of Sr^{90}. Because of this consistent bias the results obtained from the integrated weekly sample were corrected upward by a factor of 1.3 to make the results consistent with each other. The curies, then of Sr^{89-90} in the Columbia River would be expressed by

$$\text{Curies } Sr^{89-90}/day = 1.4 + 0.65 \text{ (ruptures per day)}.$$

There is no really satisfactory explanation for the consistent difference between grab sample and weekly integrated samples. It has been suggested that some carry through of shorter-lived Ba^{140} in the Sr analysis may have resulted in somewhat higher values for grab samples, and not so materially with the weekly integrated sample(*). Increasing the release rate per rupture by 1.3 gives the more pessimistic value.

During the period studied the average irradiation time was 66 days which would imply a loss of 21 curies of gross fission products per rupture at 24 hours decay.

4. Release Estimates Based Upon Composited 107 Retention Basin Outlet Analyses for Sr^{89}.

Available data of Sr^{89} content of continuous basin outlet samples obtained during 1956 were treated in a similar fashion. These data are shown in Figure 3. The correlation is very poor. The best-fit line gives 2 curies Sr^{89} released per rupture, or some 50 curies of gross fission product at 24 hours decay (average irradiation time was 130 days).

Table I summarizes the losses estimated in the studies discussed above which involved some 145 ruptures of all types.

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* Present analytical methods for Sr^{89}, Sr^{90} and Y^{90} are currently under critical review to insure accuracy at the low levels experienced.

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Figure 3

Sr^{89} at 107 Basin Outlet
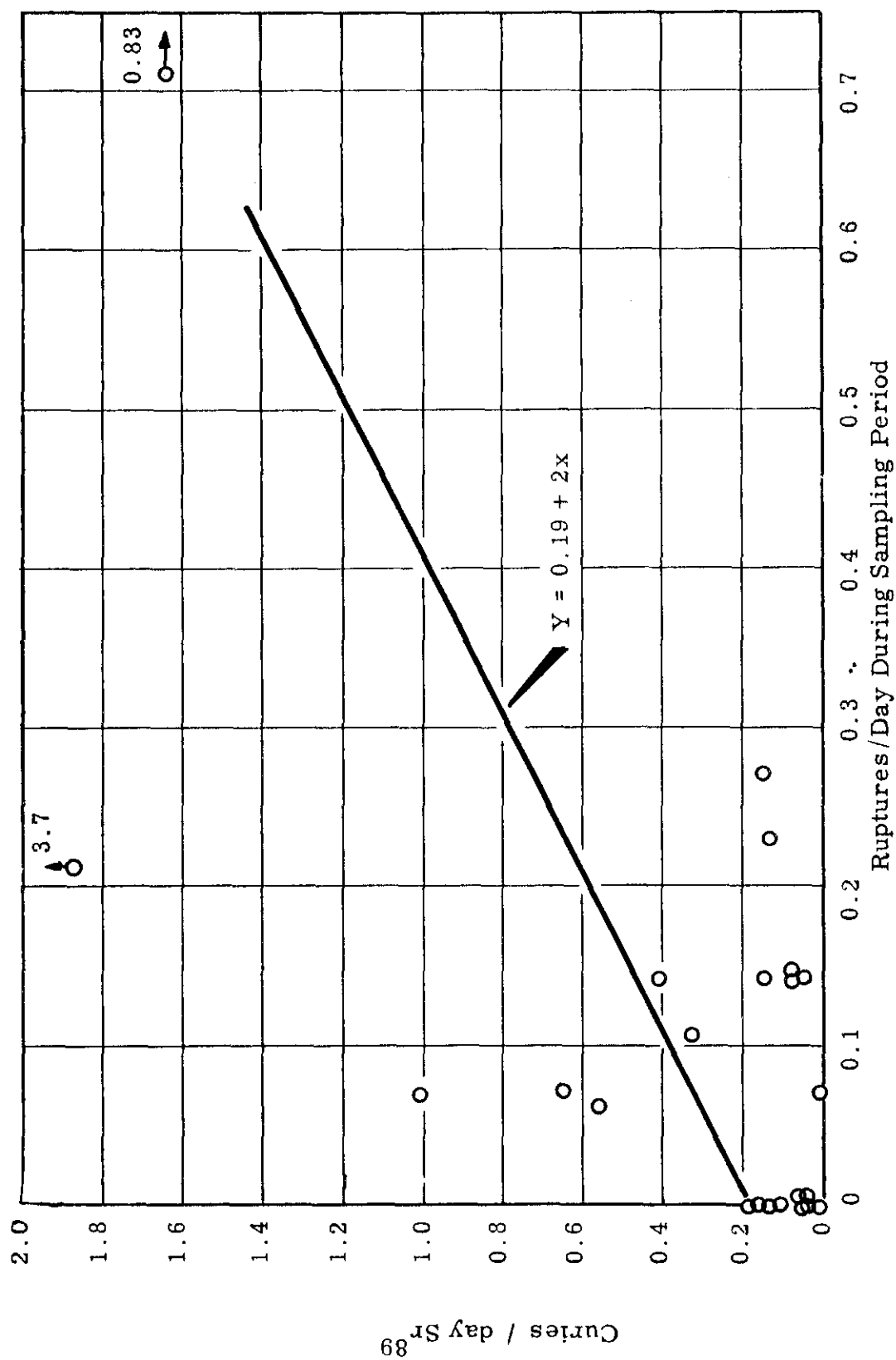Continuous Samples - 1956

$Y = 0.19 + 2x$

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TABLE I

ESTIMATES OF AVERAGE LOSS PER RUPTURE

Method	Rupture Monitor Signal	Pasco Raw Water Analyses	107 Basin Analyses	Average
Curies at 24 hours	16	21	50	29 (~30)
Grams Uranium	5	5	14	8

It is interesting to note that the average appoximates the nine gram loss assumed earlier by Healy for split and cap type failures. It might be speculated that faster detection due to the monitors may have resulted in less severe ruptures. This conclusion would be consistent with the relatively lower overall average release per rupture than the release calculated assuming a 150 gram loss from a severe rupture and nine gram loss from others.

It is again emphasized that average release per rupture as presented in Table I, although calculated from the best data available, is nevertheless subject to a significant tolerance. The magnitude of this uncertainty is not known but could be a factor of perhaps two either way from the nominal value. Simplifying assumptions required in the absence of data on such points such as distortion of fission product spectra due to preferential solubility, prior release of noble gases with significant daughter products, and particulate behavior reduce the accuracy of the estimates.

CLEAN-UP BY BASINS AND RIVER

It has been speculated in the past that the basins and the river could effect an appreciable decontamination of rupture debris by deposition of particulates. The efficacy of this cleaning would depend on the particle size of the rupture debris and solubility of the more hazardous fission products such as strontium and iodine. In 1958 Perkins and Nielsen[4] reported on self-cleaning of the river, but unfortunately neither of these species were included.

Little information on the size of rupture debris particles is available, however, samples from ruptures of unirradiated uranium in loops[5] and autoclaves[6] have been sized. Table II gives the size distributions obtained.

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TABLE II

URANIUM CORROSION PRODUCT PARTICLE SIZE

Loop		Autoclave			
Size Microns	Weight Percent	Size Microns	Weight Percent	Size Microns	Weight Percent
>30	1.5	>1190	1.5	>130	0
25-30	1.9	420-1190	2.5	70-130	22
20-25	8.2	177-420	1.2	50-70	16
15-20	28.5	149-177	6.4	40-50	26
10-15	40.1	74-149	11.2	30-40	15
5-10	16.9	44-74	10.9	20-30	9
1.6-5	2.9	<44	66.3	10-20	7
				<10	5

Because the size of oxide particles produced through the reaction of uranium
with water is relatively independent of the temperature and strongly dependent
upon mechanical attrition[7] it would seem the samples of material obtained
from the loop would most closely approach the actual size of rupture debris.
At a density of eight, 15μ particles settle at a terminal velocity of 0.1
ft/min. At a basin transit time of 100 minutes ample settling should occur
to remove uranium oxide particles of this size, resulting in perhaps 50%
decontamination, if an average size of 15 microns is assumed.

Decontamination by settling in the basins cannot, of course, be effective for
soluble fission products. Some rather qualitative estimates of fission pro-
duct solubilities have been made. These observations, which have been sum-
marized in Table III, indicate Ba, Cs, I, and Sr are soluble.

TABLE III

SOLUBILITY OF FISSION PRODUCTS FROM RUPTURES

Isotope	Soluble	Comments	Reference
I	Yes	I formed by decay of Te is soluble	IDO-16213
Te	No		
Ce^{144}	Probably	Associated with particles but plated on piping.	KAPL-M-SMS-72
Y^{91}	Probably		
Sr^{89}	Yes	No decontamination by filtering.	
Ba^{140}	Yes		
Sr	Yes	Material from J slug rupture leaving basin.	HW-32059
I	Yes		
$Sr^{89-90}+Y^{90}$	Yes	None removed by filtering.	Unpublished data. Rupture Sample KE-1
$Ba^{140}+La^{140}$	Partly	40% removed by filtering.	
Cs^{137}	Partly	70% of total amount in solution (pH 10, 300°C)	HW-59659

The concentration of rupture debris downstream of the reactors was estimated
from the Sr89-90 analyses assuming an undisturbed spectrum of fission iso-
topes. The reasonable consistency between this estimate and estimates from
107 basin analyses, and rupture monitor calibrations tends to substantiate the
solubility of Sr. The assumption of complete solubility for other fission pro-
ducts maximizes the amounts of gross fission products present in the Columbia
River.

Additional data on the size and solubility of rupture debris are needed. It
is possible that tests now being conducted with irradiated uranium in loops
by Coolant Testing Operation can provide some of these data. In view of the
apparent solubility of many of the fission products it seems advisable to
assume no clean-up by the basins and river when estimating amounts of rupture
debris downstream.

PRESENT SIGNIFICANCE OF FISSION PRODUCTS FROM RUPTURES IN THE COLUMBIA RIVER

The average value of 30 curies per rupture may be used with the reservations
indicated to estimate the present contribution of ruptures to the fraction of
MPC of isotopes in the Columbia River. The present rupture frequency approx-
imates one rupture each two days, hence an estimated 15 curies are released
on an average each day. At minimum water flow (assumed to be 36,000 cfs) this
release would result in a concentration of about 1.7×10^{-7} µc per cc. This
assumes complete mixing, no fallout in basins or in the stretch of river be-
tween the reactors and Pasco. At the average river flow (taken to be
130,000 cfs), the concentration would be about 5×10^{-8} µc/cc.

The MPC of a fission product mixture*, of 100 days irradiation and 24 hours
decay is computed to be 4×10^{-4} µc/cc for a 168 hr/week exposure (occupa-
tional) GI tract limiting(10). At the average river flow rate the contri-
bution from ruptures is thus about 0.0001 of the occupational MPC. Fission
products from ruptures thus presently contribute less than one percent of the
annual average fraction of MPC in the river.

In reference (1) Healy recommended a working limit of 0.3 curies per minute at
24 hours decay. Under average river flow this is equal to a concentration of
about 1.4×10^{-6} µc/cc, or some 30 times that presently estimated in the Col-
umbia River due to ruptures. The concentrations quoted are on a year-average
basis; however, if it is assumed that the average rupture persists for four
hours, then the concentration in the river for a 30 curies loss over this four
hour period will be about 6×10^{-7} µc/cc or 12 times the average concentration
estimated to be present.

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* Computed for the most restrictive conditions (soluble or insoluble). Decay
   products of noble gases were assumed to remain with the mixture.

UNUSUAL RUPTURE INCIDENTS

The contribution of ruptures to the river activity has been estimated above on
the basis of present frequency and severity. Here is briefly considered the
magnitude of release which may be postulated under much more serious conditions
of unknown but very low probability of occurrence. As results of the loss of
water to a tube or tubes have been noted elsewhere[8], we shall consider only
the magnitude of a "reasonably possible" rupture. In the event of a malfunction
of the rupture monitor the rupture could be detected by the changing pressure
drop across the tube. To remain undetectable by this method the corrosion
product would have to be removed in such a fashion as to avoid swelling of
the jacket and plugging of the tube. The immediate removal of a corrosion
product by the cooling water could conceivably happen for a freely exposed area.
The amount of debris released per rupture has been estimated at 30 curies or
8 gm. Examination of the rupture monitor charts indicates significant release
may typically occur during a four-hour period. Assuming an exposed area of
10 sq cm a corrosion rate of 200 mg/hr cm^2 is obtained, corresponding to the
rate at a temperature of 380°F[9] for unirradiated uranium. While this seems
high for the U-H_2O interface temperature the apparent high corrosion rate may
reflect the effects of irradiation, erosion, and internal heat generation.
Taking this as a base rate an eight inch element which split in two along the
axis would corrode with an exposed area of 150 sq cm and release 500 mg per min
or about 2 curies per min resulting in 2% of the occupational MPC in the river
at 24 hours decay until a reactor shut-down or complete corrosion of the uran-
ium in five days. Assuming three of these more serious undetected ruptures
occurring simultaneously among all reactors the quantity of debris at Pasco
would raise the river concentration to about 7% of the occupational MPC from
this source for an extended period of time.

Because the maximum amount of fission products which could be released from
any one rupture is the total content of the ruptured element or about 15,000
curies (at 24 hours decay) the resulting downstream conditions (created by the
release) are described by:

$$\text{(River concentration)(Release time + time spread)} = \text{Constant.}$$

At extremely high release rates the time spread dilution of the river is con-
trolling. That is, a quantity of material deposited in the river over a one
minute interval will arrive at Pasco during a one hour interval 24 hours later
if the transit time is 24 hours and time spread one hour. The effect of the
complete loss of fission products from an eight inch element under these sim-
plified conditions is shown in Figure 4. Taking one hour as the minimum likely
time spread*, the rapid loss (of nearly explosive violence from an unpostulated
mechanism) of a full element would result in 280% of the MPC (4×10^{-4} μc/cc)
(GI) for one hour duration at Pasco. This is the maximum concentration thought
possible, short of a release from a disastrous event, although lower concentra-
tions of proportionately longer duration may be of greater radiological con-
sequence because of increased chances for exposure. No attempt has been made
to assay the relative risk associated with the conditions shown in Figure 4.

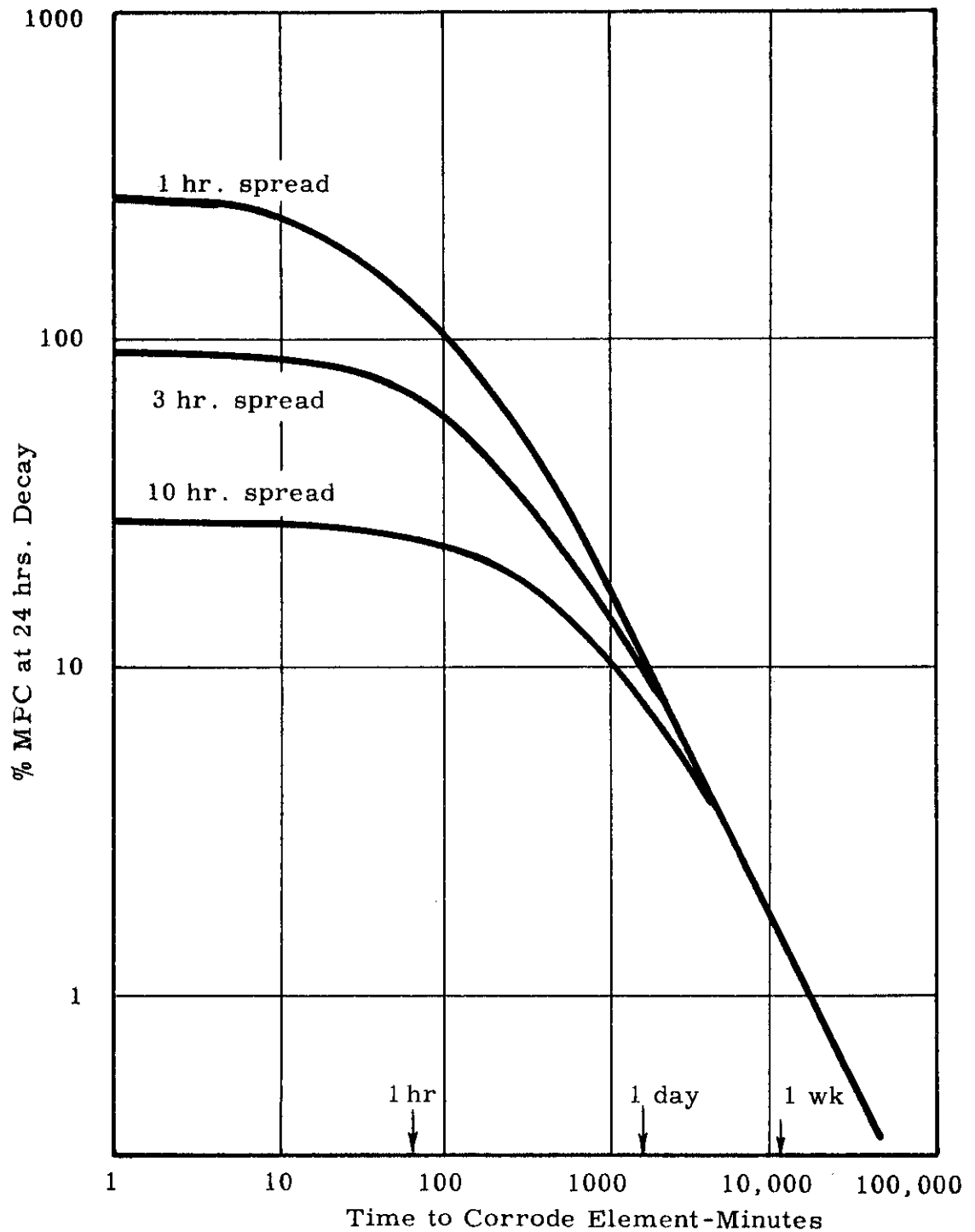* Float tests show typical time spreads of 3 to 5 hours[11].

FIGURE 4

Estimated Effect
Loss of 8" Element

While an extremely severe rupture should be readily detectable the response
time, not the sensitivity, becomes of primary importance. Undesirable emis-
sions could arise from such ruptures releasing significant amounts of material
while checking the monitor operation or obtaining confirming samples. Pro-
tection against such events can be obtained by a time delay (retention basins)
before committing such "off standard" effluent to the river or by decreasing
the time necessary to confirm up-scale monitor signals.

CONCLUSION

While the accuracy of the fission product losses from ruptures as estimated
from the various sources of data can be questioned the three methods used lend
mutual support to each other. This reasonable agreement restores some of the
confidence lost by examination of the correlations obtained, nevertheless, a
significant uncertainty still exists concerning actual release to the river.
It seems likely a sampling program designed specifically to measure rupture
debris combined with integrated rupture monitor chart readings could provide
needed additional data about both the amounts released and the gamma monitor
capabilities. Rather than to speak in terms of the "average" rupture it would
then be possible to characterize each rupture, and determine the probability
of a rupture of given severity occurring. Lacking the results of such a pro-
gram it is suggested that ruptures be assessed at a loss of 8 grams each, with
periodic reviews to determine the effect of any manufacturing or operational
changes. At the present frequency and severity ruptures do not contribute
significantly to the fraction of MPC at Pasco.

In considering downriver conditions created by unusually severe ruptures it is
estimated that a rupture releasing some 200 times the fission product currently
released from the average rupture during average river flow would result in
about 2% of the occupational MPC at Pasco from this source alone. This would
persist until the reactor were shut down, or until the element were completely
disintegrated in some five days. It is not possible to state categorically
that a rupture whose severity were 200 times that of the present average would
always be detected in a short interval after rupture; however, it is unlikely
such a rupture would go undetected. It is not only important that severe
ruptures be detected early, but that corrective action be performed quickly
after detection. This latter requirement is of some concern, since there is
presently a significant lag period between detection, and confirmation, and
reactor shut-down.

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