BENJAMIN E. WALL. Monitoring ^{35}S During the Incineration of Liquid Low Level Radioactive Wastes. (Under the direction of DR. PHILIP E. HAMRICK AND DR. STEVEN L. SIMON)

ABSTRACT

Effluents and residual ash were monitored in order to determine the fate of ^{35}S labelled liquid waste during high temperature incineration. Effluents were monitored using a modified EPA approved Method 5 isokinetic sampling probe with 3% H_2O_2 as the trapping solution. The radioactivity content in the gaseous effluents and residual ash was counted using a liquid scintillation counter calibrated for ^{35}S. Eleven trial burns of liquid waste with activities ranging from 199 to 5659 μCi[1] were conducted. An aqueous solution of ^{35}S labelled Methionine was the source of activity in eight of the trials and an aqueous solution of ^{35}S labeled sulphate was the source in the remaining three trials. Percent of the total activity incinerated contained in the effluents and ash was determined.

---

[1] Conventional Units are used in certain areas of the text in order to be consistent with licences and engineering specifications

TABLE OF CONTENTS

LIST OF FIGURES

LIST OF TABLES

ACKNOWLEDGMENTS

I would like to thank, Dr. Philip E. Hamrick for his guidance and support throughout the research project, Dr. Steven L. Simon for his assistance in the statistical analysis and editorial skills, and Todd Ivey for his help with equipment and incinerator operation throughout the project.

The study was supported by the National Institute of Environmental Health Sciences. I would like to thank many unnamed people at the Institute for their cooperation and help.

INTRODUCTION

Purpose

The National Institute of Environmental Health Sciences (NIEHS) located at Research Triangle Park, North Carolina, is the principle Federal biomedical research laboratory investigating the effects of chemical and biological agents in the environment As is common with similar facilities, the Institute uses radioactive material primarily as a tracer in various research projects. The use of radioactive material is accompanied by the production of radioactive waste.

The increased costs and restrictions of shallow land burial disposal persuaded many facilities to begin incinerating low level radioactive waste. By implementing incineration, the radioactive material in the waste is concentrated in the residual ash, released in the effluents or precipitated on the refractory (inside stack) surface, thereby significantly reducing the volume of waste. Recent developments in incineration design demonstrate a waste volume reduction of up to 90% (Cook 1984). Many studies have been conducted which advocate incineration of low level wastes as an economical and safe alternative to shallow land burial disposal (Machis 1952, Glauberman 1964, Wollen 1971, Parker 1981, Cook 1984).

In 1969 NIEHS began incinerating its low level radioactive waste, the majority of which consists of ^{14}C and ^{3}H and ^{35}S. As mentioned above, the principle of radioactive waste incineration is based on the assumption that the radionuclide is either released in the effluents during incineration, retained in the residual ash or precipitatedalong the refractory. In order to investigate this assumption, the Institute conducted two studies. In 1983, Michael Parker investigated the fate of ^{3}H and ^{14}C during incineration by monitoring the gaseous effluents and particulates. The study was carried further in 1984 by Steven Knapp. Similar to Parkers study, Knapp monitored the effluents released during the incineration of ^{3}H and ^{14}C labeled waste. Unlike Parker, Knapp also investigated the ash to confirm the effluent data. Neither study investigated ^{35}S

The original objective of this project was to develop a method for the determination of the fate of sulphur-35 during the incineration of radioactive waste. This rather broad topic was narrowed as the project progressed, resulting in the identification of more specific objectives. From this objective the following four "a-priori" questions were formulated and investigated during the course of the project:

1) What is the proportion of the total activity incinerated recovered from the ash and from the effluents?

2)Does the relationship between the proportion in the effluents and in the ash depend on the chemical form of the incinerated waste?

3)Is there a relationship between recovery efficiency and total activity incinerated? Is there a relationship between recovery efficiency and total volume of activity incinerated?

4) Is it a valid assumption that the activity can be accounted for in either the ash or the gas?

Review of Literature

Institutional Radioactive Waste Incineration in Chronological Order

Johns Hopkins University published the results of a study in 1952 which addressed the use of incineration as a method for disposal of its institutional waste (Machis 1952). Waste, with known activities of ^{32}P ranging from 100 to 2000 μCi, were incinerated at several different incinerator locations. Ash, effluents and incinerator refractory were sampled to determine the radioactivity contents. A large fraction of the activity was recovered from the ash, (20 to 100%), with as much as 32% precipitating on the stack wall. Their results indicate the effluents rarely registered activity above background.

In 1964 a study of several waste incinerators, with load capacities of 20 to 2000 pounds per hour, was contracted by the Atomic Energy Commission (AEC). Areas investigated included total volume of reduction of radioactive waste, cost effectiveness of incineration, and the retention of radioactivity in the ash (Glauberman 1964). The volume reduction reported, 80 to 100%, was considerable. Retention of the activity by the ash ranged from 95 to 100%. The specific nuclides studied and method of detection were not reported.

A study was conducted at the University of Minnesota, Minneapolis, from January 1965 through December 1969 to investigate the use of incineration for the disposal of low level radioactive waste generated by the university. Animal carcasses and combustible solid lab waste were incinerated. Based on average stack gas flow rate and maximum permissible concentration (MPC) data listed in table 2, Appendix B, 10 CFR 20, the maximum allowable activity that could be incinerated was calculated for various radionuclides (Wollan 1971). No report of effluent or ash radioactivity was made.

Bush and Hundal reported ash retention results for twenty seven radionuclides incinerated by the University of Birmingham, Birmingham, England (Bush 1973). The results ranged from 0.02 percent retention for ^{14}C and ^{125}I to 100% for ^{22}Na and ^{137}Cs. The results for ^{35}S ranged from 39.6 to 77.2% depending on the chemical form. The 77.2% retention corresponded to an aqueous solution of sodium sulphate. No mention was made about radioactivity counting techniques or effluent sampling of ^{35}S.

The Purdue University School of Veterinary Medicine conducted a study of the incineration of animal carcasses containing plastic coated radioactive microspheres labelled with ^{46}Sc (Landalt 1983). An EPA Method 5 approved sampling system was used to monitor the stack effluents during the 48 minute sampling period. The amount of activity recovered from the ash were reported to be 97.9 ±7.6%. No specific effluent data was reported.

Katsikis et. al (1984) reported on the licensing, design and use of a low level radioactive waste incinerator in North Carolina. The authors speculated that complete combustion of the waste would result in effluents consisting of CO_2, H_2O, and SO_2, with very little activity remaining in the ash. A dual chamber, controlled air incinerator operating at 1850°F in the lower chamber and 2000°F in the upper chamber, was used in the study. No activity was found in the ash; therefore, it was concluded that all the activity was released into the effluents.

In 1985, Purdue University published data concerning the release of effluent radioactivity during the incineration of animal carcasses containing microspheres (Brekke 1985). Tin-113,^{153}Gd, ^{57}Co, ^{95}Nb, and ^{103}Ru were the nuclides studied. The report concludes that less than 17% of the incinerated radioactivity was released to the effluents for all nuclides tested.

The solubility of seventeen radionuclides in ash, which resulted from the incineration of animal carcasses, was investigated at the Mayo Clinic in 1985 (Classic 1985). Twenty aliquots of ash, each weighing 0.1 gram, were placed into 5 ml of distilled water. After a period of one hour, samples were removed and counted using either a gamma or liquid scintillation counter. The percent retention of radioactivity in the ash for ^{35}S was reported as 0.0%.

An investigation of the fate of ^{14}C and ^{3}H during the incineration of liquid, low level radioactive waste, conducted at the National Institute of Environmental Health Sciences (NIEHS), reported a mean percent retention in the ash of less than 0.01% (Hamrick 1986). Swipe tests of the refractory surface indicated no significant activity above background. The result of that effluent sampling suggested that most of the activity incinerated was released into the atmosphere as tritiated water vapor or ^{14}C-labeled carbon dioxide.

The literature search identified various papers concerning the incineration of low level radioactive waste. The majority of the papers, however, addressed the topic of incineration as an economic feasibility study, rather than a scientific investigation into the fate of the incinerated radionuclides. The search did identify some documentation investigating the fate of the incinerated nuclides (^{14}C and ^{3}H), but it produced no published experimental data relating the activity in the effluents and residual ash during the incineration of ^{35}S labeled radioactive waste. The previous studies which investigated incineration of ^{35}S made the assumption that the activity is either released in the effluents or retained by the ash. Therefore, only the ash was sampled as an attempt to assess the fate of the radioactivity. The studies in the literature reported retention values ranging from 0 to 90%. One purpose of this study was to investigate the range of ash retention values.

Governmental Regulations

Two Federal agencies are responsible for regulating low-level radioactive waste incineration; The Nuclear Regulatory Commission (NRC) and the Environmental Protection Agency (EPA). The Code of Federal Regulations (10 CFR 20) contains the NRC regulations for allowed effluent radionuclide concentration. The EPA regulations concerning nonradioactive emissions are found in 40 CFR 60. The permit for NIEHS to operate an incinerator was granted by the

Environmental Management Commission under Article 21B, Chapter 143. The Nuclear Regulatory Commission also issued a license to NIEHS for operation of an incinerator (Knapp 1984).

MATERIALS AND METHODS

Method of Investigation

A modified EPA Method 5 isokinetic stack sampling system was used to monitor the gaseous effluents released during the incineration of ^{35}S labeled liquid waste. The residual ash also was sampled in order to confirm the effluent data for each trial burn. A total of eleven trial burns were conducted. During eight of the trial burns, samples of ^{35}S labeled methionine with known activities were incinerated. Aqueous solutions of ^{35}S labeled sulphates were incinerated in the remaining three burns.

Incinerator Characteristics

A modified, dual chamber, pathological waste incinerator is used by the National Institute of Environmental Health Sciences (NIEHS) to combust type IV (pathological) waste. The incinerator (Consumate Systems INC., Richmond, Virginia) which is fired by four natural gas burners in the lower chamber and one burner in the upper chamber, is a controlled air incinerator licensed by the state of North Carolina. The license limits the charge rate to 350 pounds of type IV waste per hour. NIEHS elects to limit the charge rate to 200 pounds per hour.

Waste is loaded into the primary chamber by means of a hydraulic ram loader. In the primary chamber, the waste is ignited in a starved air atmosphere by four natural gas burners which maintain the temperature at 1400°F. If this temperature is exceeded some burners will shut down. Due to the starved air environment, the oxygen concentration is held below the stoichiometric point (McRee 1986). This condition results in only partial oxidation of the incinerated waste (Koenig 1986).

The waste gas passes into the secondary chamber where excess air is introduced. The elevated oxygen concentration and high temperature (1600°F) maintained by the upper burner allows for complete oxidation of the waste gas. Under optimal operating conditions, carbon dioxide and water are the major products of combustion (McRee 1986). As the effluents ascend the exhaust stack, additional air is introduced through the air induction collar, thus cooling and diluting the gas before it is released into the environment.

The pathological incinerator has been fitted with two sampling ports situated ninety degrees apart. The sampling ports are constructed from 4 inch diameter pipe and are flush mounted to the inside surface of the 22 inch inside diameter refractory. The ports are located 42 inches above the roof and 66 inches above the air induction collar. This configuration of sampling ports (Fig. 1) allows sampling in accordance with Environmental Protection Agency guidelines specified in Appendix A of 40 CFR 60 (USEPA 1985).

The incinerator has a one hour warm-up period which insures proper chamber temperature. After the final charging, an automatic five hour burn down cycle is initiated. Table 1 lists some incinerator specifications.

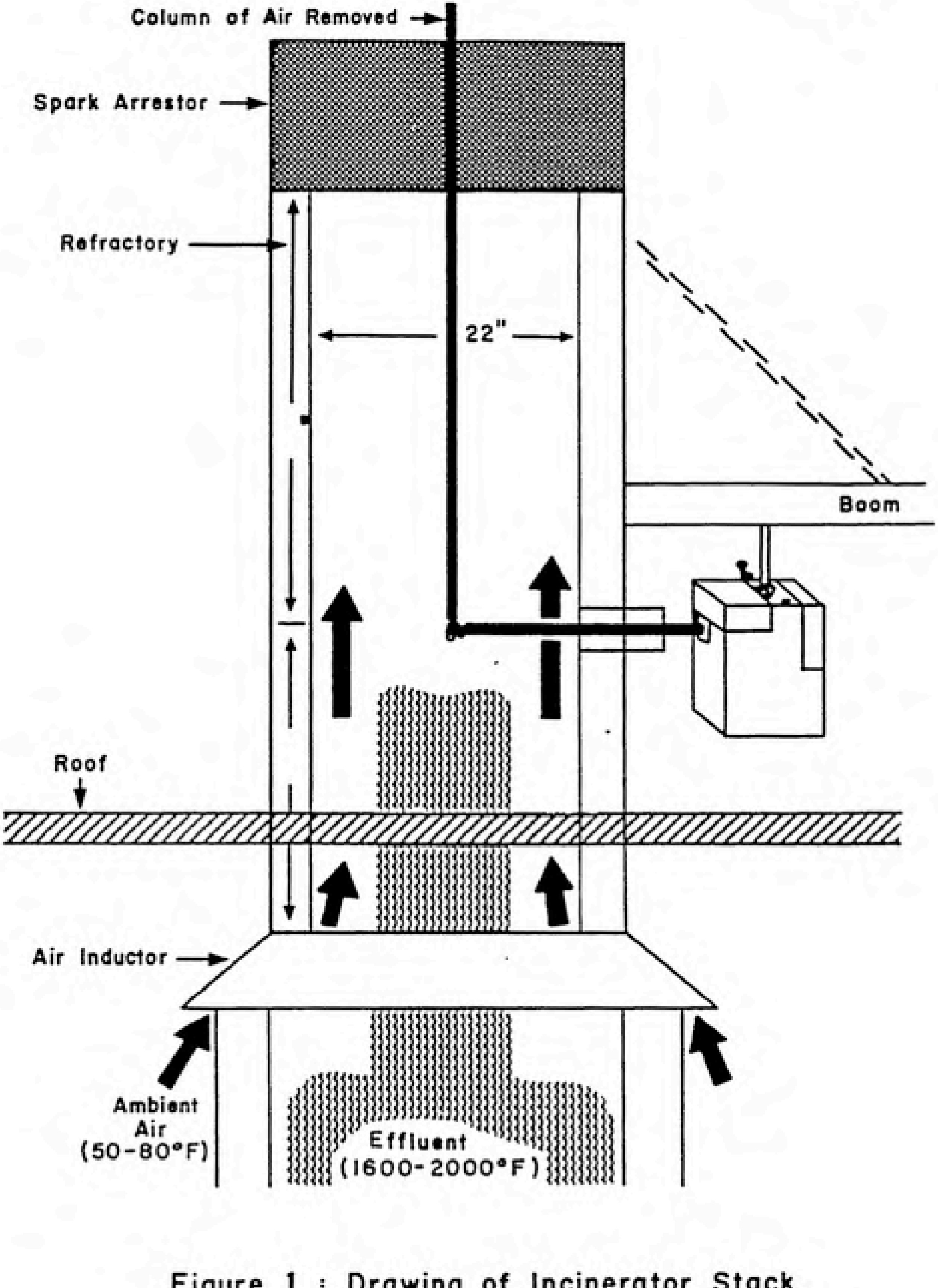


Figure 1 : Drawing of Incinerator Stack

Table 1. Incinerator Specifications

Model	Consumat C-125P
Fuel	Natural gas
Fuel Feed Rate	
a) Primary Burners	350,000 BTU/hr
b) Secondary Burner	1,000,000 BTU/hr
Lower Chamber Volume	170 ft^3
Upper Chamber Volume	102 ft^3
Lower Chamber Temperature	1400°F
Upper Chamber Temperature	1600-2000°F
Waste Type Burned	Type IV
Waste Charge Rate	350 lbs/hr

Definition of Isokinetic Sampling

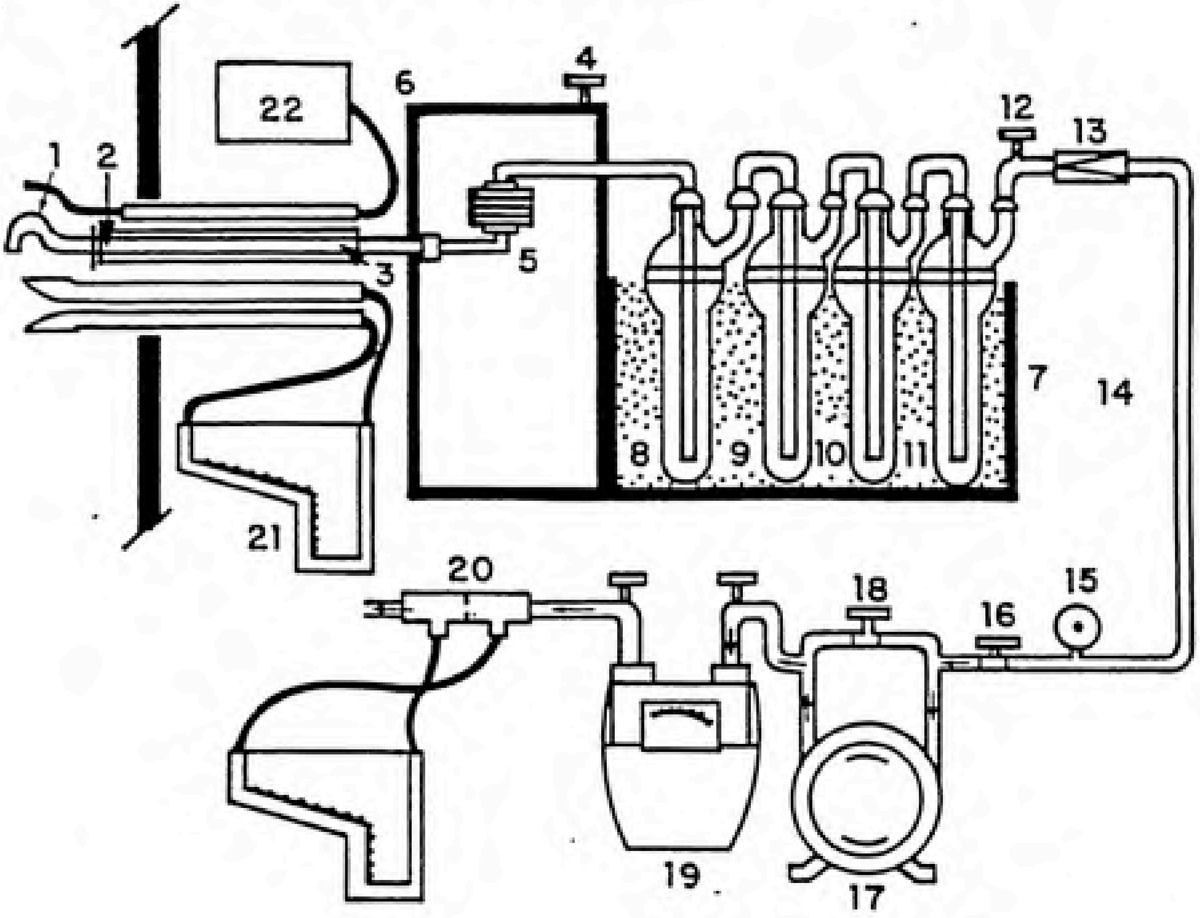
Isokinetic sampling is an equal, uniform sampling of particulates and gases in motion within the stack (USEPA 1979a). It provides an unbiased analysis of pollutants being emitted from the source and closely evaluates the various parameters that exists during the sampling interval. Isokinetic conditions are met when the velocity, V_n, of the gas stream entering the nozzle equals the velocity of the gas ascending the stack, V_s. If V_n is not equal to V_s the particulate concentration data derived from the sampling process will be biased, either positively or negatively depending on the relationship of V_n and V_s. Although the focus of this project was not directed towards particulate emissions determination, an isokinetic sampling system was selected because it allows for reliable effluent monitoring.

Sampling System

An EPA Method 5 approved stack sampler (Nutech Corporation; Durham, North Carolina, Model 201, Serial No.93-39) was chosen for stack sampling. The sampling train (Fig. 2) is organized into three different components: an in-stack sampling probe, an out of stack sample case and a meter console.

The sampling probe consists of a sampling nozzle, a S-type pitot tube to measure the velocity pressure, V_p, stack gas temperature sensor, a sampling probe sheath, and a heated sampling probe liner.

Figure 2: Schematic of EPA Method 5



1. Sampling nozzle
2. Sampling probe sheath
3. Heated sample probe liner
4. Filter compartment thermometer
5. Out of stack filter assembly
6. Heated filter compartment
7. Impinger case (ice bath)
8. First impinger
9. Second impinger
10. Third impinger
11. Fourth impinger
12. Impinger exit gas thermometer
13. Check valve to prevent back pressure
14. Vacuum line
15. Pressure gauge
16. Coarse adjustment valve
17. Leak free vacuum pump
18. By-pass valve
19. Dry gas meter with thermometer
20. Orifice meter with manometer
21. Type S pitot tube with manometer
22. Stack temerature sensor

The heated probe liner prevents condensation from occurring as the sample gas passes through the sampling probe.

The sample case (Fig. 3) consists of two chambers. A heated chamber houses a filter bell and an ice cooled chamber houses the sampling impingers. Gas from the sampling probe enters the sample case in the heated filter compartment and is cooled as it passes through the ice cooled impinger compartment.

The meter console (Fig. 4) houses the orifice meter, dry gas meter, thermometer, vacuum pump and magnehelic differential gauges. The meter console is connected to the sampling probe and sampling case via an umbilical cord.

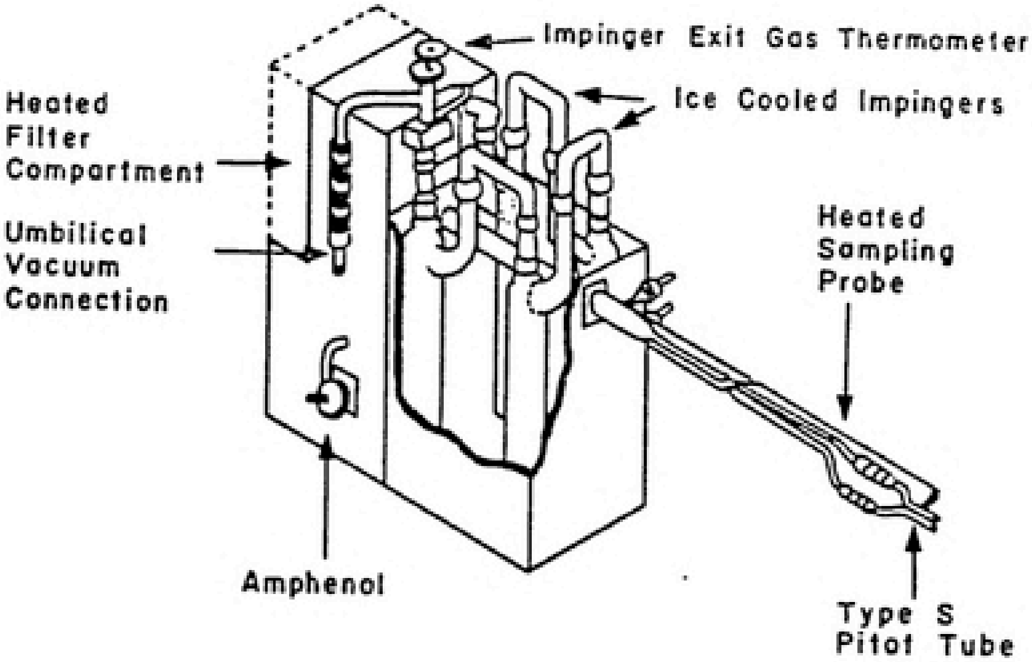


Figure 3: Impinger Case

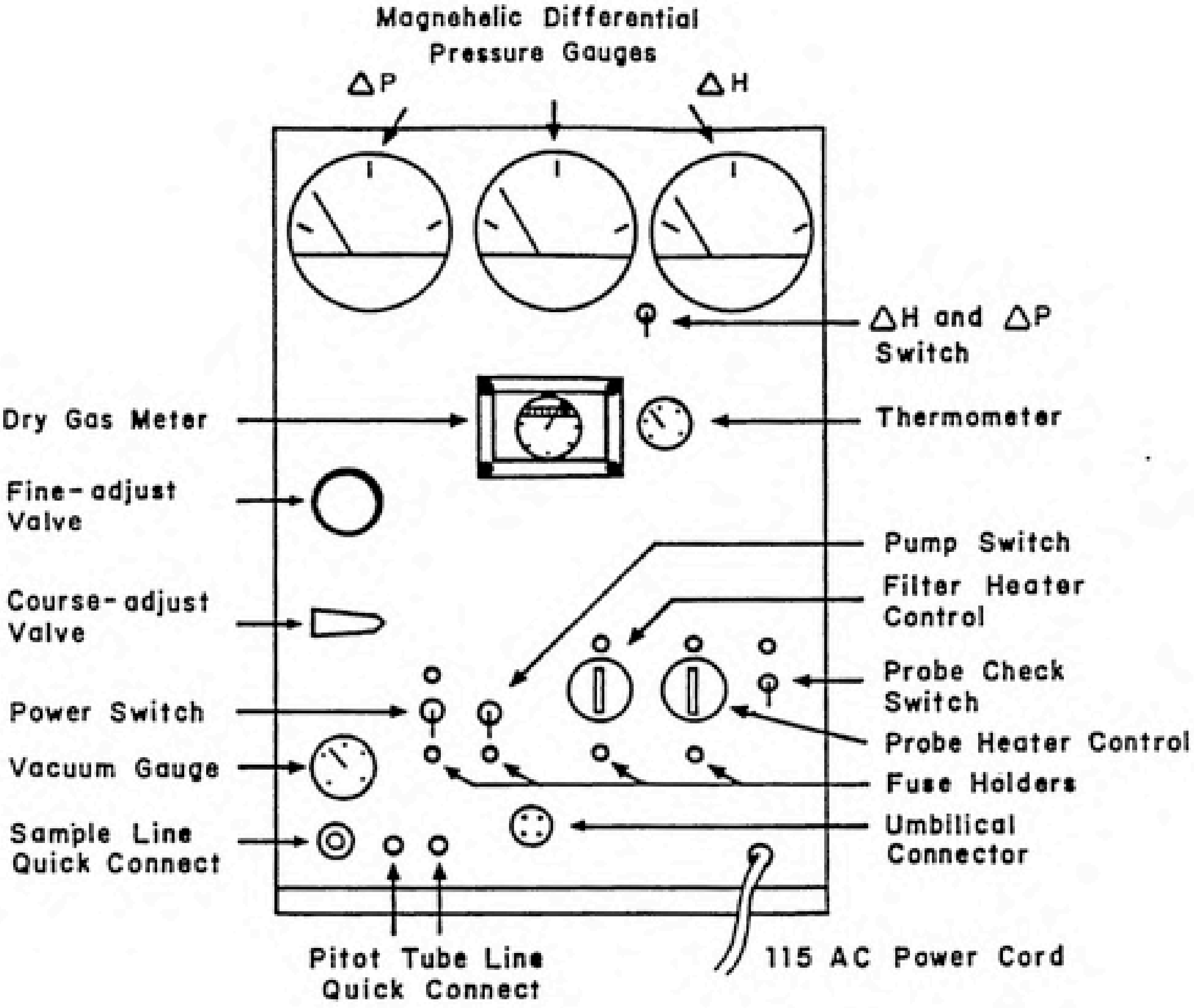


Figure 4 : Control Panel

Characterization of Conditions During Incineration

Using the Nutech sampling system, a traverse across the diameter of the stack was completed following the methodology described in 40 CFR 60 Method 1 (USEPA 1985). Data on stack gas temperature, velocity pressure and pressure differential across the orifice were collected. Using the following equations and the data described above, the velocity, flow rate and temperature were calculated.

(Eq. 1) $$V_s = K_p C_p \sqrt{\frac{T_s}{P_s M_s}} (\sqrt{\Delta p})_{av}$$

where: V_s = velocity of the stack gas
K_p = dimensional constant
C_p = pitot tube calibration coefficient
T_s = absolute temperature of the stack gas
P_s = absolute pressure of stack gas
M_s = apparent molecular weight of stack gas
Δp = average velocity pressure

(Eq. 2) $$Q_m = K_m \sqrt{\frac{T_m \Delta H}{P_m M_m}}$$

where: Q_m = volumetric flow rate
ΔH = pressure differential across the orifice
P_m = absolute pressure inches Hg
T_m = absolute temperature at the meter
K_m = proportionality factor
M_m = molecular mass of stack gas

Figures 5 and 6 illustrate the equal areas velocity and temperature profiles measured during a traverse of the stack. The velocity profile is skewed to one side as a result of air entering the stack through the 4 inch diameter sampling port. The temperature profile drops in a similar manner for the same reason. The velocity profile was used to determine the optimal point for effluent sampling. A single sampling point at the peak velocity was chosen rather than a multiple point traverse due to fluctuating emission rates characteristic of rapidly oxidizing materials. The concentration of the gas across the stack profile was assumed to be uniform, therefore an estimate of total activity released could be made. Data from both the temperature and velocity profiles were utilized in the determination of velocity and temperature correction factors (Appendix A).

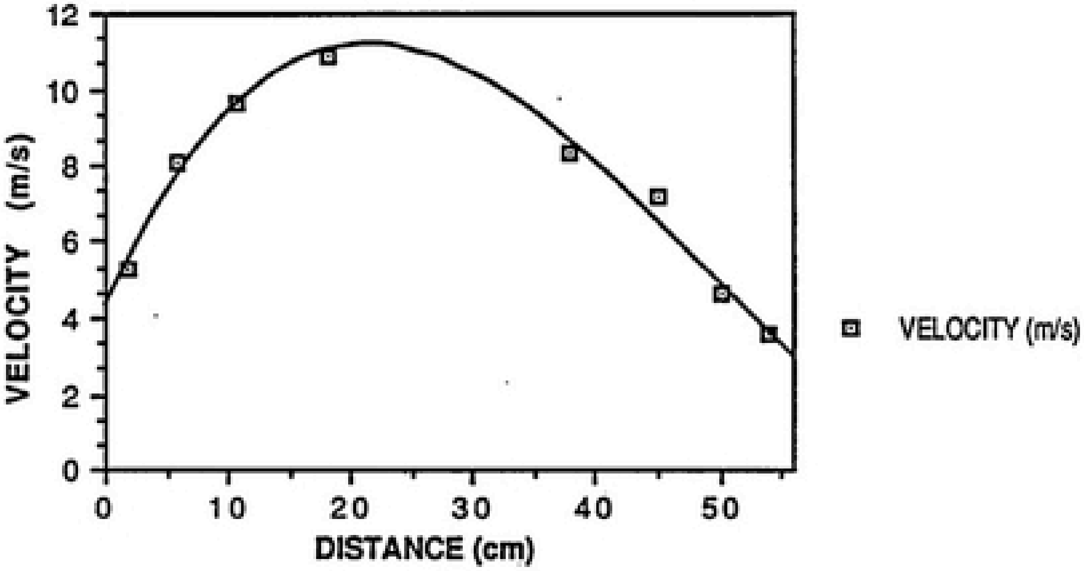


Figure 5. Stack Velocity Profile

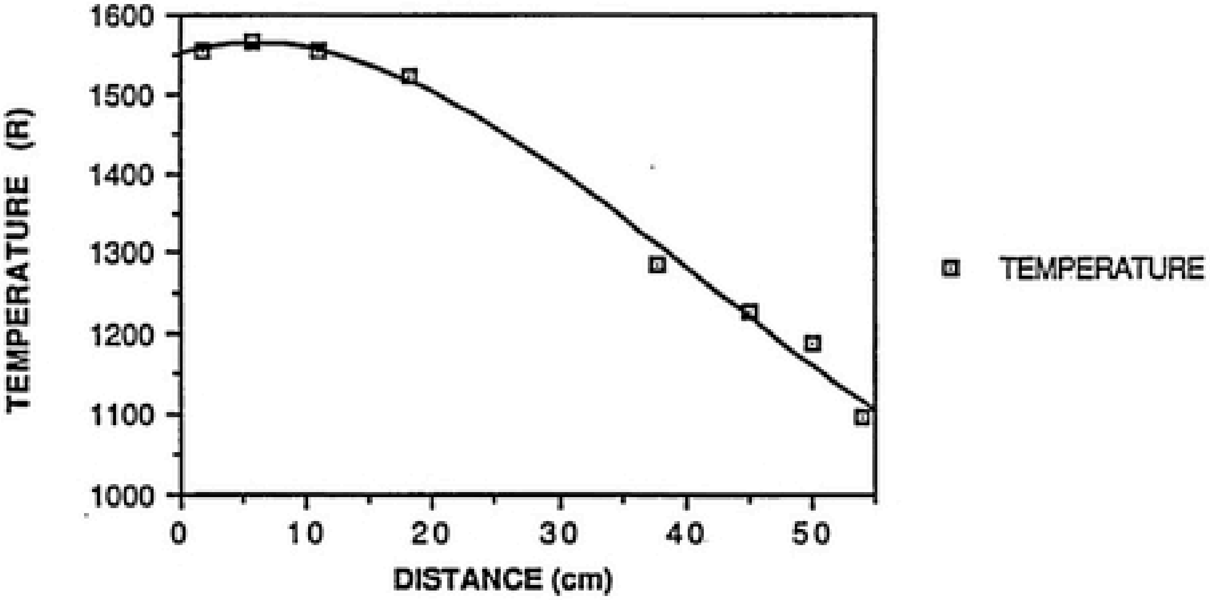


Figure 6. Stack Temperature Profile
(ºR=ºF + 460)

Radioactivity Sampling

Sample Preparation

During the data collection phase of the project, eleven trial burns were conducted. Each trial consisted of the following steps. The ^{35}S labeled waste was equally distributed among six separate samples. The volume of each sample was recorded and 1/2 ml aliquots were taken from each sample in order to determine the total activity. The activity was quantified by liquid scintillation counting. The activity was recorded and the samples were poured into individual, 3.78 liter, plastic waste jugs. Each waste jug was placed into a 1.89×10^5 cm^3 cardboard box lined with a 0.3 mm thick plastic bag. During incineration, the boxes were loaded in series spaced by a seven minute interval. The radioactive sulphur was in a methionine complex in eight of the burns and as a sulphate ion in the remaining three. During the first four trials, lab waste was the source of the incinerated ^{35}S. In the remaining seven burns the incinerated solution was made from commercially available source of ^{35}S, (Amersham/Searle Corporation, Arlington Heights, Illinois). Table 2 lists the chemical form, activity and volume for the eleven trail burns. Both the effluents and ash residue were sampled for ^{35}S content.

Effluent Sampling

The effluent gas stream was sampled using the same EPA approved sampling train employed for the velocity profile and

TABLE 2. Sample Characteristics

BURN #	FORM OF SULPHUR	TOTAL ACTIVITY (μCi)	TOTAL VOLUME (ml)
1*	M	2473	2400
2*	M	1587	675
3*	M	1254	540
4*	M	5659	1200
5	M	4276	1200
6	M	596	500
7	M	496	500
8	S	645	600
9	S	625	600
10	S	556	600
11	M	199	400

* Indicates sample made from lab waste
M=Methionine, S=Sulphate

temperature traverse. Under ideal conditions, sulphur dioxide is the major sulphur combustion product when ^{35}S is incinerated (McRee 1986). Therefore, the contents of the four impingers were selected for optimal SO_2 absorption. During the initial trail burn, the impinger contents closely followed those outlined in EPA Method 5 (USEPA 1985).

The particulates were removed from the gas stream by a 9.0 cm glass wool filter (Whatman Limited, England) housed in a filter bell which was maintained at 250°F. The filter had been desiccated and weighed prior to the burn. The gas stream was desiccated in the first impinger which contained 200 grams Silica gel in the form of of 6-16 mesh (Fisher Scientific Company, Pittsburgh, Pennsylvania). Both the second and third impingers contained 200 milliliters of liquid scintillation cocktail designed for trapping sulphur dioxide (Rauschenbach 1974). This cocktail was mixed in the lab based on methods specified in Appendix E. The fourth impinger contained 200 grams of 8 mesh Drieritte (W. A. Hammond Drieritte Company; Xenia, Ohio) desiccant. The desiccant removed excess water vapor, thus protecting the dry gas meter and vacuum pump. The four impingers were weighed before and after each trial in order to determine the amount of water vapor condensed and desiccated in the silica gel and dessicant. During the sampling interval, the four impingers were cooled in an ice bath.

After the initial trial burn, this sampling configuration was evaluated and rejected because scintillation analysis showed that essentially all of the sulphur dioxide had been trapped by the silica gel located in the first impinger. It also was noted that the trapping cocktail in the second and third impingers had degenerated resulting from evaporation caused by the high temperature of the stack gas. Based on published methods for sulphur dioxide collection (Katz 1977, USEPA 1985, Cheminoff 1978 Landalt 1983, Kusumo 1969), a new impinger configuration was designed.

A 3% hydrogen peroxide solution (3% H_2O_2) was made by diluting 100 milliliters of 30% H_2O_2 reagent with 900 milliliters of deionized, distilled H_2O (EPA 40 CFR 60 Appendix A Method 8). The silica gel in the first impinger was replaced by 200 to 250 milliliters of 3% H_2O_2 solution. The second and third impingers also contained dilute H_2O_2 of the same concentration and volume. In order to protect the vacuum pump and dry gas meter, 250 grams of 6-16 mesh silica gel were added to the fourth impinger replacing the 8 mesh Drierite desiccant used in the initial trial. The four impingers were weighed and placed into the sampling case. Ice was packed around the impingers before sampling began. The sampling train schematic is illustrated in Fig. 7. As the gas sample passed thorough the sampling train, particulates were removed by a glass filter housed in the filter bell. The filter paper had been desiccated and weighed prior to the trial burn. After particulates were removed, the gas passed into the series of impingers where the sulphur dioxide reacted with the dilute hydrogen peroxide to form H_2SO_4, thus trapping the ^{35}S.

Effluent monitoring began ten minutes prior to the loading of the first box and continued ten minutes after loading the final box. The boxes were incinerated in series at seven minute intervals. During effluent monitoring, velocity pressure, orifice pressure stack gas

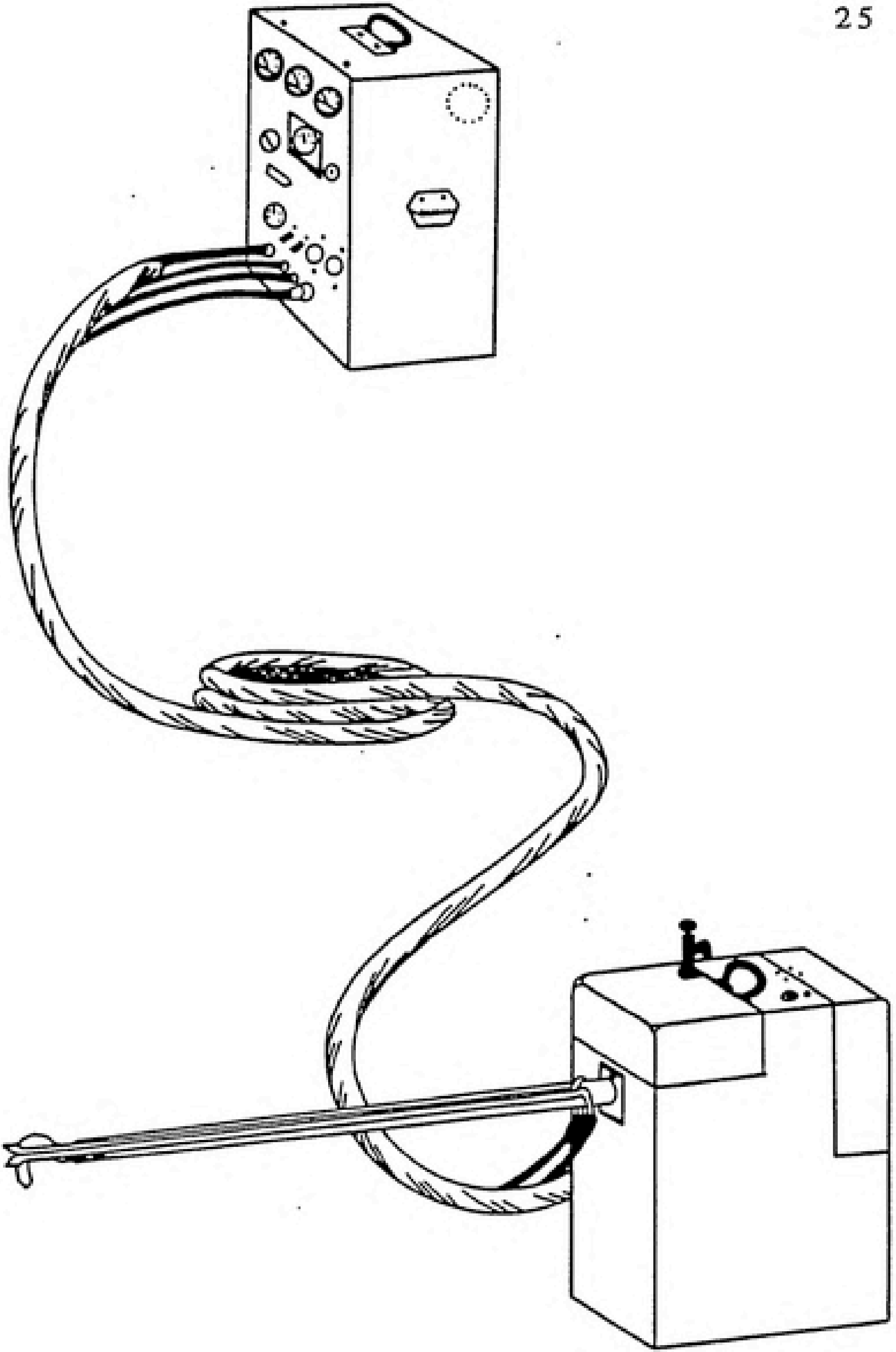


Figure 7 : Isokinetic Stack Sampler -- Model 100

temperature, temperature at the meter, and dry gas temperature data were recorded every five minutes. Dry gas volume and sampling time were also recorded. These data were used to quantify the volume of the effluents that were pulled through the sampling train (Appendix A). Upon completion of each trial burn, the masses of the impingers and their contents were determined. The change in mass for each impinger was recorded. The filter paper was removed from the filter bell, desiccated and weighed. These data were used in the determination of particulate concentration in the effluents (Appendix A).

Ash Sampling

In order to properly determine the fate of the ^{35}S during incineration, the ash was monitored for radioactivity content. To ensure that the activity remaining after the burn was due to the ^{35}S, the incinerator was cleaned by manually sweeping the refractory before each trial burn. After the trial burn was completed and the incinerator had cooled, the refractory was swept again to collect the ash residue. The mass of the ash was determined using a Metler analytical balance. Ten 200 milligram samples of ash were weighed and counted using the Packard Tricarb 4530 liquid scintillation counter. This instrument had previously been calibrated for ^{35}S determination. Using this data and the mass of the ash, the activity remaining in the ash was calculated (Appendix A).

Analytical Procedure

Liquid Scintillation Counting

Sulphur-35 is a pure beta emitter with a maximum energy of 167 keV and average energy of 55 keV. Liquid scintillation was selected as the method to asses the activity of ^{35}S because it is a simple and reliable procedure for quantifying beta activity. The Tricarb liquid scintillation counter (Model 4530 Packard Instrument Company, Downer Grove, Illinois,) allows for the determination of disintegrations per minute (DPM) and chemiluminesence correction by the external source method. By counting samples with a known activity and variable quench, this technique generates an efficiency curve specific for a desired radionuclide. Samples were made from a commercially available ^{35}S standard (Amersham Corp) and varying amounts of CCl_4 which served as a quenching agent. Originally ash was used as the source of quench, but it was determined that ash did not provide a wide range of quench values. The SIE values for the CCl_4 and Ash quench curves corresponded to similar efficiencies (Appendix F). The samples were counted and the data stored in the memory of the counter and used to determine disintegrations per minute by dividing the count rate by the efficiency determined from the quench curve. Figure 8 illustrates the calibration curve for ^{35}S.

The quench curve is specific to the radionuclide as well as the liquid scintillation cocktail. Because different counting cocktails have different efficiencies, the scintillation cocktail used to generate the quench curve should be used to count the samples. The

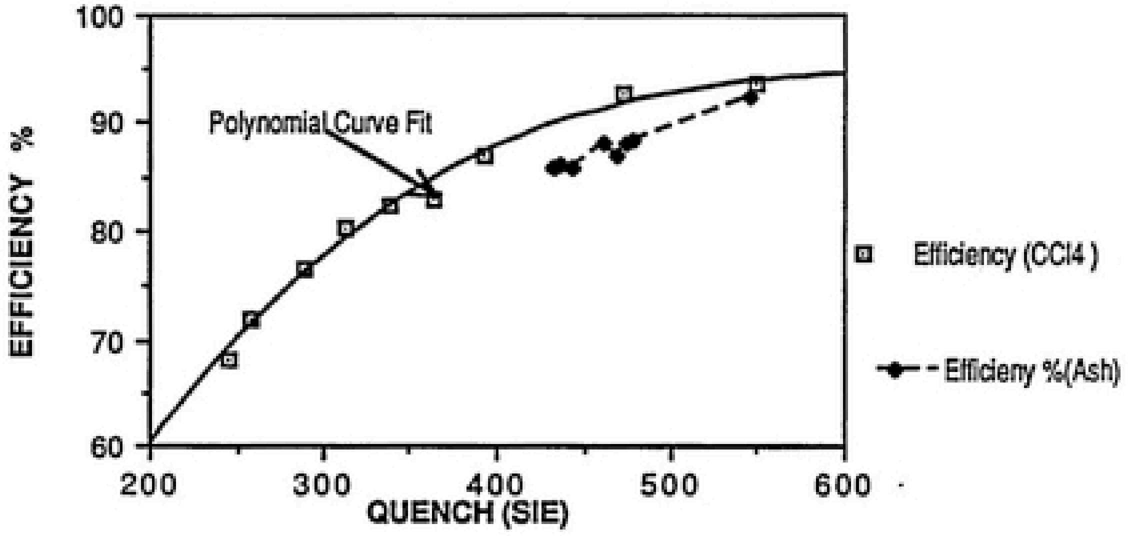


Figure 8. S-35 Quench Curve

size and shape of the counting vial should also remain constant, thus avoiding any changes in counting geometry associated with the vial (Stanely 1974). NEN formula 989 (New England Nuclear) liquid scintillation cocktail and Packard 7ml plastic vials were used throughout the project.

^{35}S In Effluents

In order to quantify the activity collected from the gaseous effluents, the volume of solution in each impinger is needed. By utilizing the mass and the density of the solution, the volume of solution in each impinger was calculated. Five one milliliter aliquots of trapping solution were pipetted from each of the three impingers and decanted into individual 7 ml liquid scintillation counting vials. Six milliliters of NEN formula 989 counting cocktail were added to each vial. To ensure a homogeneous solution, the samples were mixed using a test tube vortex. The activity per milliliter then was determined by counting each sample for a ten minute interval on the calibrated liquid scintillation counter. From these data, a mean value of DPM/ml was calculated for each impinger. By multiplying the mean DPM/ml by the volume of liquid in each impinger, the disintegrations per minute per impinger were determined. Using standard conversion factors ($2.22\text{x}10^{6}\,\frac{\text{DPM}}{\mu\text{Ci}}$), the DPM values were converted into microcuries.

The filter paper and silica gel also were counted for radioactivity in order to quantify the activity resulting from the effluents. The silica gel was counted using two methods. In the first method, samples from the silica gel were weighed and counted using the liquid scintillation counter. The total activity in the silica gel was determined by multiplying the total mass of the silica gel by the activity per gram. To eliminate possible problems with chemilluminesence due to the silica gel (Stanley 1974), a second method was developed.

In the second method, the mass of the silica gel first was determined. Distilled water then was added until saturation was exceeded. Since the density of water is about equal to 1, the change in the mass of the silica gel was used as the value for the volume of water added. One milliliter samples were removed and counted to determine the activity per milliliter of solution. By multiplying the concentration of the activity by the total volume of water, the total activity of the silica gel was determined.

To determine the activity on the filter paper, the paper was cut into six sections, placed into liquid scintillation counting vials and counted using the liquid scintillation counter. The activity per section was summed and used as the value of activity associated with the filter paper. The activity recovered in the impinger solution, the silica gel and the filter paper were used to determine the proportion of the total incinerated activity recovered from the effluents. This calculation is expressed in more detail in Appendix A.

^{35}S In The Ash

After each trail burn, the ash and residue were collected from the lower chamber of the incinerator and placed in a plastic bag of a known mass. The mass of the bag plus its contents was determined.

The change in mass was assumed be the mass of the ash. The activity remaining in the ash was determined in a manner similar to the method previously described.

Five 250 milligram samples of ash were weighed and placed into individual counting vials. The samples were chosen from five different areas of the ash to try and obtain a representative sample. Six milliliters of NEN formula 989 counting cocktail were added, and the samples were counted using the liquid scintillation counter. A mean value of DPM/250 mg was calculated and multiplied by the mass of the ash in order to determine the total activity remaining in the ash. This value was used to calculate the proportion of the total activity incinerated remaining in the ash.

Non-Routine Sampling

In order to further investigate the fate of ^{35}S during incineration of radioactive waste at the facility, several non-routine sampling procedures were performed.

Biological Oxidizer

During trial burn number ten, the gas that was exhausted from the meter console was collected in two plastic sampling bags with volumes of about 425 liters. Gas from the first sample bag was passed through a biological Material Oxidizer (Beckman Instrument, Inc. Fullerton, California 92634). By applying a vacuum, the sample gas was drawn into the combustion chamber where it was mixed with O_2 In theory, the high temperature (900°C) and excess O_2

would completely oxidize any sulphur compound to the SO_2 form. The off gas was bubbled through two impingers containing 15 milliliters of a 3% H_2O_2 solution. The SO_2 reacts with H_2O_2 to form H_2SO_4, trapping the ^{35}S in solution. The solution was analyzed for radioactivity by liquid scintillation.

Activated Charcoal

The second bag of gas collected in trial burn number ten was passed through a sample tube containing activated charcoal. Using a vacuum pump, the gas was pulled through the charcoal where the ^{35}S was trapped. Five 250 milligram samples of charcoal were weighed and counted using the liquid scintillation detection system.

Refractory Swipe Test

A swipe test was taken from the interior of the upper and lower chambers of the incinerator as well as the interior of the smoke stack. Filter discs, (4.25 cm) were inserted through the sampling port and swipes were made of the area immediately below the port. This procedure was intended to provide data for estimating the amount of activity plated out of the gaseous effluents onto the interior surface of the stack.

Statistical Analysis

Basic statistical calculations were performed on eighteen variables which had been recorded or calculated from the burn using Systat (Systat Inc. Evanston, Illinois) software package and a personal computer. For each varriable, the mean and standard deviation were calculated and the minimum, maximum, and the total number of observations were determined. The calculations were performed on all of the data from the 11 trial burns as well as the data subdivided into methionine or sulphate. Pearson correlation coefficients were calculated and Students t test statistics were determined, for several hypotheses (Appendices B, and C).

The Students t test allows for hypothesis testing by comparing the variability around the means of two sample populations. A test statistic is calculated from the following equation which utilizes the two samples means, standard deviation and number of cases in each population.

Student's Test
Statistic:

$$t = \frac{x_1 - x_2}{\sqrt{\frac{S_1^2}{n_1} + \frac{S_2^2}{n_2}}}$$

The test statistic can be used as guide in decision making. By comparing the test statistic to a predetermined critical value, the null hypothesis is either accepted or rejected. The critical region is dependent on the degrees of freedom of the sample and the size of the confidence interval (Remington 1985).

The Students t test assumes that the sample populations follow normal distributions, however, moderate departures from normality do not seriously affect results. A probability plot was generated to assess the normality of each sample population (Remington 1985), as shown in figures 9 through 11. Figure 9 illustrates the deviation from the normal expected value of the percentage of activity recovered from the ash (PCASH) for all eleven trial burns. Figure 10 similarly depicts the deviation from normality of the activity recovered from the effluents (PCGAS) for the pooled data from the methionine and sulphate burns. The deviation from the expected value of PCASH for methionine data alone is illustrated in Fig 11.

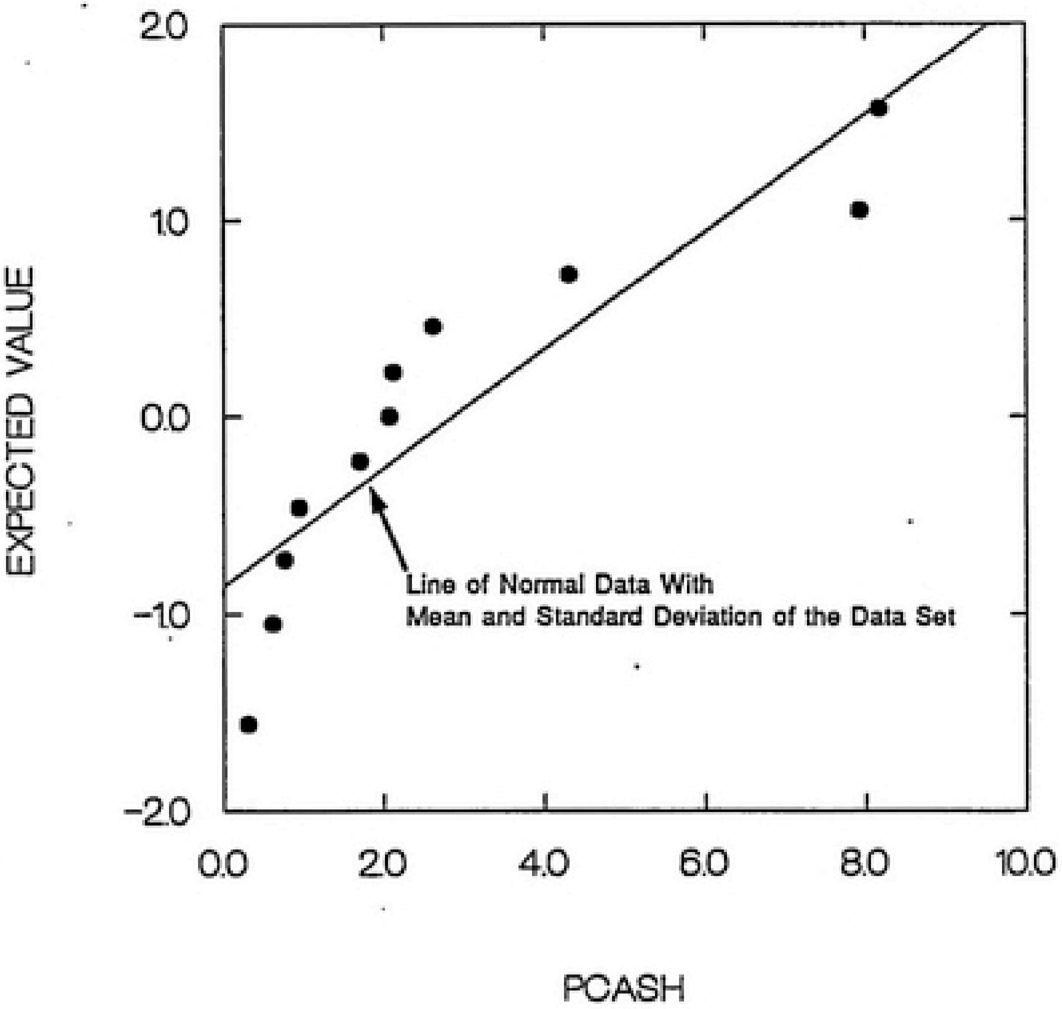


Figure 9. Pooled Samples Probability Plot % Ash
Y axis in standard deviations

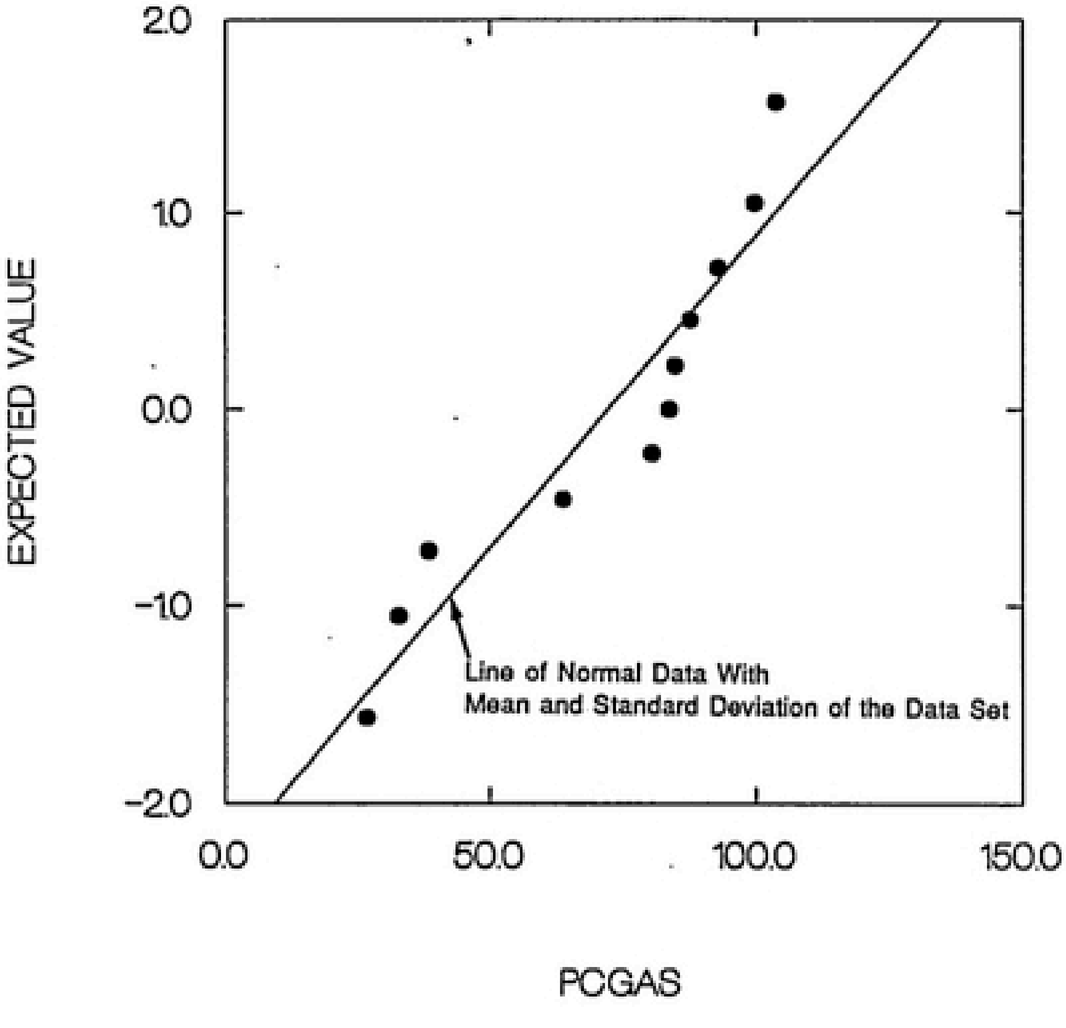


Figure 10. Pooled Samples Probability Plot % Gas
Y axis in standard deviations

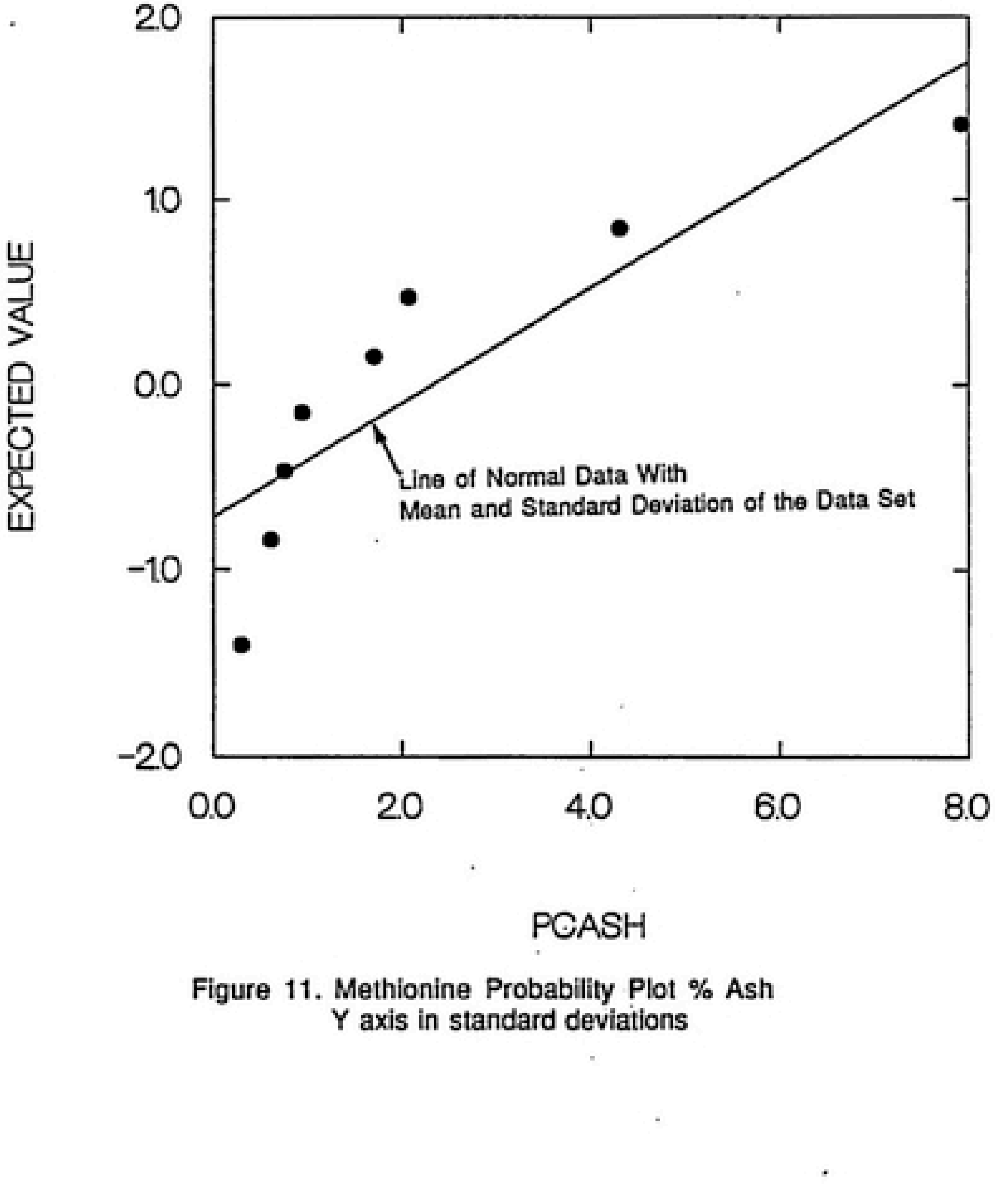


Figure 11. Methionine Probability Plot % Ash
Y axis in standard deviations

RESULTS AND DISCUSSION

Results of Hypothesis Testing

The results of the four "a-priori" questions discussed in the introduction are discussed in the following paragraphs

Question One

The first question deals with the original objective of the study, which was to determine the fate of the ^{35}S during incineration. By utilizing the computer program in Appendix D, the ash and effluent recovery efficiencies were calculated. The results of the burn calculations listed in Appendix A are summarized in Table 3. The percent effluent data represents activity trapped by the hydrogen peroxide solutions, filter paper and silica gel. The ash retention data was computed from the analysis of the ash residue which was recovered from the lower chamber of the incinerator.

The recovery efficiency (percent of total activity recovered) of the ash ranged from 8.2% to 0.29% with a mean value of 2.9% while the recovery efficiency for the gas ranged from 103% to 26.8% with a mean of 72.2%. The standard deviations of the ash and gas recoveries were 2.8 and 27.65%, respectively. When the total population is treated as two distinct groups of Methionine and Sulphate, the mean effluent recovery efficiencies are 87.02 and 32.61 respectively. The respective standard deviations are 12.47 and 5.75 respectively. The mean and standard deviation of the ash are 2.325 and 2.599 for methionine and 4.307 and 3.364 for sulphate.

Table 3: Results of Trial Burns

BURN #	FORM OF SULPHUR	ACTIVITY IN uCi	PERCENT IN EFFLUENT	PERCENT IN ASH	TOTAL PERCENT RECOVERED
1	M	2473	103.77	0.29	104.1
2	M	1587	83.59	7.93	91.5
3	M	1254	99.76	2.07	101.8
4	M	5659	92.9	0.94	93.8
5	M	4276	63.6	0.75	64.4
6	M	596	87.57	1.70	89.3
7	M	496	84.67	0.61	85.3
8	S	645	26.76	8.18	34.9
9	S	625	38.32	2.12	40.4
10	S	556	32.75	2.62	35.4
11	M	199	80.33	4.31	84.6

M=Methionine
S=Sulphate

Question Two

The question, "does the proportion of the total activity incinerated recovered from the ash and the effluents depend on the chemical form of the activity incinerated?" can be written in the form of the following null hypothesis (Ho).

$$\text{Ho: } \left(\begin{array}{c}\text{Proportion of Total} \\ \text{Activity in Ash}\end{array}\right)_{\text{Sulphate}} = \left(\begin{array}{c}\text{Proportionof Total} \\ \text{Activity in Ash}\end{array}\right)_{\text{Methionine}}$$

The value of the computed Students t test statistic for the percent of the activity recovered from the ash for the two chemical forms of sulphur was 1.050. Based on the data and the value of the test statistic, there is insufficient evidence to reject the above null hypothesis ($p>0.05$). This suggests that the proportion of activity remaining in the ash does not depend on the chemical form of the material incinerated. This is an important finding because from data reported by Bush (1973), we had previously believed that the proportion remaining in the ash for sulphates would be larger than for methionine.

The above null hypothesis can also be written to investigate the proportion in the gas by substituting "Ash" with "Gas". The test statistic for the gas, 7.095, falls in the critical region of the distribution and therefore there is sufficient evidence to reject the null hypothesis that the proportion of the total activity in the effluents for methionine equals that for sulphates. The values of % effluent recovery listed in Table 3 further support these two conclusions.

Question Three

Figures 12 and 13 show the relationship between the percent of the total activity recovered from the gas and ash as a function of the total incinerated activity for methionine and sulphate samples separately. Figure 14 in a similar manner depicts the percentage of total incinerated activity which was recovered from the gas and ash as a function of sample volume. There appears not to be a significant relationship for the methionine recovery in the gas and the ash either as a function of total incinerated activity or volume of the sample. The sulphate data are less conclusive since there were only three trial burns using this compound.

The results from the Pearson Correlation values for percent recovered from the gas and ash versus activity for methionine were -.100 and -.332, respectively. For sulphates, the Pearson Correlations in the same category were -.234 and 0.617. When the correlation was calculated for percent recovery from the gas and ash versus sample volume, the results were .366 and .402 for methionine. The correlation of sulphate recovery and sample volume was not calculated since the volume remained constant in all three trials.

These correlation values indicate that there is no significant linear relationship between these variables. In other words, the change in sample volume or total uCi incinerated appears to have no significant effect on percentage of activity recovered from the gas or from the ash for either compound.

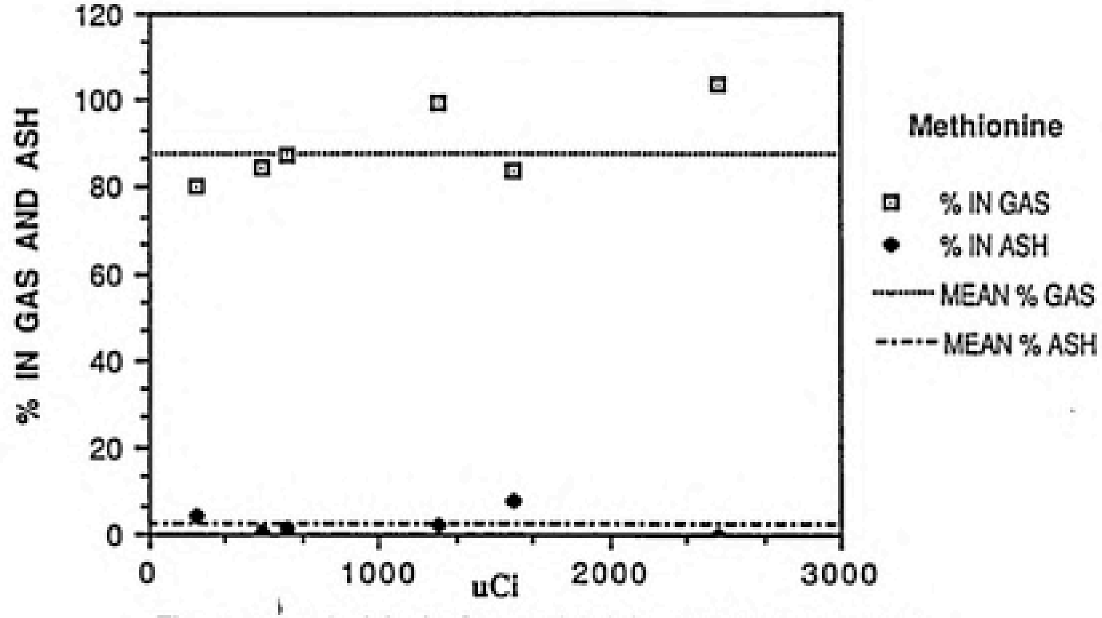


Figure 12: %Activity in Gas and Ash Vs Activity Incinerated

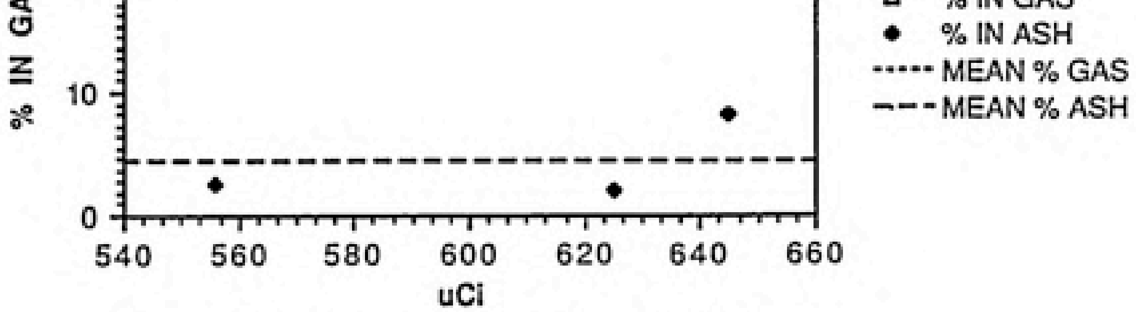


Figure 13.%Activity in Gas and Ash Vs Activity Incinerated

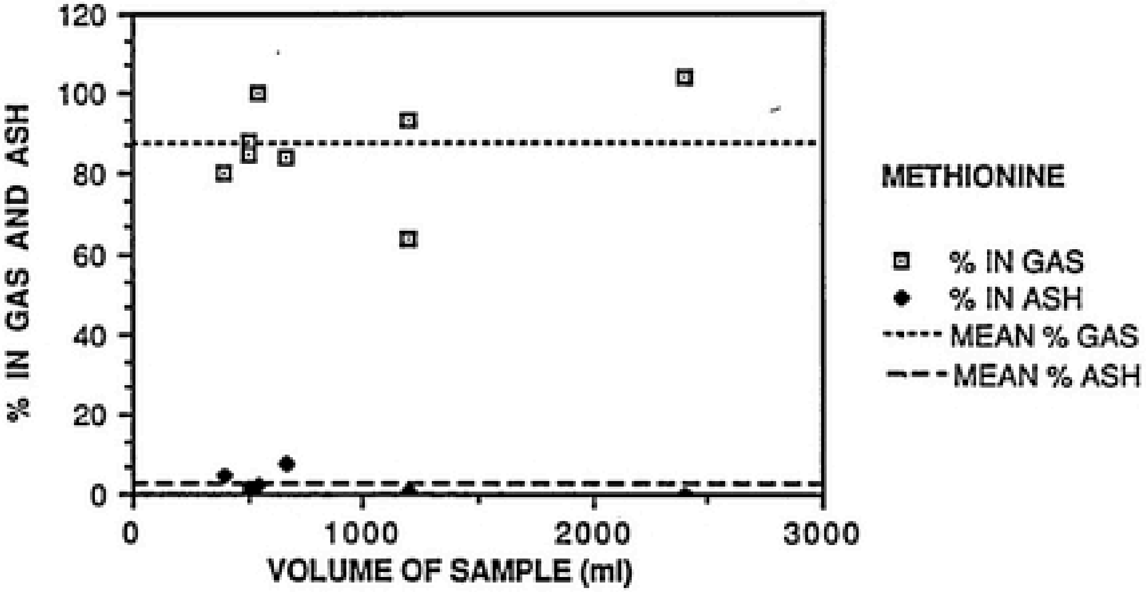


Figure 14. %Activity in Gas and Ash Vs Sample Volume

Based on the insufficient Pearson Correlation values and scatter diagrams, it maybe concluded that for those data the percentages recovered from the ash and gas were independent of the volume of sample incinerated and total activity incinerated.

Question Four

The final question addressed the assumption that the activity incinerated could be accounted for in either the ash or the effluents. Stated in the following form the hypothesis is:

$$\begin{array}{l}\text{PROPORTION OF}\\ \text{TOTAL ACTIVITIY}_{\text{ASH}}\end{array} + \begin{array}{l}\text{PROPORTION OF}\\ \text{TOTAL ACTIVITY}_{\text{GAS}}\end{array} = 1$$

In order to evaluate this question the above formula was rewritten as the following null hypothesis.

$$H_{0}\text{: (\% IN EFFLUENTS) = (100 - \%ASH)}$$

This null hypothesis was used for the sulphate and methionine data separately, as well as the combined sample population. The Students t test statistic for each group is reported in Appendix C

The t value of 2.447 for the methionine data is not in the critical region defined by the 95% confidence interval. Therefore, there is not sufficient evidence to reject the null hypothesis. The value of the test statistic for the sulphate population, which was 35.720, falls in the critical region. Based on this value and a 95% confidence interval, there is sufficient evidence to reject the null hypothesis.

These results indicate that the sulphate activity is not totally accounted for in the ash and effluents. Although the null hypothesis for methionine was not rejected, it was on the boarder line at the 95% confidence interval and, therefore, it too should be investigated further.

Discussion of Variability and Error

The fluctuation in the recovery efficiency and the results of the Students t tests data indicate that further investigation to determine the source of variation is needed. Although the data for the ash recovery had a higher deviation when compared to its mean, the effluent deviation was the area which created more concern.

An obvious explanation for the fluctuating effluent recovery could be a malfunction of the sampling system. A leak in the sampling train, for example, could be responsible for spurious results. This explanation, however, was rejected by the results of leak tests which were performed on the sampling train before and after each trial burn. Pearson correlation coefficients calculated on the dry gas volume versus time, and flow rate versus dry gas volume, suggest a strong, positive correlation, thus indicating that the system was functioning properly. A second hypothesis to explain the variability of recovery was that there was incomplete oxidation of the incinerated samples resulting in an increase in the SO_3 proportion of the waste gas (Koenig 1986, McRee 1986). As mentioned earlier, the sampling solutions of H_2O_2 are designed for SO_2 trapping. During the first eight trial burns, the activity recovered from the effluents was trapped in the first two H_2O_2 impingers. Due to this trend, the H_2O_2 in the third impinger was replaced by an eighty percent (80%)

isopropanol solution as an attempt to trap SO_3 (Cheminoff 1978, Katz 1977). This sampling modification was utilized in burns nine through eleven. In each case, however, no activity above background was detected. In addition to the SO_3 modification, during trial burn ten the exhaust gas was collected in two plastic air sampling bags as it exited the sampling train via the vacuum pump. Analysis of the gas by biological oxidation and activated charcoal also resulted in no measurable activity.

In a further attempt to explain the recovery efficiency discrepancies, swipe tests were taken from the lower and upper chambers of the incinerator as well as the stack. Although the lower and upper chambers indicated no activity, the stack swipe test picked up ^{35}S activity recording 150-200 CPM above background. These data seem to indicate that some activity had plated out along the refractory wall of the stack. The lack of measurable activity in the upper and lower chambers may be explained by the high temperatures associated with these areas. As the effluents ascend the stack, air is introduced through the air induction collar. The effluents, therefore, are much cooler than the same effluents located in the upper and lower chambers, thus increasing the potential for plating onto the interior stack surfaces.

The variation of the effluent recovery efficiency data can be explained to some degree by treating the total sample population (all eleven burns) as two discrete populations. The standard deviations of the effluent recovery efficiency for methionine and sulphate are much smaller than the standard deviation in effluent recovery efficieny calculated for the entire sample poulation. From the values listed in Table 4, it is evident that the fluctuation in the effluent recovery efficiency can be attributed to the consistently low values of the sulphate samples. This evidence, along with the results of analyzing question number two, indicates a fluctuation in effluent recovery efficiency due to chemical form.

Another factor that must be considered is the possibility of error resulting from the ash data. The Pearson correlation values for ash concentration and gas concentration versus total incinerated activity are 0.488 and 0.966, respectively. The value of 0.966 for the gas concentration indicates a significant positive correlation exists (p=0.95). If the amount of activity incinerated increases then the concentration of the activity in the gas increases. The value of 0.488 for the ash concentration suggests that there is not sufficient evidence to establish a relationship between concentration in ash and activity incinerated. Figures 15 and 16 further illustrate this idea. These results are important. The source of the ash (cardboard boxes and plastic jugs and bags) was consistent in all eleven trial burns. With the source of the ash held constant, one would expect the activity concentration in the ash, and total activity incinerated, to have a linear relationship. This was not shown conclusively to be the case, which implies that there could be a source of error in the ash quantification. Figure 15 shows that two points in particular are outliers outside the 95% confidence interval. Without these points the relationship of ash concentration and total incinerated activity would be more significant.

Table 4. Mean and Standard Deviation of Recovery Data

	MEAN(%)	STANDARD DEVIATION(%)
Methionine (M)		
ash	2.325	2.599
gas	87.02	12.459
Sulphate (S)		
ash	4.307	3.364
gas	32.61	5.781
M and S Data Pooled		
ash	2.866	2.802
gas	72.18	27.6

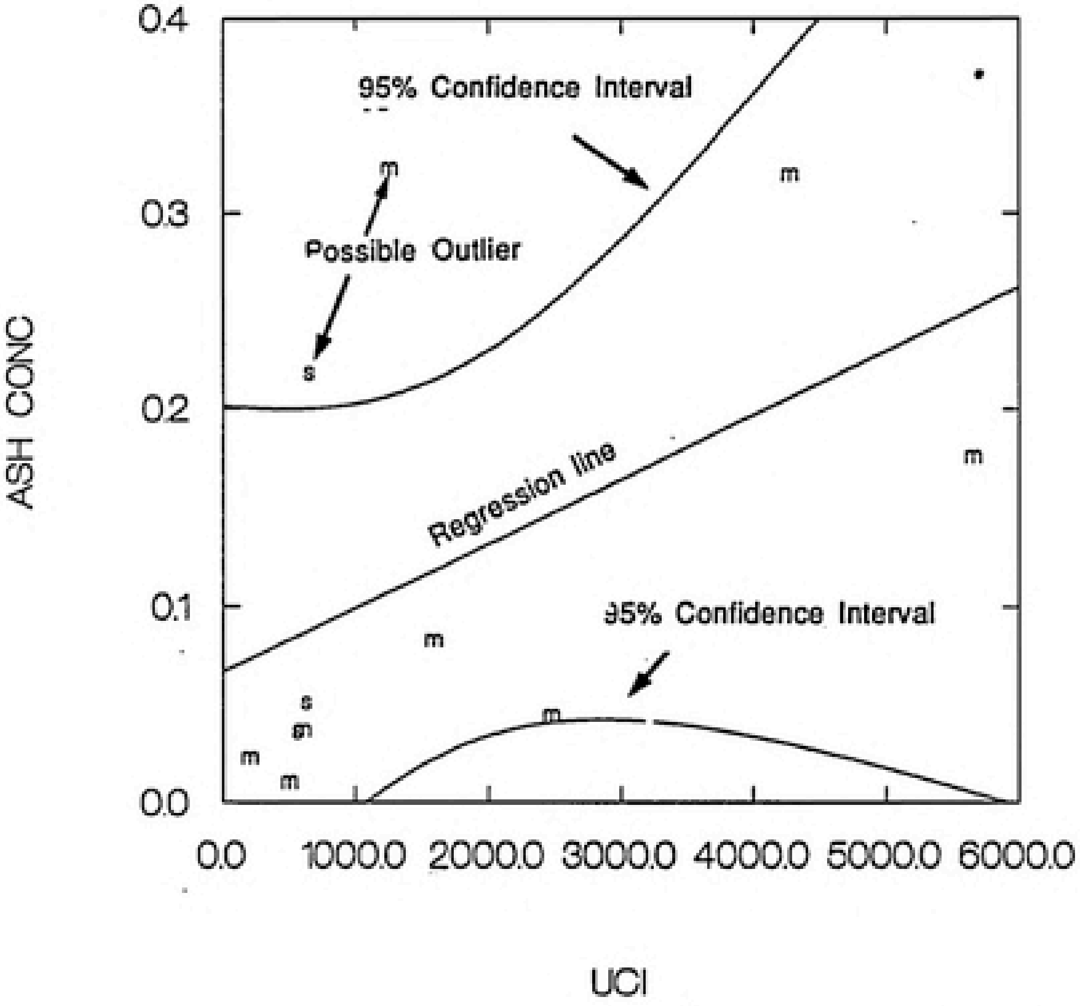


Figure 15.Ash Concentration (µCi/g) Versus Activity Incinerated

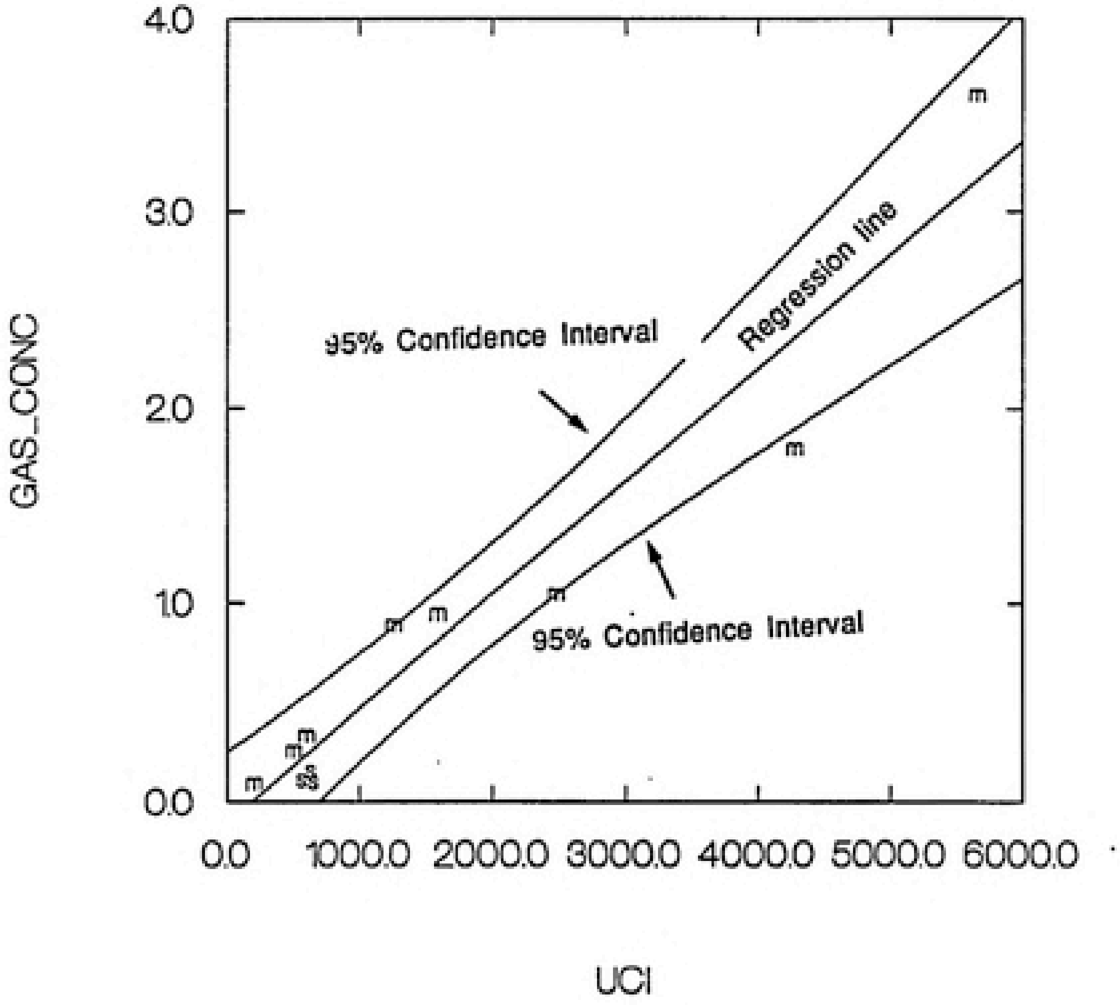


Figure 16. Gas Concentration (μCi/m^3)Versus Activity Incinerated

Figure 16, which plots the activity concentration in the gas versus activity incinerated, shows that there is a significant linear relationship.

One explanation of error in the ash data is inhomogeneity of the ash samples. The probability plots (Figs. 9-11) show that the ash data for pooled sample population and methionine fluctuate from the expected value more than the effluent data of the pooled sample population. The quantification of the effluent activity results from the trapping of the gas in H_2O_2, a homogeneous medium. Ash, however, is not in solution and is therefore the ^{35}S is distributed less homogeneous. When the activity in the ash was determined, samples were randomly selected in an attempt to obtain a representative sample. However, the low Pearson correlation data suggests that either there is no relationship or that this method of ash sampling was not very successful.

CONCLUSIONS

This investigation showed that the monitoring of ^{35}S during incineration is not a simple task. The fact that the swipe test of the stack indicated activity plating out can be used to explain, to some degree, the fluctuating recovery efficiencies. The data presented here also indicate that the effluent recovery efficiency is dependent on the chemical form of the incinerated material. The plot of percent activity recovered from the gas versus activity for the two chemical forms (Figs. 12 and 13) further supports this conclusion. It seems that the sulphate form has a higher affinity for plating out onto the interior surfaces of the incinerator stack. Further investigation into this matter is necessary in order to determine whether or not this is an attribute of the sulphates.

The result of the exhaust gas analysis implies that the sampling system satisfactorily trapped the ^{35}S in the effluents. Therefore the fluctuation of the recovery efficiency data can not be attributed to poor trapping efficiency of the 3% H_2O_2 solution.

This study also illustrated the necessity of developing a reliable ash sampling system. I feel that the counting system used to quantify the activity in the ash performed well. However, the ash sampling procedure needs further investigation in order to develop a method of homogeneous sampling.

LIST OF REFERENCES

Brekke D D., Landolt R R., and Zimmerman N J., "Measurement of Effluent Radioactivity during the Incineration of Carcasses Containing Radioactive Microspheres", Health Physics, Vol. 48 No. 3, pp 339-341, 1985

Bush D. and Hundal R.S., "The Fate of Radioactive Materials Burned in an Institutional Incinerator", Health Physics ,Vol. 24, No. 5, pp 564-568,1973

Cheminoff P.N. and Morresi A.C., Air Pollution Sampling and Analysis Deskbook, 1st. ed., Vol. 1, Ann Arbor Science, Ann Arbor, 1978, p 707

Cook F.C.,Jr. and Richard M.L., "Incineration of Low-Level Radiactive Wastes A Planning Approach", Proceedings Industrial Waste Conference, 38th, pp. 251-260,1984

Classic K., Gross G. and Vetter R.J., "Solubility of Radionuclides in Ash from the Incineration of Animals", Health Physics, Vol. 49, No. 6, pp 1270-1271,1985

Glauberman H. and Loysen P., "The Use of Commercial Incinerators for the Volume Reduction of Radioactively Contaminated Combustible Wastes", Health Physics, Vol 10, pp237-241,1964

Hamrick P.E., Knapp S.J., Parker M.G. and Watson J.E., "Incineration and Monitoring of Low Level ^{3}H and ^{14}C Wastes at a Biological Research Institution", Health Physics, Vol. 51, No. 4, pp 469-478, 1986

Katsikis E.P., Worrell L.E. and Lainhurt M.S.,"Incineration of Low Level Radioactive Waste and Scintillation Vials Under Resource Conservation and Recovery Act", Radiation Risk, Protection Int. Congr., 6th Vol. 3, pp 1397-1400, 1984

Katz M., ed., Methods of Air Sampling and Analysis, 2nd ed., Vol.1, American Public Health Association, Washington D.C., 1977 P 707

Knapp S. J., "Monitoring the High Temperature Incineration of ^{3}H and ^{14}C Liquid Scintillation Wastes," Health and Safety of Report, National Institute of Environmental Health Sciences, Research Triangle Park, North Carolina, 1984

Koenig R.A. and Vauruska J.S., "Incinerators used for Radwaste", Preliminary Program, Incineration Basics Course, Incineration of Low Level and Mixed Wastes: Charlotte, North Carolina April 22-25, 1986

Kusuno K., Shimada S., Jomoto Y., Kanayoma Y., Otabe S., Yasunaga M., Ichijima I. and Isshiki H., "Study on the Behavior of Fuel Oil Sulphur in Blast Furnace by Radioisotope ^{35}S", Iron and Steel Institute Japan-Trans, Vol.9 No.4, pp 297-305, 1969

Landolt R.R., Barton T.P., Born G.S., Morris V.R., Vetter R.J. and Zimmerman N.J., "Evaluation of a Small, Inexpensive Incinerator for Institutional Radioactive Waste", Health Physics, Vol. 44, No.6, pp. 671-675, 1983

Machis A. and Geyer J.L., "Burning Radioactive Wastes in Institutional Incinerators", American Industrial Hygiene Association Quarterly, Vol. 13, pp 199-205, 1952

McRee R.E., "Combustion", Preliminary Program, Incineration Basics Course, Incineration of Low Level and Mixed Wastes: Charlotte North Carolina April 22-25, 1986

Parker G.D., "Incineration ofHazardous and Low-Level Radioactive Waste", Pollution Engeneering vol.13, No. 22, 1981

Rauschenbach P. and Simon H., "Sample Preparation with an Automated Oxygen Flask Combustion Apparatus for Liquid Scintillation Counting of ^{3}H, ^{14}C and/or ^{35}S Labelled Material", Liquid Scintillation Counting, Vol. Date 3 pp158-163, 1973

Remington R. D. and Schork M. A., "Hypothesis Testing", Statistics with Applications to the Biological and Health Sciences, 2nd. ed. Prentice-Hall,Inc., New Jresey, 1985, pp. 165-204

Stanley P.F.and Scoggin B.A., ed. " Choice of Counting Vial For Liquid Scintillation : A Review ", Liquid Scintillation Counting, Recent Developments, 1st. ed., Vol. 1, Academic Press, New York, 1974, pp 431-446

USEPA 1979a United States Environmental Protection Agency, Source Sampling for Particulate Pollutants: Student Manual for APTI Course 450, EPA 450/2-79-006, 1979.

USEPA 1979b United States Environmental Protection Agency, Source Sampling for Particulate Pollutants: Student Workbook to APTI Course 450, EPA 450/2-79-007,1979

USEPA 1985 United States Environmental Protection Agency, Code of Federal Regulations, Title 40, Part 60, 1985

Wollen R.O., Staiger J.W. and Boge R.J. "Disposal of Low Level Radioactive Waste at A Large University Incinerator", American Industrial Hygiene Association Journal, Vol. 32, No. 9, pp 625-632, 1971

APPENDIX A
EQUATIONS FOR BURN CALCULATIONS

Eq.1: DRY GAS VOLUME MEASURED BY THE DRY GAS METER CORRECTED TO STANDARD CONDITIONS **Vmstd (ft^3)**

$$Vmstd = \frac{(Vm)(Tstd)(Pbar+(H/13.6))}{(Tm)(Pstd)}$$

Where:

Vm =Dry gas volume measured by Dry Gas meter in (Ft.^{3})
Tstd =Standard absolute temperature (528^{o}R, ^{o}R=^{o}F+460)
Pbar =Barometric pressure drop at dry gas meter (in. Hg)
H =Mean Pressure drop across orifice meter in (in. Hg)
Tm =Absolute average dry gas temperature (^{o}R)
Pstd =Standard absolute pressure (29.92 in. Hg)

Eq.2: PROPORTION OF WATER VAPOR IN THE STACK GAS STREAM **Bws**

$$Bws = \frac{Vwc(std) + Vwd(std)}{Vwc(std) + Vwd(std) + Vm(std)}$$

Where:

Vwc(std)=Volume of Water condensed at standard conditions
Vwd(std)=Volume of water dessicated at standard conditions
Vm(std)=From equation 1

Eq.3: DRY GAS MOLECULAR WEIGH OF STACK GAS **Md**

$$Md=0.44(\%CO_2)+0.32(\%O_2)+0.28(\%CO+\%N_2)$$

Where:

$\%CO_2$ =percent CO_2 by volume, (1.2%)
$\%O_2$ =percent O_2 by volume, (17.8%)
$\%CO$ =percent CO by volume,.(0.1%)
$\%N_2$ =percent N_2 by volume, (80.9%)
0.44 =molecular weight of CO_2 divided by 100
0.32 =molecular weight of O_2 divided by 100

0.28 =molecular weight of CO and N_2 divided by 200

Eq.4: WET MOLECULAR WEIGHT OF STACK GAS
Ms

$$Ms=Md(1-Bws)+18(Bws)$$

Where:

Md =Dry molecular weight of stack gas
Bws=Proportion of water vapor is stack gas
18 =Molecular weight of water

Eq. 5: AVERAGE STACK GAS VELOCITY
Vs (ft/s)

$$Vs=(0.7473)(Kp)(Cp)\sqrt{\frac{T_s}{P_s M_s}}(\sqrt{\Delta p})_{av}$$

Where:

Kp=85.49
Cp=0.85
T_s=Absolute average temperature of the stack gas corrected (°R)
Δp=Average velocity pressure (in. water)
P_s=Absolute pressure of the stack gas (in. Hg)
M_s=Wet molecular weight of the stack gas (Eq. 4)
0.7473=Velocity correction factor based on velocity profile

Eq.6: DRY VOLUMETRIC STACK GAS FLOW RATE AT STD

$$Qstd=\frac{3600(1-Bws)(Vs)(A)(Tstd)(Ps)}{(Ts)(Pstd)}$$

Where:

Bws=From equation 2
Vs= Average Stack gas velocity (Eq 5)
A=Cross sectional area of stack (2.906Ft.2)
Tstd=Standard absolute stack temperature (528°R)
Ps=Absolute stack pressure (in. Hg)

Ts=Absolute average corrected stack gas temperature based on temperature profile
Pstd=29.92

Eq. 7: PERCENT EXCESS AIR

$$\%EA=\frac{\%O_2-0.5(\%CO)}{0.264(\%N2)-(\%O2)+0.5(\%CO)}\times 100$$

Where:

$\% O_2$=percent O_2 by volume
% CO=percent CO by volume
%N2=percent N2 by volume
0.5=ratio of O_2 to CO correcting for incomplete oxidation
0.264=ratio of O_2 to N_2 in air by volume

Eq.8: PERCENT ISOKINETIC VARIATION

$$\%I=\frac{(Ts)((V1c)(K)+(Vm/Tm)(Pbar+H/13.6))}{(60)(\Theta_1)(An)(Vs)(Ps)}\ X100$$

Where:

Ts=Absolute average stack temperature (oR)
V1c=Total volume of liquid collected in impingers (ml)
K=Conversion Factor (0.002669in-Hg-Ft3/ml-oR)
Vm=Dry gas volume measured at meter (dcf)
Tm=Absolute average dry gas temperature
Pbar=Barometric pressure (in. Hg)
H=Average pressure drop across the orifice meter (in Hg)
Θ_1=Total sampling time (min.)
An=Cross sectional area of nozzle (0.000716 $ft.^2$)
Vs=Average stack gas velocity (ft/S)
Ps=Absolute stack gas pressure (in Hg)
13.6=Specific gravity of mercury
60=conversion factor (S/min)

Eq. 9: PARTICULATE EMISSION CONCENTRATION CORRECTED TO 12 %CO2

Cs12 (g/dscdf)

$Cs12=12(Mn)/(Vmstd)(\%CO2)=2.3x10\text{-}4$ g/ft3

Where:

Mn=Mass of particulates collected (grams)
Vmstd= From equation 1
12=correction factor
$\%CO_2$=percent CO_2 by volume from waste

Eq. 10 DRY STACK GAS VOLUME RELEASED DURING SAMPLING AT STANDARD CONDITION

$Vq=(Qsd)(\Theta 2)=ft^3$

Where:

Qsd=Dry volumetric stack gas flow rate at standard Conditions
$\Theta 2$=Total sampling time in Hrs.

Eq. 11 TOTAL MONITORED ACTIVITY

$Am=(Ac)(Vq)/Vmstd$

Where:

Vq=Dry stack gas volume released at standard conditions
Ac=Activity collected during sampling, (µCi)
Vmstd=Dry gas volume measured by dry gas meter

Eq.12: ACTIVITY RECOVERED IN THE ASH

Aa=(µCi/g)(mass of ash collected)

Where:

µCi=Micro curies collected per gram of ash
Mass=Mass of the ash collected

Eq. 13: Recovery efficiency

A) In Effluent Collection
$Reg = Am/Ai \times 100$
Where:
Am=total activity from the gas (μCi)
Ai=total activity incinerated (μCi)

B) In Ash Collection
$Rea = Aa/Ai \times 100$
Where:
Aa=Activity of ash
Ai=Activity Incinerated

C) Total
$Ret = (Am+Aa)/Ai \times 100$
Where:
Am=total activity from the gas (μCi)
Aa=total activity from the ash (μCi)
Ai=total activity incinerated (μCi)

APPENDIX B
RESULTS OF SIMPLE STATISTICAL CALCULATIONS

Table 1: Methionine Data

VARIABLE	NUMBER OF CASES	MIN	MAX	MEAN	STANDARD DEVIATION
Gas Vol	8	31.25	54.90	35.93	7.78
Δp	8	0.11	0.13	0.12	0.01
ΔH	8	2.86	4.08	3.27	0.44
Pbar	8	28.92	30.90	29.72	0.69
Tm	8	500.29	589.00	534.16	27.13
Ts	8	1258.90	1494.83	1363.53	83.02
Ai*	8	199.40	5659.80	2067.68	1962.69
% in Ash	8	0.29	7.93	2.33	2.59
% in Gas	8	63.60	103.77	87.02	12.47
%Isokinetic	8	106.75	148.85	121.37	13.04
Flow rate	8	0.51	0.65	0.59	0.04
Ash Conc	8	1.16	32.36	12.79	13.05
Gas Conc	8	467.32	16015.70	5018.43	5058.12

* Activity Incinerated in μCi

Table 2: Sulphate Data

VARIABLE	NUMBER OF CASES	MIN	MAX	MEAN	STANDARD DEVIATION
Gas Vol	3	32.51	34.63	33.62	1.07
Δp	3	0.12	0.14	0.13	0.01
ΔH	3	2.90	3.50	3.26	0.32
Pbar	3	29.80	30.22	29.99	0.21
Tm	3	539.90	551.58	545.83	5.84
Ts	3	1419.00	1493.25	1458.01	37.27
Ai*	3	556.40	645.18	608.94	43.58
% in Ash	3	2.12	8.18	4.31	3.37
% in Gas	3	26.76	38.32	32.61	5.78
%Isokinet	3	117.89	119.51	118.61	0.86
Flow rate	3	0.57	0.62	0.59	0.03
Ash Conc	3	3.69	21.89	10.26	10.11
Gas Conc	3	498.56	737.02	591.93	127.37

* Activity Incinerated in μCi

APPENDIX C
STUDENT'S T TEST RESULTS

I. Methionine Data: 8 Cases

A) Paired Samples t-test on :%Gas Vs% Ash

1)Mean difference = 84.699

2)Standard deviation difference = 13.149

3)T = 18.219

4)Degrees of freedom = 7

5)Prob = 0.000

B) Paired Samples t-Test On: %Gas Vs (100-%Ash)

1)Mean Difference = -10.651

2)Standard Deviation Difference = 12.311

3)T = 2.447

4)Degrees of Freedom = 7

5)PROB = .044

II Sulphate Data: 3 Cases

A) Paired Samples t-test on % Gas Vs % Ash

1)Mean Difference = 28.303

2)Standard deviation difference = 8.951

3)T = 5.477

4)Degrees of Freedom = 2

5)Prob. = .032

B) Paired Samples t-Test On %Gas Vs (100-%Ash)

1)Mean Difference = -63.083

2)Standard Deviation Difference = 3.059

3)T = 35.720

4)Degrees of Freedom = 2

5)Prob = .001

III Pooled Data : 11 Cases

A)Paired Samples t-test on: %Gas Vs %Ash

1)Mean Difference = 69.318

2)Standard Deviation Difference = 28.826

3)T = 7.975

4)Degrees of Freedom = 10

5)Prob. =0.000

B)Independent Samples t-Test On: %Ash

GROUP	N	MEAN	SD
m	8	2.325	2.599
s	3	4.307	3.364

Where m=methionine and s=sulphate

1)Separate Variances

a)T = 922

b)DF = 3.0

c)Prob = .454

2)Pooled Variances

a)T = 1.050

b)DF = 9

c)Prob = .321

C)Independent Samples t-Test On: %gas

GROUP	N	MEAN	SD
m	8	87.024	12.469
s	3	32.610	5.781

1)Separate Variances

a)T = 9.841

b)DF = 8.1

c)Prob. =0.000

2)Pooled Variances

a)T = 7.095

b)DF = 9

c)Prob =0.000

APPENDIX D
Stack Sampling Computer Program

```
$ type s-35.bas
10 REM
20 REM      PROGRAM TO PERFORM BURN CALCULATIONS
30 REM
40 REM          PROGRAM INCLUDES THE INPUT OF
50 REM          NINETEEN VARIABLES AND OUTPUTS
60 REM          DATA TO THE FILE BURNDATA.DAT
70 REM
72 OPEN "BURNDATA.DAT" FOR OUTPUT AS FILE #1
80 PRINT "THIS PROGRAM PERFORMS THE CALCULATIONS FOR SOME"
90 PRINT "STACK SAMPLING."
95 PRINT
100 PRINT " YOU WILL BE PROMPTED TO ENTER YOUR DATA AND UPON"
110 PRINT " COMPLETION OF THE INPUT, YOU WILL HAVE A CHANCE TO"
120 PRINT " LIST THE DATA AND CHANGE ANY MISTAKES THAT MIGHT"
130 PRINT " EXIST."
140 PRINT
150 PRINT
160 REM   REQUEST FOR INPUT FOLLOWS
162 PRINT "PLEASE INPUT VALUES FOR THE FOLLOWING"
164 PRINT
170 INPUT "DRY GAS VOLUME ? VM = "; VM
180 INPUT "BAROMETRIC PRESSURE ? PBAR ="; PBAR
190 INPUT "MEAN PRESSURE DROP ? H = ";H
200 INPUT "DRY GAS METER TEMPERATURE IN DEG. R ? TM = ";TM
210 INPUT "VOLUME OF WATER CONDENSED IN ML ? VWC(STD) = ";VWC
220 INPUT "VOLUME OF WATER DESSICATED IN ML ? VWD(STD) = ";VWD
230 INPUT "PERCENT OF CO2,O2,CO,N2 (SEPARATE BY COMMAS) - ";CO2,O2,CC
240 INPUT "ABSOLUTE AVG TEMP OF STACK GAS,.DEG. R ? TS = ";TS
250 INPUT "AVERAGE VELOCITY PRESSURE,P ? VP = ";VP
260 INPUT "TOTAL VOL. OF LIQUID COLLECTED IN IMPINGERS ? V1C = ";V1C
270 INPUT "TOTAL SAMPLING TIME IN MIN. ? O1 = ";O1
280 INPUT "MASS OF PARTICULATES COLLECTED, IN G. ? MN = ";MN
290 INPUT "ACTIVITY COLLECTED IN TRAPS IN MICRO CI ? AC = ";AC
300 INPUT "MASS OF ASH COLLECTED ? MASS = ";MASS
310 INPUT "DISINTEGRATIONS PER MINUTE IN ASH ? DPM = ";DPM
320 INPUT "ACTIVITY INCINERATED IN MICRO CI ? AI = ";AI
330 REM
340 REM     OPTION TO LIST THE DATA
350 REM
360 PRINT
370 INPUT "WOULD YOU LIKE TO LIST THE DATA?  Y OR N"; A$
380 IF A$ = "Y" THEN GOTO 390 ELSE GOTO 600
390 PRINT "VM = ";VM
400 PRINT "PBAR = ";PBAR
410 PRINT "H = ";H
420 PRINT "TM = ";TM
430 PRINT "VWC = ";VWC
440 PRINT "VWD = ";VWD
450 PRINT "CO2 = ";CO2
```

```
460 PRINT "O2 = ";O2
470 PRINT "CO = ";CO
480 PRINT "N2 = ";N2
490 PRINT "TS = ";TS
500 PRINT "VP = ";VP
510 PRINT "V1C = ";V1C
520 PRINT "O1 = ";O1
530 PRINT "MN = ";MN
540 PRINT "AC = ";AC
550 PRINT "MASS = ";MASS
560 PRINT "DPM = ";DPM
570 PRINT "AI = ";AI
580 REM
590 REM      CHANGE VALUES IF DESIRED
592 REM
600 PRINT
610 INPUT " ARE THESE VALUES CORRECT?  Y OR N";B$
620 IF B$ = "Y" THEN GOTO 646 ELSE GOTO 160
630 REM
640 REM     BEGIN CALCULATIONS
645 REM
646 LET VP = VP/2.54
647 LET TS = TS * .8915
650 LET H = H/2.54
660 LET VM = VM/2.83E-2
670 LET VMSTD = (VM * 528 * (PBAR + (H/13.6)))/(TM * 29.92)
680 LET VWC = VWC * .04715
690 LET VWD = VWD * .04715
700 LET BWS = (VWC+VWD) / (VWC+VWD+VMSTD)
800 LET MD = .44*CO2 + .32*O2 + .28 * (CO+N2)
810 LET MS = MD* (1-BWS) + 18* BWS
820 LET VS = .7473 * 85.49 *.85 * SQR((TS*VP)/(PBAR*MS))
830 LET QSTD =(3600*(1-BWS) * VS *2.906 *528 * PBAR)/ ( TS * 29.92)
840 LET EA = ((O2-.5*CO)/((.264*N2) - O2+(.5*CO))) * 100
850 LET INUM = (TS*.002669*V1C)+(VM/TM)*(PBAR + (H/13.6))
860 LET IDEN = 60 *O1*.000716*VS*29.92
870 LET I = (INUM/IDEN) * 100
880 let CS12 = 12*MN/(VMSTD*CO2)
890 let VQ = QSTD * ( O1/60)
900 let AM = AC*VQ/VMSTD
910 LET UC1 = (DPM/2.22E+12) *1E+6
920 LET AA = UC1 * MASS
930 LET REG = AM/AI *100
940 LET REA= AA/AI *100
950 LET RET =((AM + AA)/AI)* 100
1000 REM
1010 REM          PRINT SOLUTIONS TO OUTPUT FILE
1020 REM
```

```
1200 PRINT #1
1210 PRINT #1, " DATA FOR BURN CALCULATIONS"
1220 PRINT #1
1390 PRINT #1, "VM = ";VM
1400 PRINT #1,"PBAR = ";PBAR
1410 PRINT #1,"H = ";H
1420 PRINT #1,"TM = ";TM
1430 PRINT #1,"VWC = ";VWC
1440 PRINT #1,"VWD = ";VWD
1450 PRINT #1,"CO2 = ";CO2
1460 PRINT #1,"O2 = ";O2
1470 PRINT #1,"CO = ";CO
1480 PRINT #1,"N2 = ";N2
1490 PRINT #1,"TS = ";TS
1500 PRINT #1,"VP = ";VP
1510 PRINT #1,"V1C = ";V1C
1520 PRINT #1,"O1 = ";O1
1530 PRINT #1,"MN = ";MN
1540 PRINT #1,"AC = ";AC
1550 PRINT #1,"MASS = ";MASS
1560 PRINT #1,"DPM = ";DPM
1570 PRINT #1,"AI = ";AI
1580 PRINT #1
1585 PRINT #1
1590 PRINT #1, " OUTPUT OF SOLUTIONS TO BURN"
1600 PRINT #1
1610 PRINT #1
1620 PRINT #1, "DRY GAS VOLUME STD = "; VMSTD
1630 PRINT #1, "PROPRTION OF WATER VAPOR IN STG = ";BWS
1640 PRINT #1, "DRY GAS MOLECULAR WEIGHT = ";MD
1650 PRINT #1, "WET MOLECULAR WEIGHT OF STACK GAS =";MS
1655 PRINT #1, "AVERAGE STACK GAS VELOCITY =. ";VS
1670 PRINT #1, "DRY GAS VOLUMETRIC FLOW RATE = ";QSTD
1680 PRINT #1, "PERCENT EXCESS AIR = ";EA
1690 PRINT #1, "PERCENT ISOKINETIC VARIATION = ";I
1700 PRINT #1, "PARTICULATE EMISSION CONCENTRATION = ";CS12
1800 PRINT #1, "DRY STACK GAS VOLUME = ";VQ
1810 PRINT #1, "TOTAL MONITORED ACTIVITY = ";AM
1820 PRINT #1, "ACTIVITY RECOVERED IN ASH = ";AA
1830 PRINT #1
1840 PRINT #1
1850 PRINT #1, "RECOVERY EFFICIENCY"
1860 PRINT #1, "EFFLUENT COLLECTION = ";REG
1870 PRINT #1, "ASH COLLECTION = ";REA
1880 PRINT #1, "TOTAL RECOVERY EFFICIENCY = ";RET
1890 CLOSE #1
1892 PRINT
1894 PRINT "THE DATA HAS BEEN CALCULATED AND YOU WILL"
1896 PRINT " FIND THE SOLUTIONS IN A FILE CALLED BURNDATA.DAT"
1898 PRINT "  IN YOUR DEFAULT DIRECTORY."
1900 END
```

APPENDIX E

Preparation of Trapping Cocktail

1.	180 ml	2Phenylethylamine
2.	250 ml	Methanol
3.	570 ml	Toluene
4.	5 g	PPO (2,5-Diphenenyloxazole)
5.	0.5 g	Dimethyl-POPOP (1,4-bis-2-(4-Methyl-5-phyenyloazolyl)-benzene)

APPENDIX F

^{35}S Quench Curve