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GEORGIA INSTITUTE OF TECHNOLOGY
Engineering Experiment Station

PROJECT INITIATION

Date: October 16, 1973

Project Title: "Iodine - 129 Study"

Project No.: A-1570

Project Director: Dr. David M. Walker

Sponsor: Emory University

Effective: September 15, 1973 Estimated to run until: January 15, 1974

Type Agreement: Letter Agreement Amount: $ 16,291

NOTE: two phase project

REPORTS: Progress Report due about January 15, 1974

CONTACT PERSON: Dr. J. M. Palms, Chairman
Room 204, Physics Building
Physics Department
Emory University
Atlanta, Georgia 30322

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GEORGIA INSTITUTE OF TECHNOLOGY
Engineering Experiment Station

PROJECT TERMINATION

Date ~~August 14, 1974~~

PROJECT TITLE: "Iodine - 129 Study "

PROJECT NO: A-1570

PROJECT DIRECTOR: Dr. David M. Walker

SPONSOR: Emory University

TERMINATION EFFECTIVE: ~~7/31/74~~

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IODINE-129 STUDY Project A-1570

R. C. McFarland and D. M. Walker

Progress Report

February 19, 1974

Nuclear Applications Group
Engineering Experiment Station
GEORGIA INSTITUTE OF TECHNOLOGY

Background

As part of a study of I-129 about the site of the Barnwell Nuclear Plant, methods for obtaining valid iodine samples and for measuring the required low concentrations of I-129 are being developed. In particular, the following tasks comprise this phase of the I-129 study:

1. Development of practical measurement methods for I-129 and I-127 in biological media such as thyroid tissue.

2. Development of practical methods for I-129 measurements in HV-70 filter papers and charcoal cartridges.

3. Preliminary evaluation of acceptable flow rate for air sampling system. (More detailed studies to be conducted later).

4. Planning, in conjunction with the Emory University team, the study of I-129 in the environment about the BNFP plant.

Recent literature and information from other groups involved in I-129 studies are also being reviewed.

Progress Report-October 16, 1973 to February 15, 1974

Since this program was initiated in October, attention has been directed to the development of practical I-129 measurement methods. Existing methods offer sensitivity levels of about 4×10^{-6} pCi, but require extensive chemistry and coincidence counting techniques which raise the cost to $400-600 per sample. The desired number of samples anticipated for the BNFP study make these cost levels prohibitive; alternate methods with sensitivity levels of about 10^{-3} pCi and a cost of $200-300 per sample are being developed. These levels appear to be adequate, except for isolated cases, for detecting the I-129 levels which have been reported by groups at BNWL[1], N. Y. Radiological Sciences Laboratory[2], and others.

Procedures now being investigated for I-129 and I-127 in tissue and vegetation are given in Appendix A. Procedures for analyses of filter paper and charcoal cartridges are given in Appendix B. A brief summary of the development effort is given below.

1. Measurements in Biological Media

Practical I-129/127 measurement methods for biological sample media are being investigated based on two procedures currently in general use: first, sodium hydroxide fussion and, second, oxygen combustion.

Our experimental investigations have concentrated on the sodium hydroxide fussion method which seems to be a practical method applicable to a variety of samples. The oxygen combustion method, on the other hand, appears to be the best method for determining I-129 in soil samples, and will also be used for comparison with the sodium hydroxide method for other sample media. An oxygen combustion system is under construction at Georgia Tech.

The procodures given in Appendix A were used to determine I-129 content of thyroid tissue, leaves, and

soybean flour. Due to the 5 MW conversion at the GTRR, the neutron activation was conducted at Argonne National Laboratory CP-5 Reactor, a heavy water reactor similar to the GTRR. For thyroid tissue the sensitivity of this method is of the order of 2×10^{-4} pCi I-129.

Several problems were encountered with the iodine measurements in leaves and flour. The chemical yield was very low, resulting in poor sensitivity. This reduction in chemical yield may be due to the possibility that the sample material did not go into solution in the sodium hydroxide, but combusted with a resulting loss of iodine. Alternate procedures involving acid distillation are being investigated to increase the chemical yield. Procedures for analyses of I-129 in milk and water involve the absorption of inorganic iodine on ion exchange resin with similar extraction and activation procedures as for tissue and vegetation.

2. Measurement of Airborne Iodine

The purpose of this portion of the study is to provide a rapid evaluation of the present BNFP air sampler design consisting of an HV-70 paper filter and a charcoal cartridge. A more detailed study of airborne I-129 sampling is planned for the next phase of this study. A test trap loop for airborne iodine will be constructed and used to determine the most desirable media for collection and retention of airborne iodine.

a. A double inlet air pump system is being used in a preliminary test to determine the effects of air flow rate and sampling time on the iodine collection efficiency of HV-70 filter paper and activated charcoal. Each sampling train of the system consists of an HV-70 particulate filter, two MSA charcoal canisters

in tandem, and a flow meter.

For the flow rate studies, both trains are run simultaneously at different flow rates. Flow rate samples obtained to date are 15 liters per minute, 40 liters per minute, and 60 liters per minute, with a wider range of flow rates planned.

To determine the effect of sampling time on collection, simultaneous samples have also been taken where the components of one sampling train are changed at shorter intervals than those of the other sampling train. Another effect of sampling time, iodine break through, is detected and measured by the quantity of iodine on the second charcoal cartridge in the train as compared to the first. These samples are being stored for analyses when the GTRR resumes operation. A decision on desired operating conditions for the present sampler can be made based on these results.

b. Procedures for the measurement of airborne iodine sampling materials have been investigated, and are presented in Appendix B.

c. Analysis of the particulate phase will utilize an ammonium hydroxide leach of the HV-70 filter paper. Analysis of the vapor phase will be broken into two fractions: the inorganic fraction can be removed from the charcoal by leaching and the total iodine can be determined by sodium hydroxide fussion.

Due to strong activation interferences from sodium, potassium, and manganese in the charcoal, the Szilard Chalmers reaction

technique used in stable iodine determination has not proven practical for I-129 measurements. Alternatively, the separation of inorganic fractions is being developed based on selective leaching procedures.

I-129 Activities by Other Groups

Several other groups in the United States are involved in I-129 studies. The major active programs are at BNWL (an AEC funded study of the NSF site), the EPA lab at Las Vegas, and the present study. Other groups who were active earlier in 1973 but have discontinued their I-129 programs are the EPA lab in Cincinnati (Bernd Kahn's group), the New York State Radiological Sciences Laboratory, and the EPA laboratory in Montgomery, Alabama. The Savannah River Lab recently contacted our group relative to I-129 studies and analyses.

Brief reviews of interesting papers not included in our earlier report[4] are given below:

J. M. Matuszek, J. C. Daly, S. Goodyear, C. J. Paperiello, and J. J. Gabay, "Environmental Levels of I-129", IAEA/SM-130/39, (1973).

Measured I-129 concentrations in milk, water, fish, algee, and thyroids from deer, cows, and small animals from the vicinity of the NSF plant are reported and related to human thyroid dose estimates. Onsite deer thyroid had an average of 492 pCi/g for 15 samples. Measured levels of I-129 in liquid samples ranged from less than the sensitivity limit ($\sim$ .01 pCi/l) to highs of 2.3 p Ci/l for milk, and 8.3 pCi/l for drainage water. Direct counting of I-129 with LEPS type detectors, liquid scintillation counting, and a limited amount of activation analysis were used as measurement methods for I-129 (and I-125).

Calculated thyroid doses in a child due to I-129 are calculated based on 2.3 pCi/L as 500-800 mrem/yr. Thyroid doses from fish consumption of fish with 2 pCi/g of I-129 are 3500-5600 mrem/yr to the child thyroid. These estimates differ drastically from the estimates of the BNWL group.

F. P. Brauer and J. H. Kaye, "Detection Systems for the Low Level Radiochemical Analysis of Iodine-131, Iodine-129, and Natural Iodine in Environmental Samples, BNWL-SA-4726," November 1973.

Alternate methods of γ-γ and B-γ coincidence counting for I-130 in the presence of interference gammas from I-126 and I-128 are discussed. The methods are based on the fact that I-130 decays with several coincident gammas of high energy. Detection limits of 0.1 to 10 dpm for I-130 are estimated for the various systems, with the B-γ systems giving the best sensitivity.

F.P. Brauer, J. K. Soldat, H. Tenny and R. S. Strebin, Jr., "Natural Iodine and Iodine-129 in Mammalian Thyroids and Environmental Samples Taken from Locations in the United States", IAEA-SM-180/34, (1973).

Data is given for I-129 and I-129/I-127 ratios for animal and human thyroid tissue, vegetation, soil, rain water, milk, and air filters and cartridges. The I-129/I-127 ratio measurements on thyroids show an increase from $<10^{-9}$ for pre-1945 samples, to 15-44000 x 10^{-9} for 1962-1964 samples. Average ratios were: Washington State $\sim$ 1700, South Carolina $\sim$ 800, other areas $\sim$ 20. All samples from near the Hanford plant show increased I-129 except river water samples. Calculated thyroid doses of less than 1 Mrem/year for adult and infant cases in the Hanford area is given based on average dietary habits.

Measurement methods are described which give an estimated I-129 sensitivity of 4 x 10^{-6} pCi. The air sample data is questionable because of the filter pore size (7 microns) and the charcoal face velocity (150-300 cm/sec).

F. P. Brauer, H. G. Rieck, Jr. and R. L. Hooper, "Particulate and Gaseous Atmospheric Iodine Concentrations", BNWL-Sa-4723, August 1973.

Measured atmospheric concentrations of I-127, I-129, and I-131 are reported for a series of samples taken with a very high volume sampling system. The sampling face velocity ranged from 12-16 ft./sec., a sampling rate which is difficult to reconcile with published data showing significant loss of collection efficiency at greater than about 1 ft/sec. I-127 concentractions in the nanogram per cubic meter range are given (these valves are in accord with earlier measurements by our group and others). The filter paper pore size of 7 microns caused most of the iodine to be collected on the charcoal, since the reported mean iodine partical size is less than one micron. I-129 concentrations of less than 1 $pCi/10^6$ m^3 to 43 $pCi/10^6$ m^3 near the Hanford plant are reported.

APPENDIX A

PROCEDURES FOR THE ANALYSES OF IODINE-129 AND IODINE-127 IN TISSUE AND VEGETATION

When the iodine-129 content of a sample is known to exceed 5-10 pCi, then determination by liquid scintillation counting is recommended. For smaller amounts of iodine-129, determination by activation analysis is recommended. When the stable iodine content of a sample exceeds one mg then it may be determined by titrimetry; otherwise it can be determined simultaneously with the iodine-129 by activation analysis.

Sample Preparation

1. Cut the sample (5-10 grams) into small pieces and place in a 250 ml nickel crucible. If the sample is to be analyzed by neutron activation, add about 1000 dpm of carrier-free ^{131}I for determination of the chemical yield.
2. Add ten ml of water, 30 grams of NaOH pellets and carefully, ten ml of ethyl alcohol.
3. Heat gently on a hot plate, stirring as needed to dissolve the mass. Evaporate off the ethyl alcohol.
4. Place the crucible in a muffle furnace at 250°C. Raise the temperature to 600°C in increments of 100 degrees at 15-30 minute intervals.
5. Remove the crucible from the muffle furnace and cool. Dissolve the melt with water and transfer to 800 ml beaker. Wash the crucible with water.

Iodine Extraction

1. Heat the solution from Step 5 of the Sample Preparation procedure to just below boiling on a hot plate. Add additional water if necessary. (Volume should be about 400 ml). Slowly pass chlorine gas through the solution for about 30 seconds.

2. Cool the solution. Acidify using concentrated nitric acid (pH < 1) and transfer to a 1000 ml separatory funnel.

3. Add 50 ml of carbon tetrachloride and 10 ml of 1 M hydroxylamine hydrochloride. Shake for two minutes to extract the iodine into the organic phase. Draw off the organic layer into a 250 ml separatory funnel.

4. Add 25 ml of carbon tetrachloride and 5 ml of 1 M hydroxylamine hydrochloride to the first separatory funnel and shake for two minutes. Combine the organic phases. Discard the aqueous phase.

5. Add 25 ml water and ten drops of 1 M sodium sulfite to the separatory funnel containing the carbon tetrachloride. Shake for 1-2 minutes. Discard the organic phase.

6. Add 20 ml of toluene, one ml of 1 M nitric acid, and ten drops of 1 M sodium nitrite to the separatory funnel. Shake for one-two minutes. Discard the aqueous.

7. Add ten ml of water and ten drops of 1 M sodium sulfite to the separatory funnel and shake for one-two minutes. Transfer the aqueous phase to a 60 ml separatory funnel. Discard the organic phase.

8. Add exactly ten ml of toluene, one ml of 1 M nitric acid and ten drops of sodium nitrite to the separatory funnel and shake for two minutes. Discard the aqueous.

9. Add ten ml of 0.01 M nitric acid to the separatory funnel and shake for 30 seconds. Discard the aqueous.

10. Repeat step 9.

11. If the sample is known to contain more than 5-10 pCi of ^{129}I, proceed with the "Determination of ^{129}I by Liquid Scintillation Counting;" otherwise proceed with the "Determination of ^{129}I by Neutron Activation Analysis". If there is a distinct pink color to the toluene layer use the procedure for the "Volumetric Determination of Stable Iodine;" otherwise determine the stable iodine (iodine-127) simultaneously with the iodine-129 by neutron activation.

Determination of ^{129}I and ^{127}I by Neutron Activation Analysis

1. Take a 5-10 ml aliquot of the toluene layer from step 10 of the "Iodine Extraction" procedure and place this aliquot in a 60 ml separatory funnel.

2. Add 4 ml of 1 M amonium sulfite to the separatory funnel. Shake for two minutes and transfer the aqueous phase to a 10 ml volumetric flask.

3. Repeat step 2, combining the aqueous portions in the volumetric flask and dilute to volume with 1 M ammonium sulfite. Transfer five ml of this solution to a polyethylene irradiation vial and heat seal.

4. Prepare standards of ^{127}I and ^{129}I as follows: Pipet exactly five ml of a stock solution containing a known amount of ^{127}I (10 ug/ml) in 1 M ammonium sulfite into a polyethylene vial and heat seal. Similarly, pipet

exactly five ml of a stock solution containing a known amount of ^{129}I (0.01 ug/ml) in 1 M ammonium sulfite into another polyethylene vial and heat seal.

5. Irradiate the vials containing the samples and the standards in a constant neutron flux of 1-2 x 10^{13} $n/cm^2/sec.$ for eight hours. Record the exact time of removal from the flux.

6. Cut open a vial carefully and transfer the solution (pipet exactly 4 ml of the standard solutions) to a 60 ml separatory funnel containing 10 ml of water, 10 mg of iodine carrier as (potassium iodide) and 20 ml of toluene.

7. Acidify with 6 ml of 1 M nitric acid. Add 2 ml of 1 M sodium nitrite to completely oxidize the iodine to free iodine. Shake for about two minutes. Discard the aqueous.

8. Wash the organic phase three times, shaking for 30 seconds with 10-15 ml 0.01 nitric acid each time. Discard washings.

9. Add 15 ml of 1 M ammonium sulfite. Shake for one minute.

10. Add 15 ml of 1 M ammonium sulfite to the separatory funnel containing the toluene and shake for one minute. Combine the aqueous portion with the aqueous from step 9 and acidify with sulfuric acid.

11. Add an excess of palladium chloride solution to precipitate palladium iodide.

12. Wash and mount the palladium iodide precipitate for counting of the iodine-128, iodine 130, and iodine 131.

Note 2: The solution must be cooled before the excess bromine is destroyed with formic acid. If the solution is too warm, the formic acid will also reduce some of the iodate giving low results. It is also important that the solution be colorless after the bromine is destroyed with formic acid.

Note 3: The 0.02 N thiosulfate solution is prepared as follows: Dissolve 2.5 grams of $Na_2S_2O_3 \cdot 5H_2O$, 0.2 grams Na_2CO_3, and three drops of chloroform in water and make up to one liter. Standardize by titrating against a known amount of iodine.

Determination of ^{129}I by Liquid Scintillation Counting

1. Pipet exactly four ml of toluene from step 10 of the "Iodine Extraction" procedure into a liquid scintillation counting vial. Add 1 ml of 2-methyl-1-butene and 10 ml of scintillation solution. (Note 1)
2. The vial is then capped and placed under a fluorescent light for about two hours to decolorize the iodine.
3. Count for ^{129}I in a liquid scintillation counter.

Note 1: The scintillation solution is prepared by dissolving seven grams of 2,5-diphenyloxazole (PPO) and 0.1 grams of 1,4-bis-2 (5-phenyloxozolyl) - benzene (POPOP) in one liter of toluene.

exactly five ml of a stock solution containing a known amount of ^{129}I (0.01 ug/ml) in 1 M ammonium sulfite into another polyethylene vial and heat seal.

5. Irradiate the vials containing the samples and the standards in a constant neutron flux of 1-2 x 10^{13} $n/cm^2/sec.$ for eight hours. Record the exact time of removal from the flux.

6. Cut open a vial carefully and transfer the solution (pipet exactly 4 ml of the standard solutions) to a 60 ml separatory funnel containing 10 ml of water, 10 mg of iodine carrier as (potassium iodide) and 20 ml of toluene.

7. Acidify with 6 ml of 1 M nitric acid. Add 2 ml of 1 M sodium nitrite to completely oxidize the iodine to free iodine. Shake for about two minutes. Discard the aqueous.

8. Wash the organic phase three times, shaking for 30 seconds with 10-15 ml 0.01 nitric acid each time. Discard washings.

9. Add 15 ml of 1 M ammonium sulfite. Shake for one minute.

10. Add 15 ml of 1 M ammonium sulfite to the separatory funnel containing the toluene and shake for one minute. Combine the aqueous portion with the aqueous from step 9 and acidify with sulfuric acid.

11. Add an excess of palladium chloride solution to precipitate palladium iodide.

12. Wash and mount the palladium iodide precipitate for counting of the iodine-128, iodine 130, and iodine 131.

Volumetric Determination of Stable Iodine

1. Pipet a four ml aliquot of the toluene layer from step 10 of the "Iodine Extraction" procedure, into a 60 ml separatory funnel. Add about 20 ml of water and five ml of 1 M sodium sulfite dropwise to completely reduce the iodine to iodide. Shake for about two minutes.
2. Collect the aqueous portion in a 125 ml Erlenmeyer flask.
3. Add another 20 ml of water to the organic phase, and ten drops of sodium sulfite. Shake for two minutes. Combine the aqueous phase with the aqueous from step 2. Discard the organic phase.
4. Add about five ml of bromine-sodium acetate solution (note 1) and heat to boiling to insure conplete oxidation of the iodide to the iodate.
5. Cool in an ice bath. Add formic acid (88-90 per cent) dropwise until the yellow color of the bromine disappears (Note 2).
6. Add about three ml of ten per cent potassium iodide solution, ten ml of 1 M sulfuric acid, and about five drops of one per cent starch solution.
7. Titrate the liberated iodine with a standardized 0.02 N sodium thiosulfate solution (Note 3). Color change is from blue to colorless at the endpoint.

Note 1: The bromine-sodium acetate reagent is prepared by dissolving 10.0 grams of sodium acetate trihydrate and two ml of Br_2 in 100 ml of glacial acetic acid.

APPENDIX B

PROCEDURES FOR THE ANALYSES OF AIRBORNE IODINE-129 AND IODINE-127

I. Filter Paper

1. Add about 1000 dpm of carrier free ^{131}I and leach the filter paper for 15 minutes in concentrated ammonium hydroxide (approximately 20 ml).
2. Filter the solution and add sodium hydroxide to keep it basic.
3. Heat the solution to just below boiling on a hot plate. Add additional water if necessary. (Volume should be about 40 ml). Slowly pass chlorine gas through the solution for about 30 seconds.
4. Cool the solution. Acidify using concentrated nitric acid (pH < 1) and transfer to a 125 ml separatory funnel.
5. Add 25 ml of carbon tetrachloride and 5 ml of 1 M hydroxylamine hydrochloride. Shake for two minutes to extract the iodine into the organic phase. Draw off the organic layer into a 125 ml separatory funnel.
6. Add 25 ml of carbon tetrachloride and 5 ml of 1 M hydroxylamine hydrochloride to the first separatory funnel and shake for two minutes. Combine the organic phases. Discard the aqueous phase.
7. Add 25 ml water and ten drops of 1 M sodium sulfite to the separatory funnel containing the carbon tetrachloride. Shake for 1-2 minutes. Discard the organic phase.
8. Add 20 ml of toluene, one ml of 1 M nitric acid, and ten drops of 1 M sodium nitrite to the separatory funnel. Shake for one-two minutes. Discard the aqueous.
9. Add ten ml of water and ten drops of 1 M sodium sulfite to the separatory funnel and shake for one-two minutes. Transfer the aqueous phase to a 60 ml separatory funnel. Discard the organic phase.
10. Add exactly ten ml of toluene, one ml of 1 M nitric acid and ten drops of sodium nitrite to the separatory funnel and shake for two minutes. Discard the aqueous.
11. Add ten ml of 0.01 M nitric acid to the separatory funnel and shake for 30 seconds. Discard the aqueous.
12. Repeat step 11.

Determination of ^{129}I and ^{127}I by Neutron Activation Analysis

1. Take a 5-10 ml aliquot of the toluene layer from step 12 of the above procedure and place this aliquot in a 60 ml separatory funnel.
2. Add 4 ml of 1 M ammonium sulfite to the separatory funnel. Shake for two minutes and transfer the aqueous phase to a 10 ml volumetric flask.
3. Repeat step 2, conbining the aqueous portions in the volumetric flask and dilute to volume with 1 M ammonium sulfite. Transfer five ml of this solution to a polyethylene irradiation vial and heat seal.
4. Prepare standards of ^{127}I and ^{129}I as follows: Pipet exactly five ml of a stock solution containing a known amount of ^{127}I (10 ug/ml) in 1 M ammonium sulfite into a polyethylene vial and heat seal. Similarly, pipet exactly five ml of a stock solution containing a known amount of ^{129}I (0.01 ug/ml) in 1 M ammonium sulfite into another polyethylene vial and heat seal.
5. Irradiate the vials containing the samples and the standards in a constant neutron flux of 1-2 x 10^{15} $n/cm^2/sec.$ for eight hours. Record the exact time of removal from the flux.
6. Cut open a vial carefully and transfer the solution (pipet exactly 4 ml of the standard solutions) to a 60 ml separatory funnel containing 10 ml of water, 10 mg of iodine carrier as (potassium iodide) and 20 ml of toluene.
7. Acidify with 6 ml of 1 M nitric acid. Add 2 ml of 1 M sodium nitrite to completely oxidize the iodine to free iodine. Shake for about two minutes. Discard the aqueous.
8. Wash the organic phase three times, shaking for 30 seconds with 10-15 ml 0.01 nitric acid each time. Discard washings.
9. Add 15 ml of 1 M ammonium sulfite. Shake for one minute.
10. Add 15 ml of 1 M ammonium sulfite to the separatory funnel containing the toluene and shake for one minute. Combine the aqueous portion with the aqueous from step 9 and acidify with sulfuric acid.
11. Add an excess of palladium chloride solution to precipitate palladium iodide.
12. Wash and mount the palladium iodide precipitate for counting of the iodine 130 and iodine 131.

II. Charcoal - Inorganic Fraction

1. Add about 1000 dpm of carrier free ^{131}I and leach the charcoal for 15 minutes in concentrated ammonium hydroxide (approximately 50 ml).
2. Filter the solution and add sodium hydroxide to keep it basic.
3. Heat the solution to just below boiling on a hot plate. Add additional water if necessary. (Volume should be about 80 ml). Slowly pass chlorine gas through the solution for about 30 seconds.
4. Cool the solution. Acidify using concentrated nitric acid (pH < 1) and transfer to a 250 ml separatory funnel.
5. Add 50 ml of carbon tetrachloride and 10 ml of 1 M hydroxylamine hydrochloride. Shake for two minutes to extract the iodine into the organic phase. Draw off the organic layer into a 250 ml separatory funnel.
6. Add 25 ml of carbon tetrachloride and 5 ml of 1 M hydroxylamine hydrochloride to the first separatory funnel and shake for two minutes. Combine the organic phases. Discard the aqueous phase.
7. Add 25 ml water and ten drops of 1 M sodium sulfute to the separatory funnel containing the carbon tetrachloride. Shake for 1-2 minutes. Discard the organic phase.
8. Add 20 ml of toluene, one ml of 1 M nitric acid, and ten drops of 1 M sodium nitrite to the separatory funnel. Shake for one-two minutes. Discard the aqueous.
9. Add ten ml of water and ten drops of 1 M sodium sulfite to the separatory funnel and shake for one-two minutes. Transfer the aqueous phase to a 60 ml separatory funnel. Discard the organic phase.
10. Add exactly ten ml of toluene, one ml of 1 M nitric acid and ten drops of sodium nitrite to the separatory funnel and shake for two minutes. Discard the aqueous.
11. Add ten ml of 0.01 M nitric acid to the separatory funnel and shake for 30 seconds. Discard the aqueous.
12. Repeat step 11.

Determination of ^{129}I and ^{127}I by Neutron Activation Analysis

1. Take a 5-10 ml aliquot of the toluene layer from step 12 of the above procedure and place this aliquot in a 60 ml separatory funnel.
2. Add 4 ml of 1 M ammonium sulfite to the separatory funnel. Shake for two minutes and transfer the aqueous phase to a 10 ml volumetric flask.
3. Repeat step 2, combining the aqueous portions in the volumetric flask and dilute to volume with 1 M ammonium sulfite. Transfer five ml of this solution to a polyethylene irradiation vial and heat seal.
4. Prepare standards of ^{127}I and ^{129}I as follows: Pipet exactly five ml of a stock solution containing a known amount of ^{127}I (10 ug/ml) in 1 M ammonium sulfite into a polyethylene vial and heat seal. Similarly, pipet exactly five ml of a stock solution containing a known amount of ^{129}I (0.01 ug/ml) in 1 M ammonium sulfite into another polyethylene vial and heat seal.
5. Irradiate the vials containing the samples and the standards in a constant neutron flux of 1-2 x 10^{15} $n/cm^2/sec.$ for eight hours. Record the exact time of removal from the flux.
6. Cut open a vial carefully and transfer the solution (pipet exactly 4 ml of the standard solutions) to a 60 ml separatory funnel containing 10 ml of water, 10 mg of iodine carrier as (potassium iodide) and 20 ml of toluene.
7. Acidify with 6 ml of 1 M nitric acid. Add 2 ml of 1 M sodium nitrite to completely oxidize the iodine to free iodine. Shake for about two minutes. Discard the aqueous.
8. Wash the organic phase three times, shaking for 30 seconds with 10-15 ml 0.01 nitric acid each time. Discard washings.
9. Add 15 ml of 1 M ammonium sulfite. Shake for one minute.
10. Add 15 ml of 1 M ammonium sulfite to the separatory funnel containing the toluene and shake for one minute. Combine the aqueous portion with the aqueous from step 9 and acidify with sulfuric acid.
11. Add an excess of palladium chloride solution to precipitate palladium iodide.
12. Wash and mount the palladium iodide precipitate for counting of the iodine-128, iodine 130, and iodine 131.

III. Charcoal-Organic Fraction

1. Place the charcoal in a 250 ml nickel crucible. Add 1000 dpm of carrier-free ^{131}I for determination of the chemical yield.
2. Add ten ml of water, 30 grams of NaOH pellets and carefully, ten ml of ethyl alcohol.
3. Heat gently on a hot plate, stirring as needed, to the mass. Evaporate off the ethyl alcohol.
4. Place the crucible in a muffle furnace at 250°C. Raise the temperature to 600°C in increments of 100 degrees at 15-30 minute intervals.
5. Remove the curcible from the muffle furnace and cool. Dissolve the melt with water and transfer to 800 ml beaker. Wash the crucible with water.
6. Heat the solution to just below boiling on a hot plate. Add additional water if necessary. (Volume should be about 400 ml). Slowly pass chlorine gas through the solution for about 30 seconds.
7. Cool the solution. Acidify using concentrated nitric acid (pH < 1) and transfer to a 1000 ml separatory funnel.
8. Add 50 ml of carbon tetrachloride and 10 ml of 1 M hysroxylamine hydrochloride. Ahake for two minutes to extract the iodine into the organic phase. Draw off the organic layer into a 250 ml separatory funnel.
9. Add 25 ml of carbon tetrachloride and 5 ml of 1 M hydroxylamine hydrochloride to the first separatory funnel and shake for two minutes. Combine the organic phases. Discard the aqueous phase.
10. Add 25 ml water and ten drops of 1 M sodium sulfite to the spearatory funnel containing the carbon tetrachloride. Shake for 1-2 minutes. Discard the organic phase.
11. Add 20 ml of toluene, one ml of 1 M nitric acid, and ten drops of 1 M sodium nitrite to the separatory funnel. Shake for one-two minutes. Discard the aqueous.
12. Add ten ml of water and ten drops of 1 M sodium sulfite to the separatory funnel and shake for one-two minutes. Transfer the aqueous phase to a 60 ml separatory funnel. Discard the organic phase.
13. Add esactly ten ml of toluene, one ml of 1 M nitric acid and ten drops of sodium nitrite to the separatory funnel and shake for two minutes. Discard the aqueous.
14. Add ten ml of 0.01 M nitric acid to the separatory funnel and shake for 30 seconds. Discard the aqueous.
15. Repeat step 14.

Determination of ^{129}I and ^{127}I by Neutron Activation Analysis

1. Take a 5-10 ml aliquot of the toluene layer from step 10 of the "Iodine Extraction" procedure and place this aliquot in a 60 ml separatory funnel.

2. Add 4 ml of 1 M ammonium sulfite to the separatory funnel. Shake for two minutes and transfer the aqueous phase to a 10 ml volumetric flask.

3. Repeat step 2, combining the aqueous portions in the volumetric flask and dilute to volume with 1 M ammonium sulfite. Transfer five ml of this solution to a polyethylene irradiation vial and heat seal.

4. Prepare standards of ^{127}I and ^{129}I as follows: Pipet exactly five ml of a stock solution containing a known amount of ^{127}I (10 ug/ml) in 1 M ammonium sulfite into a polyethylene vial and heat seal. Similarly, pipet exactly five ml of a stock solution containing a known amount of ^{129}I (0.01 ug/ml) in 1 M ammonium sulfite into another polyethylene vial and heat seal.

5. Irradiate the vials containing the samples and the standards in a constant neutron flux of 1-2 x 10^{15} $n/cm^2/sec$. for eight hours. Record the exact time of removal from the flux.

6. Cut open a vial carefully and transfer the solution (pipet exactly 4 ml of the standard solutions) to a 60 ml separatory funnel containing 10 ml of water, 10 mg of iodine carrier as (potassium iodide) and 20 ml of toluene.

7. Acidify with 6 ml of 1 M nitric acid. Add 2 ml of 1 M sodium nitrite to completely oxidize the iodine to free iodine. Shake for about two minutes. Discard the aqueous.

8. Wash the organic phase three times, shaking for 30 seconds with 10-15 ml 0.01 nitric acid each time. Discard washings.

9. Add 15 ml of 1 M ammonium sulfite. Shake for one minute.

10. Add 15 ml of 1 M ammonium sulfite to the separatory funnel containing the toluene and shake for one minute. Combine the aqueous portion with the aqueous from step 9 and acidify with sulfuric acid.

11. Add an excess of palladium chloride solution to precipitate palladium iodide.

12. Wash and mount the palladium iodide precipitate for counting of the iodine-128, iodine 130, and iodine 131.