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PRELIMINARY I-129 MEASUREMENTS IN THE SRP ENVIRONS (U)

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PRELIMINARY I-129 MEASUREMENTS IN THE SRP ENVIRONS

INTRODUCTION

Recently a preliminary program was begun to measure I-129 concentrations in a variety of environmental samples. The objectives of the study were threefold: First, to at least qualitatively estimate the impact of Savannah River Plant (SRP) operations on the I-129 inventory in the surrounding area; second, to determine prominent pathways of I-129 to man and to obtain, where possible, quantitative estimates of their associated dose rates; and third, to provide necessary input data to help in the design and implementation of more comprehensive follow-up studies.

Reasons for the recent increased interest in releases of I-129 to the environment and possible effects on man are evident from the following general comments.

Unlike I-131, I-129's very long half-life of 10^7 years means that it poses a long term potential hazard, particularly in view of the expanding nuclear power industry. As a beta emitter followed by only a weak 40 keV gamma-ray, the primary hazard to man is thru ingestion with the thyroid being the critical organ since it contains about 20 percent of the body's total iodine. Prior to 1945, I-129 concentrations in soil, arising from cosmic interactions and natural fission, were estimated to be about 10^{-12} pCi/gm.[1] On the other hand, the contribution to present background levels from atmospheric weapons testing fallout is calculated to be about 1×10^{-6} pCi/gm in soil, based on the uniform deposition of 8.4 Ci of I-129 (40mCi/kt x 2.1 x 10^5 kt) over land and water in the northern hemisphere.[1] This concentration is in good agreement with measurements made by Batelle Northwest Laboratories (BNWL)[1] where levels around Morris,

Illinois were found to be about 5×10^{-6} pCi/gm in the top 1-inch of soil.

Noting the dramatic increase in I-129 background levels caused by weapons testing, it is logical to investigate the radiation dose-to-man that might result from world-wide nuclear fuel reprocessing. The environs of SRP are a particularly valuable area for study because of the two decades of fuel reprocessing at this site.

SUMMARY

A preliminary program for measuring I-129 concentrations in various types of environmental samples, including air, soil, vegetation, milk and deer thyroids, in an area extending to 100-miles from SRP has been completed. Measurements were based on neutron activation analyses of iodine separated from these samples.

The technique involves separation of iodine by charcoal trapping following oxygen combustion of samples. Quartz ampoules containing iodine from samples and standards were irradiated in K-Area reactor. The irradiated iodine was separated from other activation products and counted on a Ge(Li) detector. Programs were written to use a programable calculator for direct reduction of the spectral data.

Measured I-129 concentrations in various types of samples were compared to dispersion model predictions. The agreement for air and soil samples was within a factor of five out to about 25-miles. Vegetation samples were within a factor of ten. In all cases measured values exceeded predicted values and agreement became worse with increasing distance from the plant. Within about a 25-mile radius of the plant, I-129 levels are expected to be moderately high and measured values should be fairly accurate. The overall good agreement between calculated and measured values in that area tends to support the validity of the data. At greater distances, however, the larger discrepancies are attributed to sample contamination which was difficult to avoid with previous laboratory facilities, especially for vegetation. Efforts are presently underway to remedy this problem.

Averaged measured values for air, vegetation and milk samples, in the Plant perimeter - 25 radius sector, were used to obtain human thyroid dose estimates for infants and adults from a critical pathway model. The doses obtained for infants and adults were 0.30 and 3.95 mrem/year respectively. A specific activity model gave 0.8 and 3.0 mrem/year based on I-129/I-127 atom ratios in infant and adult thyroid equal to the average found in on-plant deer.

This study was intended to be preliminary in nature and to give only a broad and cursory view of I-129 levels in the immediate area. Plans for a more comprehensive and in-depth study of I-129 in the environment are being developed.

SAMPLE ANALYSIS

METHOD

Since the uptake of I-129 in man is governed by the ratio of radioactive to natural iodine in the environments that supply the sources of his bodily iodine, a method which could measure both I-129 and natural I-127 was needed. Neutron activation analysis was found to be best suited to this task since it allows both iodine concentrations to be measured, and provides the high sensitivity needed for environmental samples.

Figure 1 shows a list of the reactions involved. The short half-life of I-128, from reaction 1, requires that samples be counted within about 5-6 hours after irradiation if a natural iodine concentration is to be determined.

Reactions 1 and 2 are the heart of the method since it is the comparison of I-128 and I-130 count rates in samples, with standards, which allows the I-127 and I-129 concentrations in the samples to be calculated. As is usually the case in activation analyses, several interfering reactions (3, 4 and 5) can cause problems due to increased counting backgrounds. Reaction 3 can be minimized by using very small amounts (1 nCi) of I-125, a tracer spike added to samples to determine chemical yield. Reaction 4 is induced by fast neutrons, and can best be avoided by irradiating samples in a flux with a high slow to fast neutron ratio. Since environmental samples usually contain a significant amount of bromine, reaction 5 can only be avoided thru chemical separation of iodine either before and/or after irradiation.

PROCEDURE

A simplified flow chart outlining the processing of samples is shown in Figure 2. Samples are initially spiked with a known amount of I-125 which is counted in the final step to determine the iodine chemical yield thru the procedure. Air samples are actually activated charcoal canisters which collect the iodine from a known amount of air. Soil and vegetation samples are dried initially while deer thyroids are analyzed on a wet-weight basis. Iodine from water and milk samples is collected on anion-exchange resin.

Samples or ion-exchange resin are then loaded into a quartz furnace tube and combusted in an O_2 stream at 1000°C. A charcoal trap following the furnace tube collects the iodine driven off the sample. This charcoal is inturn burned and the iodine trapped a second time on about 1.5 gm of charcoal in a small diameter quartz tube. The tube is sealed at one end, evacuated, and then heated to drive the iodine off the charcoal to the sealed end of the tube which is immersed in liquid nitrogen. A second seal is then made to form an ampoule about an inch long containing the iodine.

Four sample ampoules and two containing I-127 and I-129 standards are welded into a single aluminum irradiation capsule. Following irradiation for two hours at a thermal neutron flux of about 10^{14} in the K-Area rabbit, ampoules are removed from the capsule and brought back to the laboratory for iodine separation from other activation products by multiple extractions into CCl_4. Samples and standards are mounted on source cards as a AgI precipitate and then counted for I-128 and I-130 on a Ge(Li) detector in a low-level counting facility. After a several day decay period, a low-energy photon detector is used to count the I-125 spike for a yield determination.

DATA REDUCTION

To facilitate handling and reducing the data, programs were written to utilize a programable calculator interfaced directly to the multichannel analyzer used to accumulate the iodine sample spectra. An example of typical output from the data printout and peak integrator program is given in Figure 3. The operator first inputs the sample name along with the times out of the reactor and counted so that decay corrections can be made. Next the channel numbers in

which the analytical peaks for I-128 and I-130 are to be found are assigned. The calculator then prints these channels, followed by a request for an energy slope factor. After the value is input, the calculator echos it and prints the results of the peak integrations. The peak at 443 keV belongs to I-128 as does the smaller one at 527 keV. The 536 keV peak belongs to I-130.

After the above program is run for all four samples and the two standards, and the I-125 values are obtained in a similar way, the data is input into a second program as shown in Figure 4. The calculator asks for the sample name and values for counts/min and sigma for I-125, I-128 and I-130, echoing them as they are input. The program then prints out the yield and corrected count rates for I-128 and I-130. After this is done for the samples and standards, the calculator again asks for a sample name and sample size, and then calculates the μg of I-127 and I-129 along with the pCi of I-129 and the I-129/I-127 atom ratio in the sample.

RESULTS

F and H STACK AIRS

Measurements of I-129 concentrations released to the atmosphere by the separations areas were obtained from analyses of one-week long primary and back-up charcoal canisters routinely taken by Health Physics personnel. The total I-129 concentrations are summarized in Table 1. The data for the various months are really only a reflection of a single weeks' activity in that month, and can only be expected to average-out over a number of months. Hence extrapolated values at the bottom of the table should be viewed as order-of-magnitude type numbers.

Estimated releases for 1975 were not available at the time of this writing. The lower extrapoluted total I-129 concentrations measured for 1975 compared with the estimated 1974 concentration reflect the reduced operations in one of the separations areas during the year.

AIR and SOIL

To interpret the other data obtained in this study, dispersion models which predict desired air concentrations based on meteorlogical conditions and known source terms were used. One such model developed by R. E. Cooper,[2] here at SRL, utilizes known average weather conditions for the plant and surrounding area to calculate χ/Q, or concentration per unit source values at various distances from the stacks. Multipling these values by the estimated stack release rates, gives air concentrations at specified distances which can be compared with measured values. Similarly, once air concentrations are known, one has only to multiply by an appropriate iodine deposition velocity and total exposure time to determine soil concentrations.

A comparison between calculated and measured air and soil I-129 concentrations at various distances from the stacks is shown in Table 2. The comparison is quite good with the single exception of air at 100 miles where the measured value is high by about a factor of 40. It is suspected that this is due to a contamination problem associated with handling low-level samples in the laboratory where I-129 air concentrations are moderately high. However, agreement between air samples can be expected to be somewhat poorer since one is

comparing short term measurements with long term average calculations.

VEGETATION

Extending such model calculations along the ecological chain to vegetation can be done, but results are less reliable since each step in the chain introduces additional parameters, which in general can only be estimated. A better approach is to use a measured transference number which relates vegetation to air concentrations directly. Such a number has been determined by W. L. Marter[3] from studies made following the 1961 SRP I-131 release.

The comparison between calculated and measured I-129 values in vegetation samples is given in Table 3. Calculated values are based on the measured transference number, determined by Marter, and the model predicted air concentrations at the specified distances. While agreement is not as good as it was with air and soil, neither is the overall quality of the data. Calculated values are more assumptive by nature and the potential for sample contamination, which has been attributed as the cause of the large discrepancy at 100 miles, is more likely in view of vegetations' large affinity for airborne iodine.

MILK

Insufficient data were available to attempt similar comparisions for milk, since neither samples of actual cattle forage nor specific information on dietary make-up was sought in this preliminary study. However, milk samples were classified as originating from single cows or dairies, and it was found that in general I-129 concentrations in milk from single cows were higher. This of course is reasonable since single cows are mostly on pasture whereas dairy cattle usually receive supplimental feed.

Dairy milk samples, all from within a 25-mile radius of the plant, had an average I-129 concentration of 6×10^{-2} pCi/l, while the highest value, that from a single cow near the plant perimeter, was 9 pCi/l. This shows there is a wide variation in milk values, but all are well below the ERDA RCG value of 60 pCi/l for water.

DOSE ESTIMATES

Using the average measured values for I-129 concentrations in various types of samples in the plant perimeter - 25 mile radius sector, human thyroid dose estimates were obtained from a critical pathway model for both infants and adults. (For a detailed discussion of this model and methods of dose calculations see Reference 4.)

Results are summarized in Table 4. Values are based on assumed thyroid masses of 2 gm for infants and 20 gm for adults along with the expected average intake rates via the pathways shown at the bottom of the Table. The major pathway for adults appears to be thru leafy vegetables, whereas normally milk is considered the dominant pathway. This is due to the higher than expected measured vegetation values noted above. In any event, the totals of 0.30 and 3.95 mrem/year are small compared to the revised limits cited in Technical Standard DPSTS-RH-W-0.1 (30 mrem/year).

As a footnote, a worst-case upper limit to the dose rates in Table 4 can be obtained from a specific activity model[4] if one assumes that the average I-129/I-127 atom ratio, from measurements of on-plant deer thyroids, is not likely to be exceeded in man residing off-plant. Sixteen on-plant deer thyroid samples were analyzed. The average I-129/I-127 ratio was 4.2×10^{-5}. An outline of the calculations and the results of the model estimates are shown in

REFERENCES

1. J. K. Soldat Et Al., "The Radioecology of Iodine-129." An Interim Report, USAEC Report BNWL-1783, Battelle Memorial Institute, Pacific Northwest Laboratories, September, 1973.

2. R. E. Cooper, "Computer Programs at SRL to Evaluate Environmental Effects of SRP Operations and Postulated Accidental Releases." DPST-75-384 (1975).

3. W. L. Marter, "Radioiodine Release Incident at the Savannah River Plant." Health Phy., 9:1105-1109 (1963).

4. J. M. Palms Et Al., "The Environmental Impact ^{129}I Released by a Nuclear Fuel Reprocessing Plant." Nucl. Safety, 16-5: 593-602 (1975).

TABLE 1

MEASURED I-129 STACK RELEASES

Month/Year	I-129 (mCi)
01/75	2.9
02/75	2.1
03/75	3.3
04/75	5.6
05/75	*
06/75	2.7
07/75	*
08/75	5.6
1975 Total To Date	22.2
1975 Extrapolated	44
1974 Estimated	170

* Not measured.

TABLE 2

COMPARISON OF MEASURED AND CALCULATED I-129 CONCENTRATIONS IN AIR AND SOIL

Distance	Soil(pCi/gm)*	Soil(pCi/gm)	Air(pCi/m^3)*	Air(pCi/m^3)
Plant Perimeter	1.1×10^{-3}	2.1×10^{-2}	4.7×10^{-5}	6.5×10^{-5}
25 Mile Radius	3.1×10^{-3}	5.0×10^{-3}	1.3×10^{-5}	5.6×10^{-5}
100 Mile Radius	3.1×10^{-4}	1.0×10^{-3}	1.3×10^{-6}	5.7×10^{-6}

* Calculated values.

TABLE 3

COMPARISON OF MEASURED AND CALCULATED I-129 CONCENTRATIONS IN VEGETATION

Distance	Calculated(pCi/g Dry)	Measured(pCi/g Dry)
Plant Perimeter	2×10^{-3}	9×10^{-3}
25 Miles Radius	5×10^{-4}	4×10^{-3}
100 Miles Radius	5×10^{-5}	5×10^{-3}

TABLE 4

ANNUAL THYROID DOSES FROM VARIOUS PATHWAYS

Pathway	Dose(mrem/year) Infant	Adult
Inhalation	1.4×10^{-3}	2.5×10^{-3}
Ingestion		
Via Milk*	0.30	0.15
Via Leafy Vegetables**	0	3.80
TOTAL	0.30	3.95

* Based on 1 liter/day local dairy milk @ 5.5×10^{-2} pCi/l.

**Based on 73 kg/year leafy vegetables @ 7 pCi/kg.

LIST OF REACTIONS INVOLVED IN NEUTRON ACTIVATION ANALYSIS OF IODINE SAMPLES

PERTINENT REACTION FOR IODINE ACTIVIATION ANALYSIS

1) $^{127}I(n,\gamma)\ ^{128}I \rightarrow\ ^{128}Xe$ $\quad t_{\frac{1}{2}} = 25$ min.

2) $^{129}I(n,\gamma)\ ^{130}I \rightarrow\ ^{130}Xe$ $\quad t_{\frac{1}{2}} = 12.4$ hr.

INTERFERING REACTIONS

3) $^{125}I(n,\gamma)\ ^{126}I \rightarrow\ ^{126}Xe$ $\quad t_{\frac{1}{2}} = 13$ d.

4) $^{127}I(n,2n)\ ^{126}I \rightarrow\ ^{126}Xe$

5) $^{81}Br(n,\gamma)\ ^{82}Br \rightarrow\ ^{82}Kr$ $\quad t_{\frac{1}{2}} = 36$ hr.

FIGURE 1

FLOW SCHEME FOR I-127, 129 SAMPLE ANALYSIS

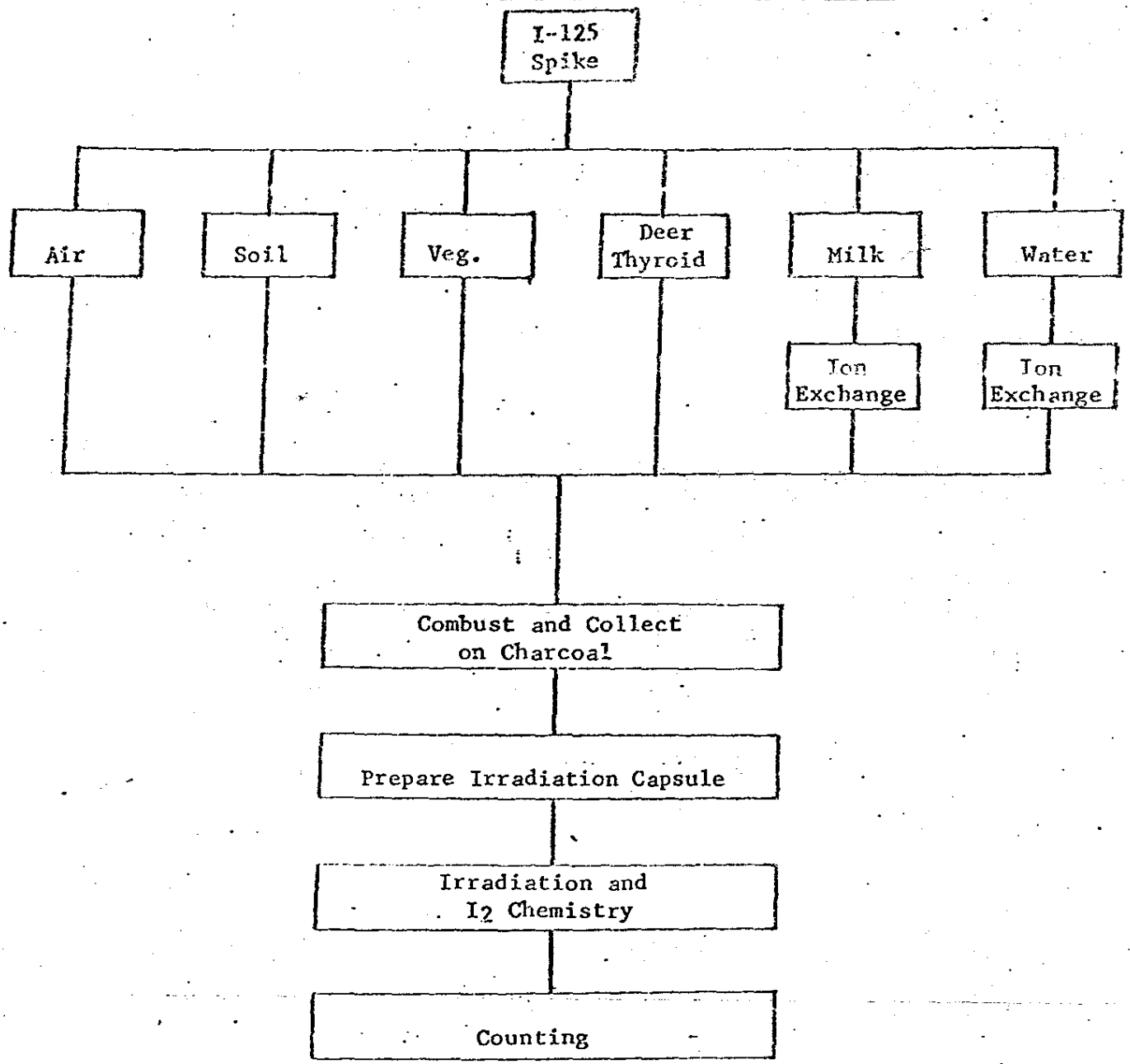

FIGURE 2

I-129, SERIES #348, SAMPLE #4
TIME OUT 07/08/75/08/28
COUNTED 07/08/75/14/07

TYPICAL OUTPUT FROM DATA PRINTOUT AND PEAK INTEGRATOR PROGRAM

600										
430	146	168	182	188	167	158	173	168	152	150
440	170	239	568	891	692	251	164	135	130	160
450	151	147	153	126	135	131	168			
522	103	121	100	123	154	157	152	132	114	94
532	111	128	118	135	193	190	130	118	104	107
542	96	102	103	105	100					

KEV/CH 1.0000
PEAK INTEGRATOR

COUNTING TIME	600.0000
ENERGY	443.0437
COUNTS/MIN.	193.0833
SIGMA	6.3397
ENERGY	527.4303
COUNTS/MIN.	17.5000
SIGMA	3.8588
ENERGY	536.4898
COUNTS/MIN.	24.4000
SIGMA	3.9038

FIGURE 3

SAMPLE NAME

355-2

TYPICAL OUTPUT FROM IODINE DATA REDUCTION PROGRAM

```
ENTER C/M I-125
      3.966000000+02
ENTER SIGMA
      1.460000000+01
ENTER C/M I-128
      4.048028800+07
ENTER SIGMA
      1.767774000+06
ENTER C/M I-130
      1.000700000+04
ENTER SIGMA
      3.470000000+02
YIELD=      3.495813134-01
+OR-       1.353548851-02

I-128=      1.157964870+08
+OR-       6.758231069+06

I-130=      2.862567196+04
+OR        1.487868144+03
```

SAMPLE NAME

355-2

```
ENTER SAMPLE SIZE
      2.125000000+00
UG I-127    =       1.253631260+01
+OR-       8.304983388-01

UG I-129    =       4.583760353-03
+OR-       2.664936499-04

PCI I-129   =       7.639600589-01
+OR-       4.441560831-02

RATIO I-129/I-127=       3.656386450-04
+OR-       3.222772965-05
```

FIGURE 4

SPECIFIC ACTIVITY MODEL - WORST CASE UPPER LIMIT

I-129/I-127 ON-PLANT DEER THYROIDS = 4.2×10^{-5}

ADULT THYROID $\approx 7 \times 10^{3}$ μg IODINE

$4.2 \times 10^{-5} \times 7 \times 10^{3} = 0.29$ μg I-129

THYROID DOSE RATE = 3 MREM/YR

INFANT THYROID $\approx 1.8 \times 10^{2}$ μg IODINE

$4.2 \times 10^{-5} \times 1.8 \times 10^{2} = 7.6 \times 10^{-3}$ μg I-129

THYROID DOSE RATE = 0.8 MREM/YR

FIGURE 5