GAS QUALITY ANALYSIS AND EVALUATION PROGRAM FOR PROJECT GASBUGGY*

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ABSTRACT

Experimental results of the gas quality analysis program for Project Gasbuggy through August 1969 are presented graphically, addressing the questions raised by the preshot program goals. The chemical composition and the concentrations of tritium, krypton-85, carbon-14 and argon-37, 39 are presented as a function of time and gas production from the nuclear chimney. Chemically, the presence of CO_2, CO and H_2 served to dilute the formation gas and caused reactions which significantly altered the gas composition at early times. The radionuclide content of the chimney gas at reentry was some 800 pCi/cm^3 of which about 80% was CH_3T. Lesser quantities of tritium were observed as HT, C_2H_5T and C_3H_7T. The other major contaminant was Kr^{85} which was present at about one-fifth the level of CH_3T. Small quantities of carbon-14 and argon-39 were also identified. The only other radionuclides identified in the gas were relatively short-lived rare gases.

During the production testing, about two and one-half chimney volumes of gas at formation pressure were removed. This removal, accompanied by dilution, has reduced the radionuclide concentrations to about 7% of their levels at reentry. The production characteristics of the Gasbuggy environment prevented an adequate test of the effectiveness of chimney flushing. However, the rapid drawdown concept is supported by the available data as an effective means of reducing contaminant levels. The changes in composition during production or testing are seen to be consistent with a model involving a non-uniform gas influx rate and flow distribution over the chimney region. Mixing times are estimated to be on the order of a few days, so that increasing concentrations following a sudden gas influx can be explained.

I. INTRODUCTION

Since inception, plans for Project Gasbuggy have included a program of one sort or another for analysis of the gas in the postshot nuclear chimney. The first formalization of this program was made by the Gasbuggy Feasibility Study[1] issued by El Paso Natural Gas Company in May of 1965. Among the experimental objectives listed in that study were the measurement of (1) the "extent of radioactive contamination of the produced gas," and (2) "extent of mixing of formation gas with contaminated chimney gas, and investigation of production techniques for controlling the degree of mixing." More recently, these objectives have been re-stated as a need "to determine the gas quality

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with regard to contamination by radioactivity and to evaluate various techniques suggested for reducing this contamination.[1,2,3]

In this paper, the accumulated data of the last two years will be discussed in relation to these specific program objectives. Interpretation and evaluation of the Gasbuggy results in more general terms, related to the expanded goals of the Gas Quality program, are discussed in subsequent papers.[4,5]

II. EXPERIMENTAL PROCEDURES

The physical operations involved in the procurement of data are sampling, chemical analysis, radiochemical analysis, and monitoring. Each operation depends to some extent upon the others, and each must be optimized to ensure reliable experimental results. Conversely, appreciation of the significance of these results is dependent upon an appreciation of these operations and their associated limitations and uncertainties. No useful purpose is served here by a detailed discussion of these procedures; they are so discussed elsewhere.[6,7] I shall, however, attempt to summarize some of the significant aspects which are necessary to the discussions which follow. An overview of the gas production and our analytical results is shown in Fig. 1.

Sampling

Because of the potential problems associated with early release of chimney gas, initial samples of the gas were obtained using a downhole sampling system. These problems did not materialize, and surface snap sampling, following sufficient production to flush the production tubing, was attempted shortly after chimney reentry was completed in January 1968. No significant differences in quality of samples obtained by these two methods were seen; therefore, all samples obtained since that time have been taken by the surface snap method.

Eighteen samples were obtained prior to the start of the first production test in June 1968, constituting four independent sets of replicate samples. During short-term production testing, we have attempted to define each test period with at least three samples (start, middle, and end). Bi-weekly samples were taken during the long-term production test beginning in February of 1969. Analyses of these selected samples were used to anchor the day-by-day trends observed by the monitoring system.

The data included in this paper have been obtained through analysis of 57 of the 75 samples obtained for LRL through August 1969.[8] Eighteen samples have been preserved for historical record.

Chemical Analysis

The chemical composition of Gasbuggy samples was determined by mass spectrometry, as was the purity of the separated components. Because of the implementation of routine snap sampling, a small quantity of air is collected along with the chimney gas. The quantity of air is estimated from the oxygen concentration, and is subtracted to obtain a true sample volume. Chemical compositions reported are normalized to 100% exclusive of air. Routinely, this correction is of the order of 1% or less, but occasional samples contained considerably more air. In the extreme, the first four samples obtained from the sealed annulus atop the emplacement well at 1 day following detonation were about 80% air.

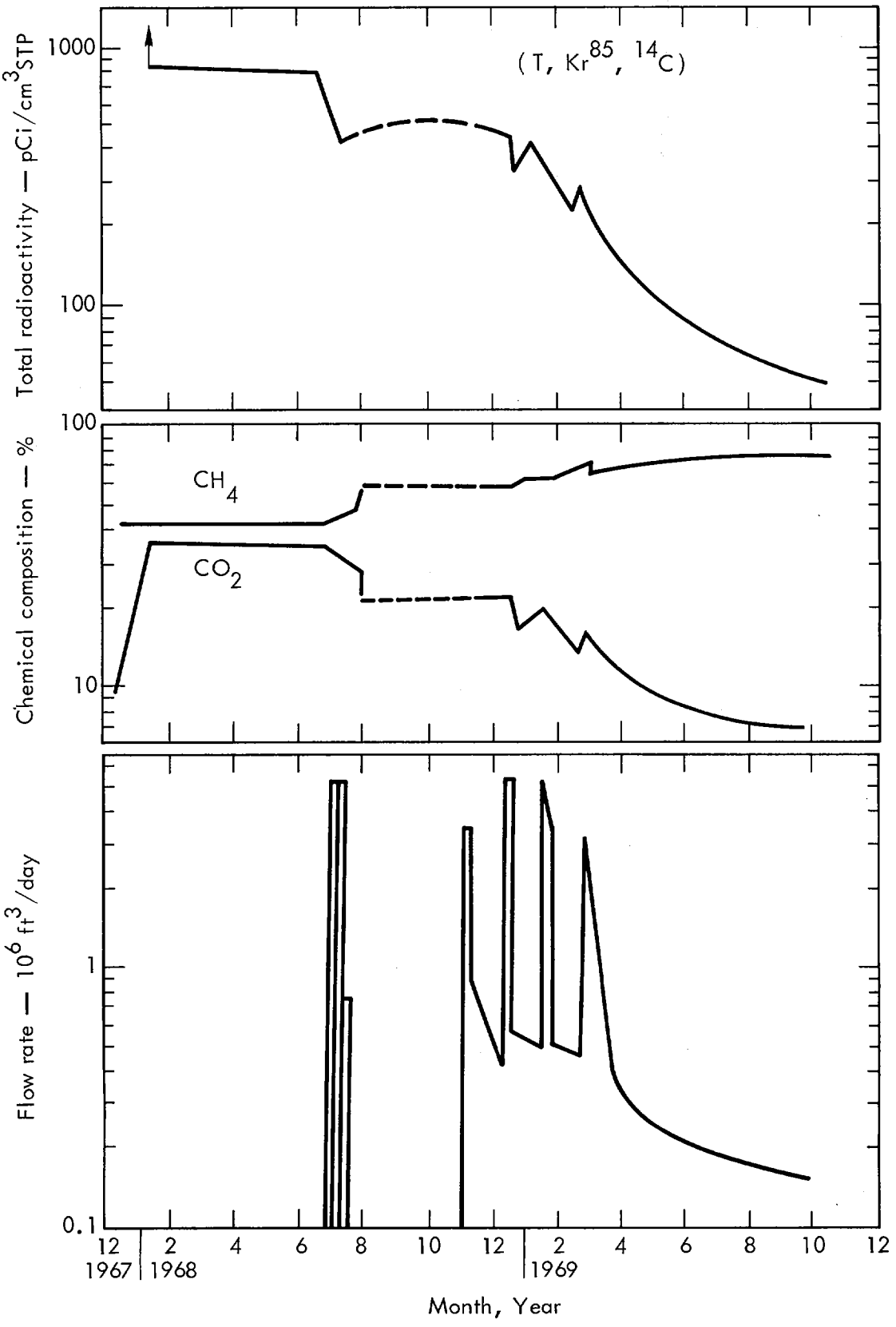

Fig. 1. Project Gasbuggy Gas Quality analysis results.

Radiochemical Analysis

Radiochemical analysis of a Gasbuggy sample begins with separation and purification of the desired components by elution chromatography. The purified fractions are then placed in appropriate counters for radio assay. Krypton-85 is determined in quadruplicate by thin-window beta proportional counting. Compounds containing tritium and C^{14} are determined by internal proportional counting of duplicate or quadruplicate fractions.

For intercomparison of the radionuclide concentrations, all data have been corrected for decay to the time of detonation.

Monitoring

The Stallkat system for monitoring the concentration of Kr^{85} and tritium in the gas flowing from the Gasbuggy nuclear chimney has been in operation since the onset of production testing in June 1968. The instrument was conceived, and has been most valuable, for continuous monitoring in connection with the on-site safety program. Reports of the Eberline Instrument Corporation should be consulted in this regard.[9] The principal value of the Stallkat system to the Gas Quality Analysis program has been to delineate trends in the concentrations of Kr^{85} on a day-to-day basis. The ability to normalize these trends to our laboratory results has been a significant factor in reducing the number of analyses required for adequate documentation and investigation of phenomenology.

III. LIMITATIONS AND UNCERTAINTIES OF THE ANALYTICAL RESULTS

The absolute uncertainty associated with determination of chemical composition by mass spectrometry is related to the actual concentration, but for those species of interest here can be assumed to be on the order of ±5% or less. Precision of the radiochemical determinations is measured by replicate analyses, the uncertainty being generally on the order of a few percent. The absolute uncertainty of these measurements is not known, but is probably less than ±10% for the species listed.

The data summary graphs which are to be presented here are based upon information published in tabular form for the open file system of Project Gasbuggy.[8] This tabulation includes actual values for the precision of radiochemical determinations for each sample. Error bars have, in general, been omitted from the graphical presentation because they typically fall within the plotted point symbols. Where such bars do appear, they represent the spread observed in results obtained from analyses of two or more presumably similar samples, and do not reflect the precision of a determination for an individual sample or component.

Potentially, large uncertainties may be present due to the very nature of the experiment. Among these are the single sampling point at the top of the nuclear chimney, non-ideal gas influx and mixing, and an extremely complex environment in which to study chemical and radiochemical reactions. It is extremely difficult to separate those phenomena associated with peculiarities of the Gasbuggy environment from observations characteristic of gas stimulation by nuclear means in the general case. The need for further detailed experimentation, both in the laboratory and in the field, is particularly evident in this area.

IV. RESULTS AND DISCUSSION

For the present discussion, results from Project Gasbuggy may be grouped according to major gas production periods. The initial 200-day

shut-in period has provided significant chemical and radiochemical data. A high flow-rate test followed shut-in, and is a partial test of the concept of chimney flushing. Two periods of production testing, the first composed of three test segments and the second a long-term production test, are useful in defining the relationship between flow rate and concentration of chimney gas components.

Chimney Shut-In Period (12/10/67 to 6/28/68)

During the first 200 days following detonation, the Gasbuggy chimney served as a great reaction vessel in which essentially all important chemical and radiochemical reactions observed to-date took place. Considerable interest has, therefore, been accorded to the information gained from this period.[4,5,6]

There are only five sets of samples taken during this period, including that obtained at 1 day via the cable leak, and the initial (after 5 million ft^3 of gas had been produced) sample of the June/July production test. Although the method of obtaining the 1-day sample was far from ideal and considerable uncertainty regarding its true worth persists, we feel that it merits consideration relative to establishment of trends in the chimney gas composition prior to the reentry samples (at 34 days). However, a detailed description of the actual changes in composition is not known. Arbitrary dashed lines are used to connect points, without intent to define the true rate of composition change.

Chemical composition of the chimney gas during this period is presented in Fig. 2. Immediately apparent is the fact that the most significant changes in composition occurred during the first month. The lack of reliable experimental data during this period may preclude an unambiguous interpretation of these results. Quite evident, however, is the early, rapid conversion of CO to CO_2, probably by a reaction with steam. This reaction appears to have reached equilibrium prior to the 1-month datum point. The observed decrease in H_2 concentration is slight, possibly due to a combination of its production in the aforementioned reaction and its reduction through various methane-producing reactions involving CO and CO_2. Methane and ethane as components of both the chimney gas and the formation gas are less easily interpretable. The illustrated trend probably is misleading, and methane was essentially constant. Within the uncertainties involved with the exceptionally large air correction for these early samples, it is quite possible to assume that the observed behavior of both methane and ethane can be accounted for by influx of natural gas from the formation as a result of cooling of the chimney.

Following chimney reentry, only H_2 and CO exhibit significant changes in abundance. The decreases observed are consistent with the assumption that they react to produce methane and CO_2 at a relatively slow rate.[5] Additional formation gas influx is expected as chimney cooling occurs, and indeed must be assumed to explain the extent of the increase in the percentages of CH_4 and C_2H_6. The observed decrease in C_3H_8 is probably due to fractionation of the sample in transit through the production tubing.

The concentrations of Kr^{85}, and of tritium in various chemical forms are presented in Fig. 3. Again, the most significant changes are observed during the first month. The most dramatic of which is exhibited by HT which decreases by a factor of about 20. This tritium is partially incorporated in methane and ethane, but is primarily removed from the gas presumably by incorporation in water, via exchange reactions.[4] Most probably these reactions have reached equilibrium prior to chimney reentry. The concentration of C_3H_7T is essentially constant, although the data are influenced by the same fractionation effects noted previously.

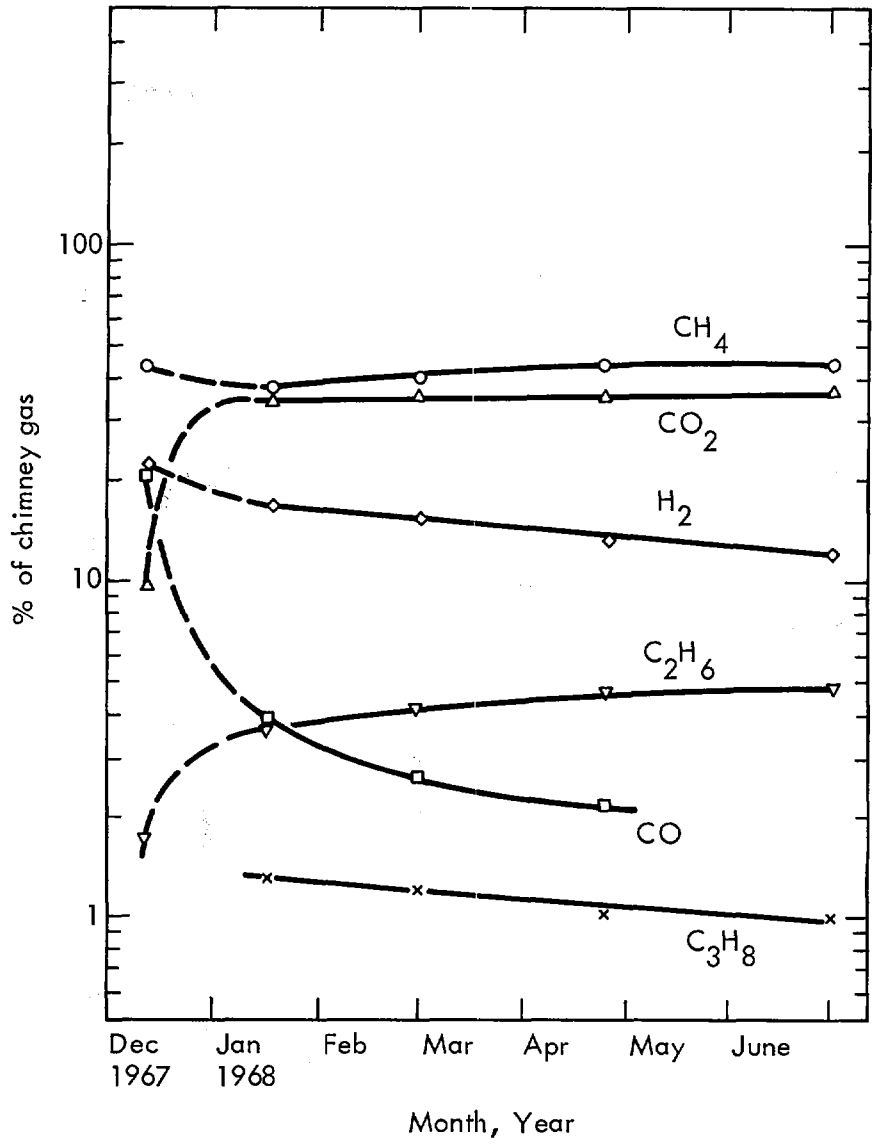

Fig. 2. Initial shut-in period, chemical composition.

The total tritium content of the gas decreased during the first month from about 30% to about 5% of the expected total of 4 g of this isotope. Presumably, the remaining 95% is in the form of water. The principal contaminant of the gas after the first month is seen to be CH_3T.

The near constancy, within experimental uncertainty, of the Kr^{85} concentration over the entire shut-in period implies mixing with a constant volume of gas and rapid pressurization of the chimney. Essentially all the mass-85 fission yield is Kr^{85} by 10 minutes following the detonation. Although entrapment of the gas in solidified melt could occur, the measured total Kr^{85}, using a known amount of Xe^{127} tracer added to the device preshot, was 350 ± 20 Ci, which corresponds to the preshot estimate. The actual volume of gas with which the Xe^{127} must have mixed to give the observed concentrations was $3.0 \pm 0.2 \times 10^9$ liters at standard temperature and pressure ($1.06 \pm 0.05 \times 10^8$ ft^3 STP). This quantity of gas would be contained in a void

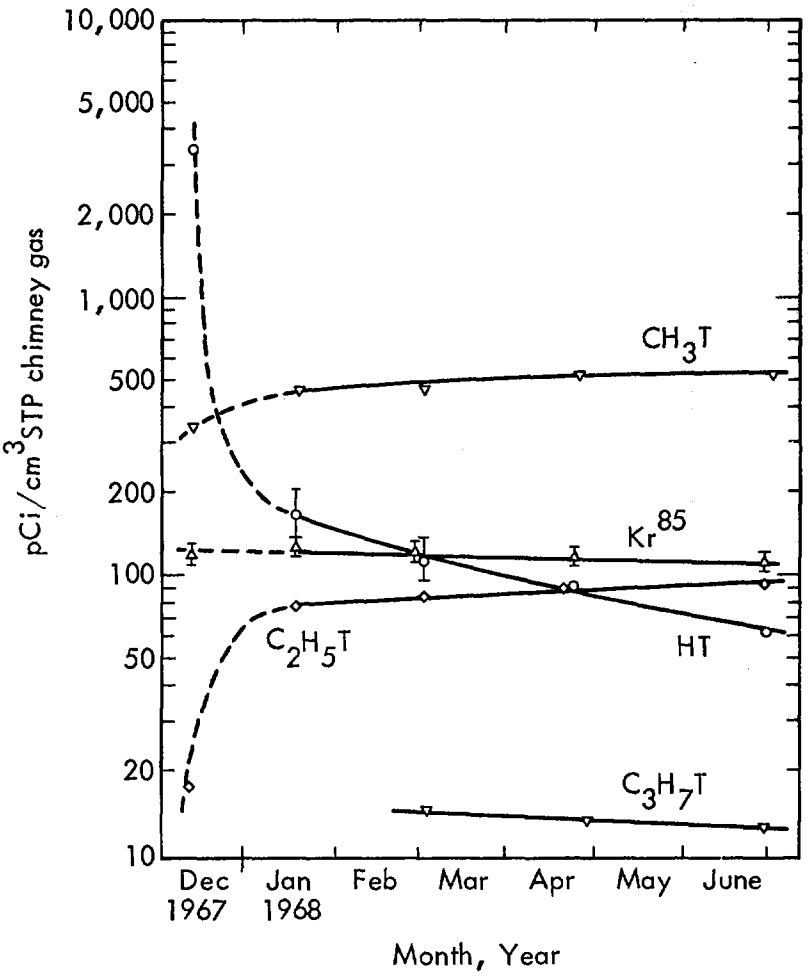

Fig. 3. Initial shut-in period, radionuclide concentrations.

of $5.8 \pm 0.3 \times 10^7$ liters ($2.0 \pm 0.1 \times 10^6$ ft^3) at 150°F and 950 psig (the observed condition on Jan. 23, 1968). Such a void was estimated preshot, and is in reasonable agreement with volumes derived during chimney drawdowns.

Experimental concentrations of C^{14} in the principal carbon-containing gases are presented in Fig. 4. Clearly apparent is the decreasing $C^{14}O$ and correspondingly increasing $C^{14}H_4$. These data support the observation that methane is being produced. Total C^{14} in the gas is essentially constant at about 2.5 pCi/cm^3, implying a total C^{14} content in the gas of about 7.5 Ci. No indication of the actual total C^{14} produced is available, since some unknown fraction of this isotope may exist in one or more non-gaseous forms in the nuclear chimney.

Other radionuclides have been identified in the chimney gas during this period. During the first few months, appreciable quantities of short-lived

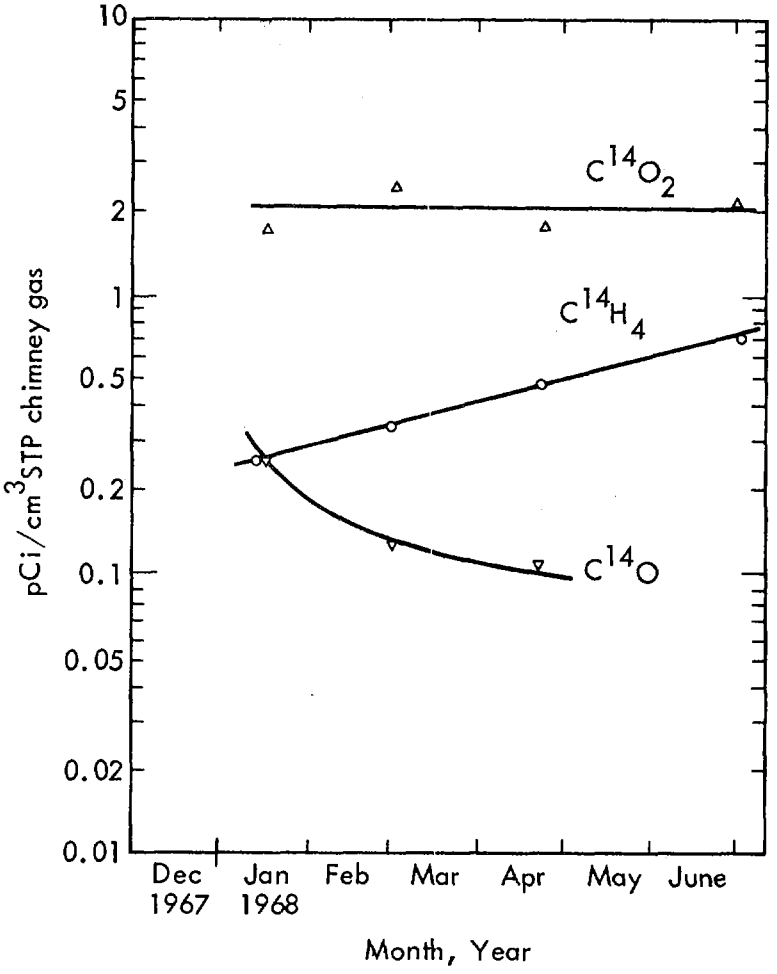

Fig. 4. Initial shut-in period, radionuclide concentrations.

fission product gases and activation products, principally Ar^{37} and Ar^{39}, were present. By the end of the 6-mo period, essentially all of the fission product gases except Kr^{85} had decayed. Concentrations of Ar^{37} were comparable to those of Kr^{85}, while the Ar^{39} was some 1000 times less abundant. Aside from Rn^{222}, which is normally present in natural gas, no other radionuclides have been detected. A search for the possible presence of non-gaseous isotopes was made during this period by gamma spectroscopy.[10] No such species was identified, and maximum possible concentrations based upon detection limits were established. These range from 0.04 pCi/cm^3 for I^{131} to about 10^{-5} pCi/cm^3 for Cs^{137}. More recently, a beta-count survey of filters taken during a rapid drawndown in November 1969 was made. Total beta activity on these particulate samples is estimated to be less than or equal to 10^{-7} pCi/cm^3 of gas passing the filter.

In summary, the principal radioactive contaminants of the Gasbuggy chimney gas at about 6 mo following the detonation are T, Kr^{85}, Ar^{37} and C^{14}. The relative abundances are listed in Table I.

Table I. Relative contribution to total gaseous radioactivity at 6 mo.

Tritium (principally CH_3T)	78%
Kr^{85}	12.5%
Ar^{37}	9.2%
C^{14} (principally $C^{14}O_2$)	0.25%
Other	0.05%

High Flow Rate Production Tests (6/28/68 to 7/14/68)

At the end of the shut-in period, the chimney gas was flared at the rate of 5 million ft^3 per day for 6 days, shut-in for 2 days, then produced for 5 more days. The flow rate was then dropped to 3/4 million ft^3 per day for 4 days, at which time the well was closed off. The observed changes in chemical composition during this period are shown in Fig. 5. At the high flow rate, the produced gas is

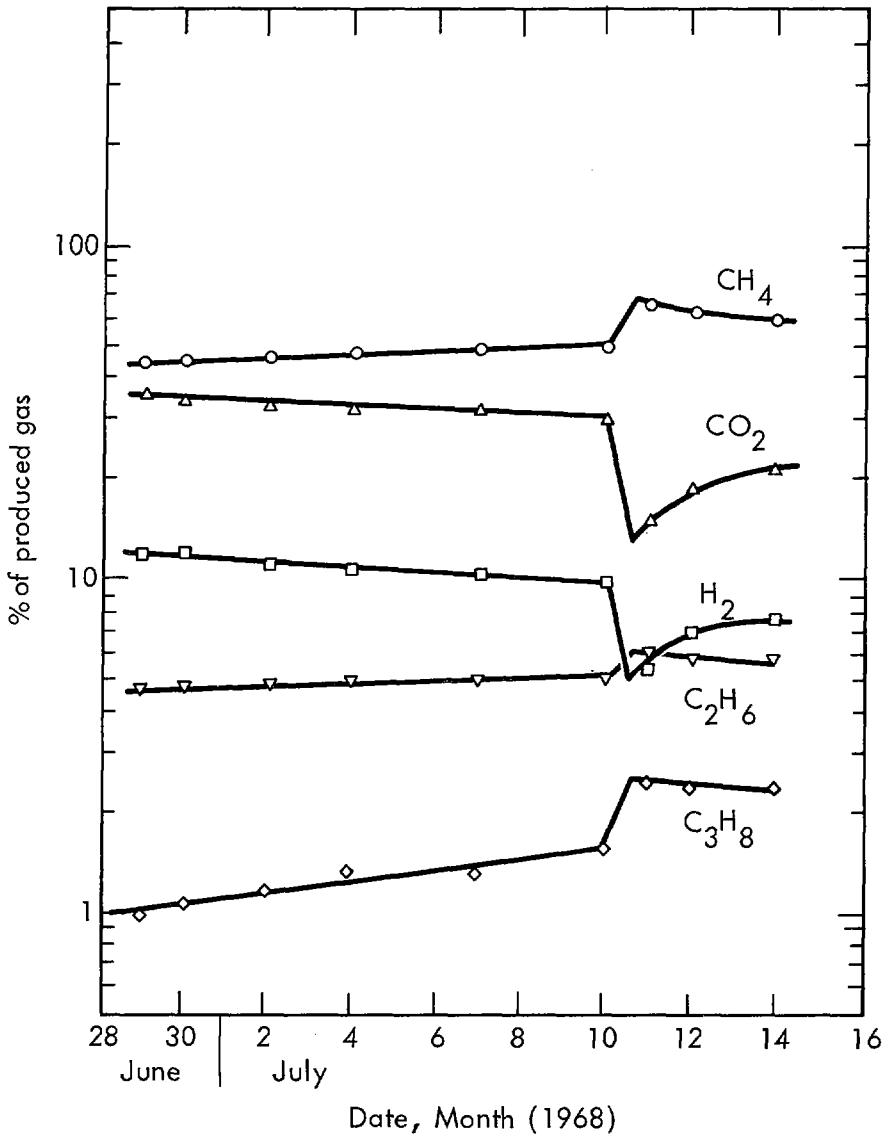

Fig. 5. High rate production test, chemical composition.

drawn principally from the chimney. Only 17% of the gas removed actually came from the formation. A significant dilution in the produced gas was observed when the flow rate was cut back on July 10. The concentrations of the various species after the 4-day lower flow period are about what would be expected by applying a dilution correction, based upon top hole pressure measurements, to the final high flow rate composition. We are forced to conclude that when the flow was reduced, the dilution was greater than could be explained by simple formation gas influx and rapid mixing. The observed behavior can be explained by a significant influx of gas into the top of the chimney at about the time the flow was reduced, causing the large dilution observed. Over the 4-day period the components of the chimney become essentially mixed, resulting in an apparent increase in concentration of CO_2 and H_2. These conclusions are supported by gas analysis data from subsequent production tests. The increased productivity of the upper portions of the Pictured Cliffs as related to deeper sections is made plausible by comparison of pre- and postshot caliper logs[11] and by production results[12] of satellite wells.

Concentration of radionuclides during this period are presented in Fig. 6. Lines of similar shape are drawn through data for each component. These are based upon the trends observed in CO_2 and H_2 in the preceeding figure. No difference in shape is warranted by the fit of these lines to the data, indicating absence of the effects of chemical reactions.

Inferences drawn from these data with regard to the effectiveness of chimney flushing suffer two main drawbacks. In the first place, the duration of the high-rate tests were not sufficient to demonstrate the effects of formation gas influx at relatively low chimney pressures. Although 40% of a chimney volume of gas was produced, more than 80% of this was initially within the chimney. The degree and rate of mixing of formation gas with chimney gas under high production rate conditions thus remains undefined.

The second drawback is due to the geology of the Gasbuggy site. Production of gas occurs principally via fractures, with the matrix permeability being quite low. Postshot investigations[11] have deduced a sketch of the chimney which reveals sagging geologic bedding planes rather than a void in the upper portion of the chimney. Since the reentry well penetrates only the uppermost layers of these planes, horizontal communication with formation gas through fractures is probable. Furthermore, access of chimney gas to this region may be impeded (permeability within the chimney is finite). We may, therefore, be observing a mixture of gases in a flow situation which is related to the true chimney gas composition via the relative values of horizontal and vertical permeabilities. In other words, at a given influx rate, the gas observed during high flow rate tests may contain a higher proportion of the true chimney gas than that observed during low production rates. Presumably, mixing occurs under static conditions so that the concentrations following shut-in are essentially uniform throughout the chimney.

Having stated these caveats, and with the limited data available, it is imprudent to speculate upon the extent to which the Gasbuggy chimney could have been decontaminated by a long-term, high-rate production experiment. However, decontamination by rapidly removing a contaminated chimney volume or two of gas appears to be effective, so long as production well in excess of gas influx is possible. The process is useful only when gas in this quantity can economically be flared, produced to controlled usage or produced to storage.

Constant Bottom Hole Pressure Production Tests — (11/7/68 to 2/18/69)

Between July 14 and November 11, 1968 the Gasbuggy chimney remained shut-in. A series of three production tests followed in which an attempt to maintain a constant bottom hole pressure by varying flow rate was

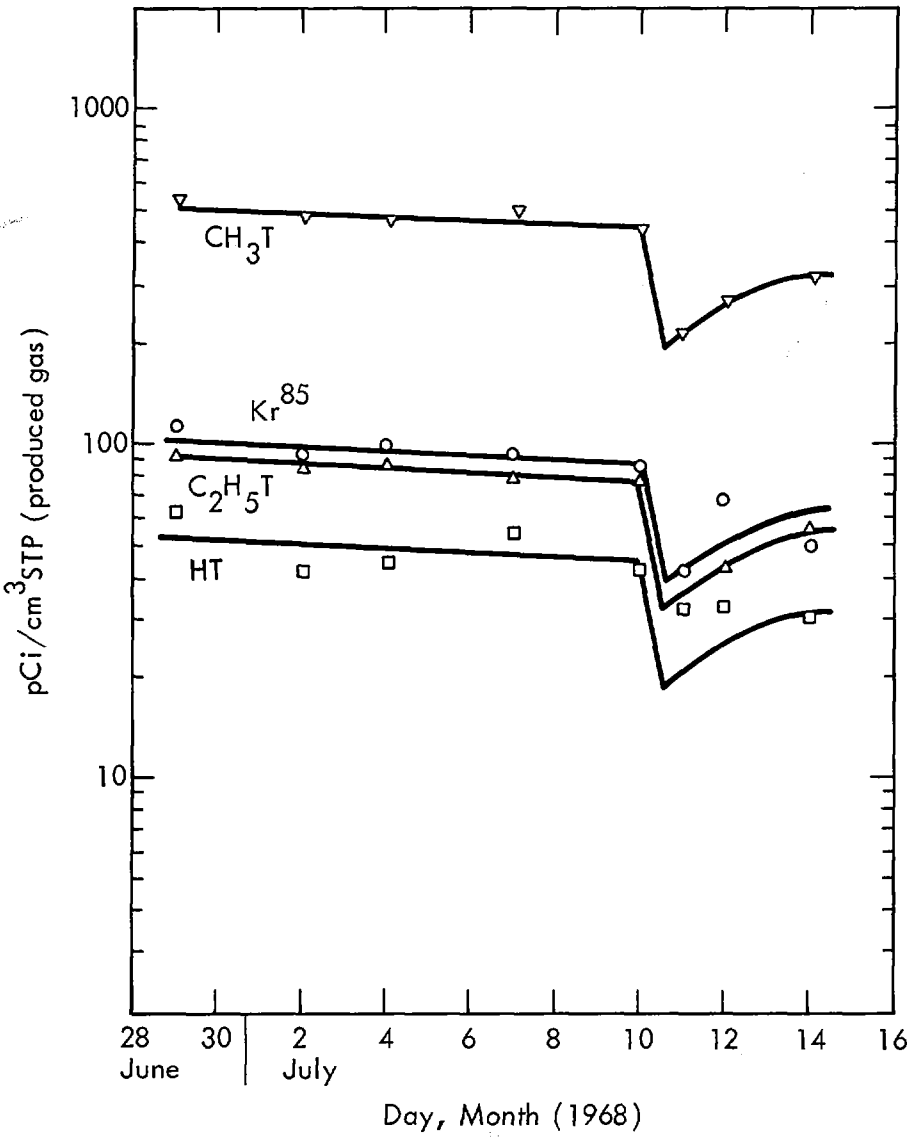

Fig. 6. High rate production test, radionuclide concentrations.

made. Each test was preceded by a rapid drawdown to adjust the initial pressure. Presented in Fig. 7 are fractional abundances of the principal components of formation gas. Aside from the dilution which occurred following the second rapid drawdown, no distinct trends are obvious. These species are, however, quite insensitive as indicators of dilution since their concentrations in the chimney are approaching those in the formation.

Much more sensitive are the components of chimney gas alone, CO_2 and H_2, as illustrated in Fig. 8. The complex behavior of these concentrations is plausibly explained by the following chain of events: During the initial test period only a slight admixture of formation gas occurred, and essentially only chimney gas was being produced. Little mixing and dilution of the chimney gas took place, as evinced by the apparent return of the concentrations to their initial levels during the rapid drawdown in December. The pressure drop resulting from this drawdown induced a rapid influx of formation gas and produced

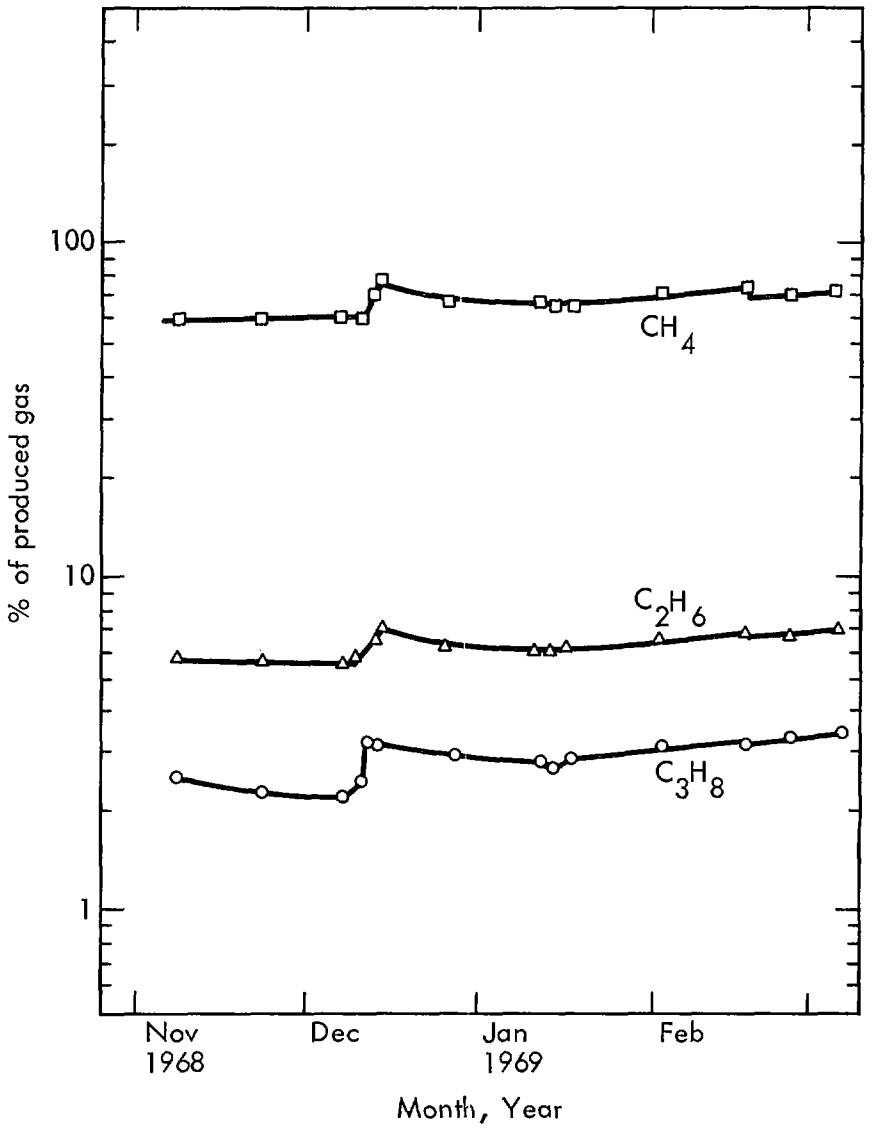

Fig. 7. Constant bottom hole pressure production tests, chemical composition.

the initial large dilution observed. Gradual mixing of this diluent with the chimney contents produced the observed increase in concentrations during the second test period. These observations are analogous to those made previously from the June/July production test data. The rapid drawdown of January 1969 was apparently not followed by a rapid gas influx from the formation. Nevertheless, the decreased pressure stimulated the flow of diluent gases through the third month of production, and concentrations of these species in the produced gas continued to decrease. Illustrated at the end of February is the effect of the high production rate which began a long-term drawdown of the Gasbuggy chimney. Again the lack of efficient mixing is demonstrated by increasing concentrations. This is due to inclusion of relatively more chimney gas in the produced mixture as a result of the high withdrawal rate.

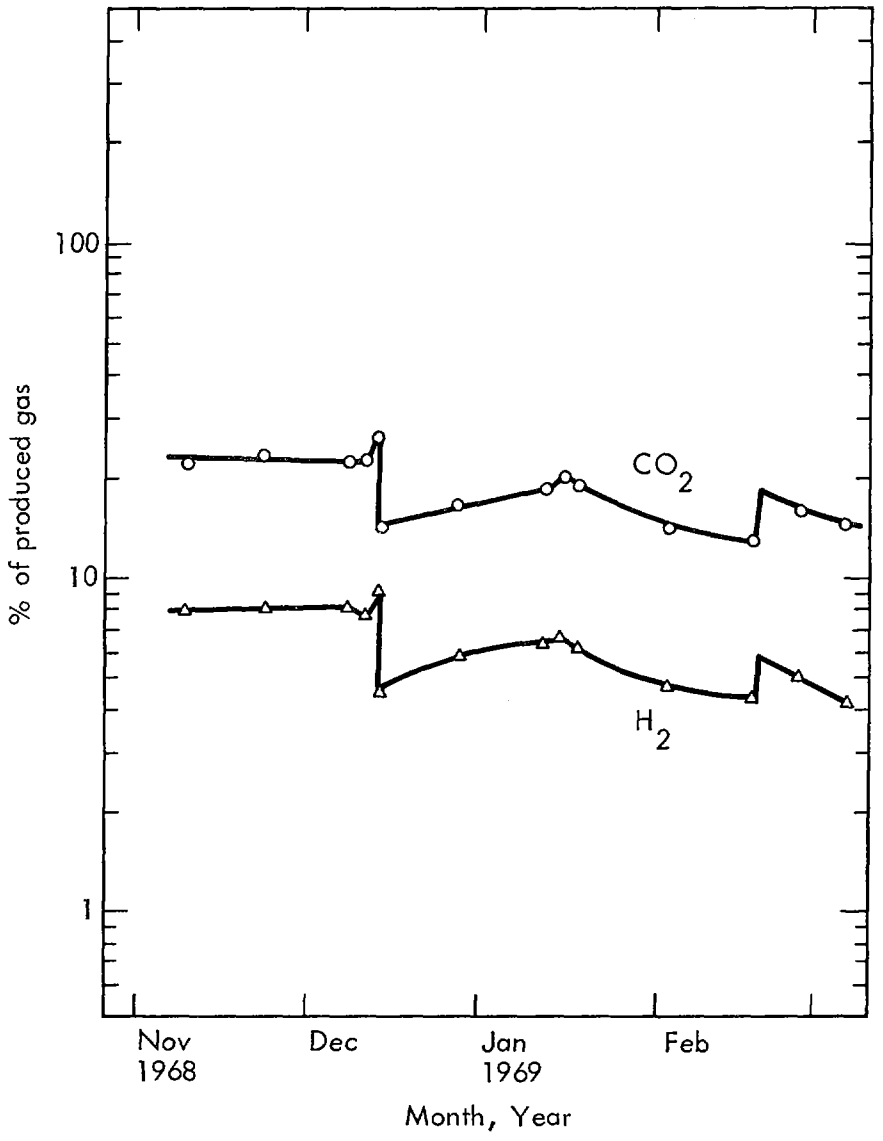

Fig. 8. Constant bottom hole pressure production tests, chemical composition.

Radionuclide concentrations through these tests are plotted in Fig. 9. As expected, the changes observed in these concentrations generally parallel those observed for CO_2 and H_2. Although some differences appear they are not described by a consistent trend, and are therefore probably not related to chemical reactions in the gas. We conclude that no significant effect other than dilution is detectable, within experimental uncertainty, during this period.

Long Term Production Test (2/18/69 to 10/28/69)

An 8-mo production test was begun in February 1969 at a flow rate of 3.5 million ft^3 per day. This production more or less gradually decreased until June where it was essentially stabilized at about 160,000 ft^3 per day. The percentage composition of components of the chimney gas through August

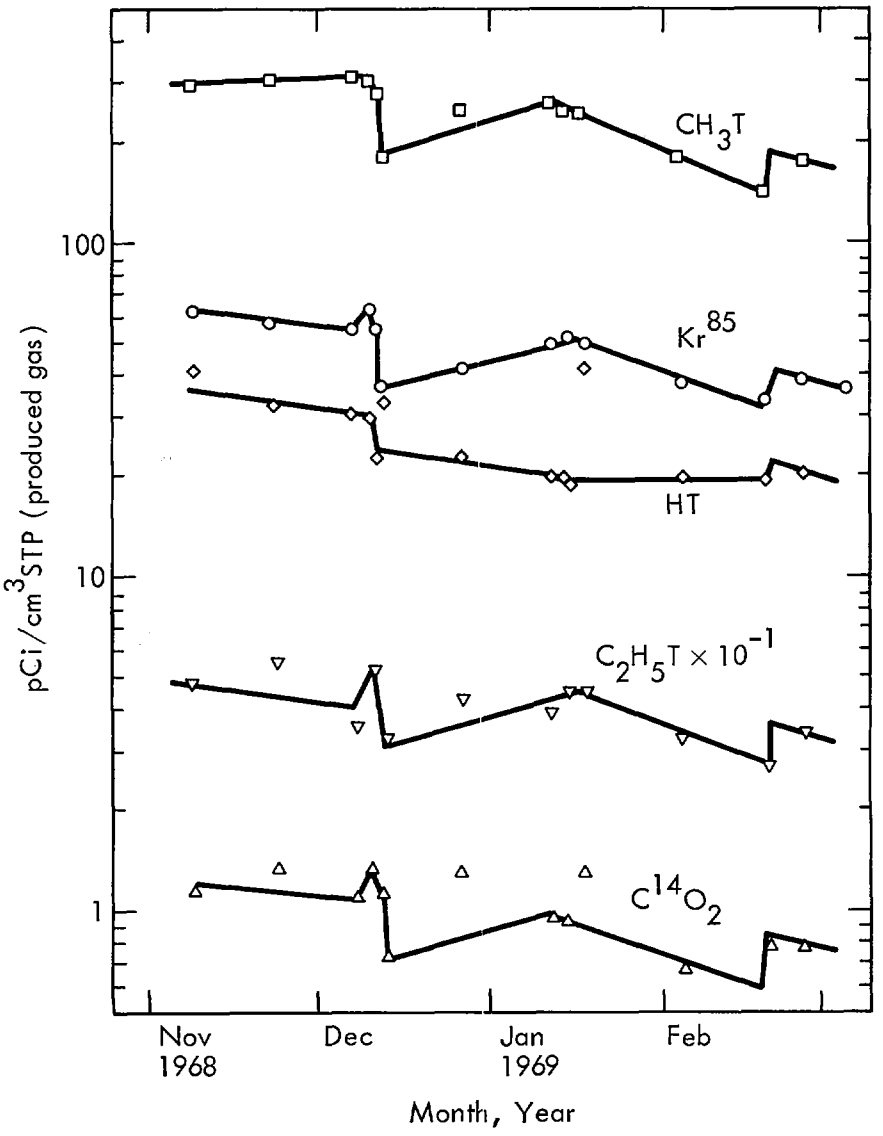

Fig. 9. Constant bottom hole pressure production tests, radionuclide concentrations.

is shown in Fig. 10. Effects of dilution are demonstrated by the gradually decreasing concentrations of CO_2 and H_2. Formation gas influx was decreasing throughout this period, as evidenced by the necessity to adjust flow rates downward to maintain a reasonably constant down-hole pressure. Concentrations of CO_2 and H_2 are seen to reflect this decreasing availability of diluent gas from the formation. Note, for future reference, that the overall decline in concentrations of CO_2 and H_2 amounts to a dilution of a little more than a factor of two, and that they appear to be changing relatively little during the final few months. The components of the formation gas exhibit increasing concentrations, as expected.

Concentration changes of the radioactive chimney gases are shown in Fig. 11 for this production period. The similarity of the rates of decrease of these concentrations is indicative of the effects of dilution with formation gas.

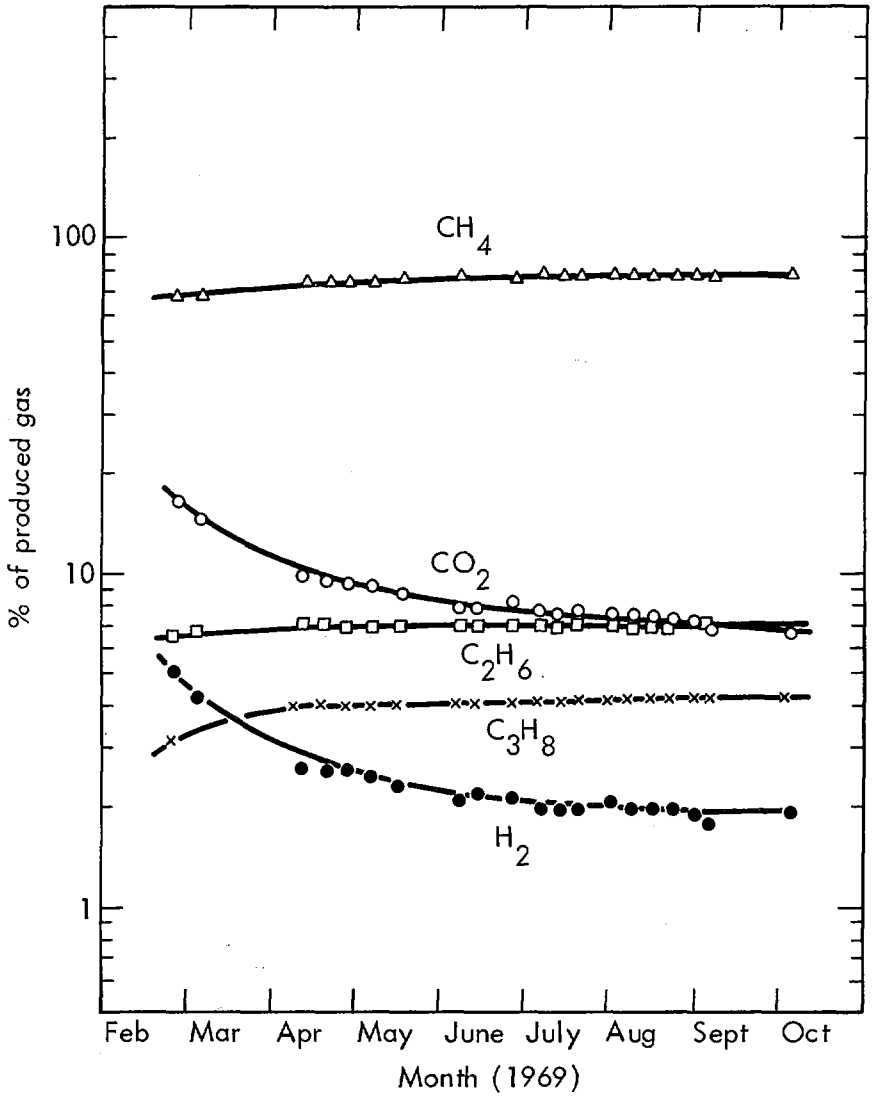

Fig. 10. Long-term production test, chemical composition.

Comparison of the shape of these curves with that shown for CO_2 and H_2 in Fig. 10 presents an interesting anomaly. Relative changes for the radioactive components amount to more than a factor of four due principally to a greater rate of decrease during the final months. To illustrate, the ratio of Kr^{85} to H_2 and to CO_2 is plotted in Fig. 12 over the entire Gasbuggy postshot period. Although the deviation is most marked toward the end of the extended production period, the trend is observed throughout. Evidently, the concentrations of CO_2 and H_2 are decreasing less rapidly than the concentration of Kr^{85} during production. This trend implies a source of CO_2 and H_2 which is not initially mixed with the Kr^{85} in the chimney gas. Such a source for CO_2 may be gas dissolved in the water within the chimney. H_2 is much less soluble and the soda water concept cannot explain its similarity to the CO_2. A possible source for additional H_2 could be diffusion into the formation at early times and subsequent diffusion back to the chimney when the pressure was significantly

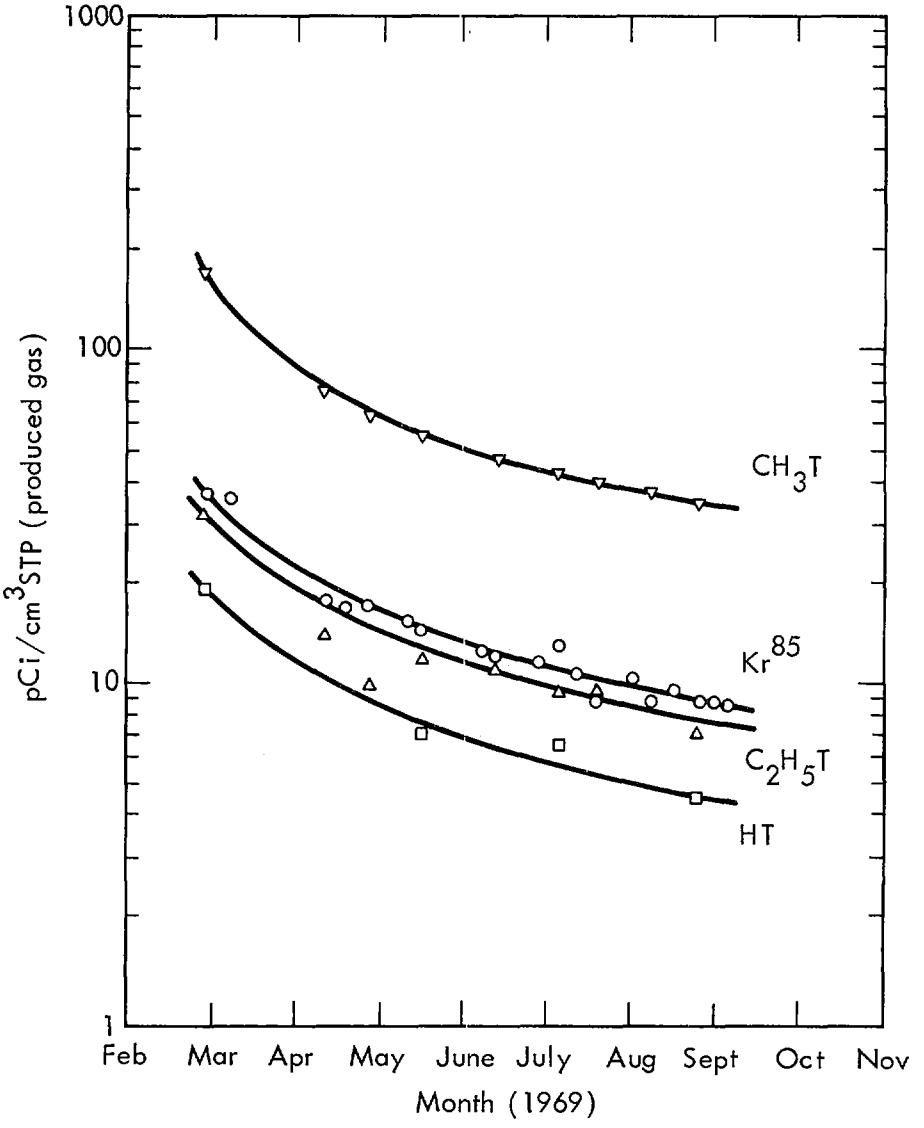

Fig. 11. Long-term production test, radionuclide concentrations.

lowered by production. All radionuclide concentrations including the Kr^{85} have been corrected for radioactive decay to the time of detonation.

V. SUMMARY

Drawing from the data presented here, it is possible to address the questions raised by the principle goals of the Gas Quality Program. The gas quality, both with regard to chemical composition and radionuclide concentrations, has been well defined. Chemically, the presence of CO_2, CO and H_2 served to dilute the formation gas and participated in reactions which significantly altered the gas composition at early times. The radionuclide content of the chimney gas at reentry was some 800 pCi/cm^3 of which about 80% was CH_3T. Lesser quantities of tritium were observed as HT, C_2H_5T and C_3H_8T.

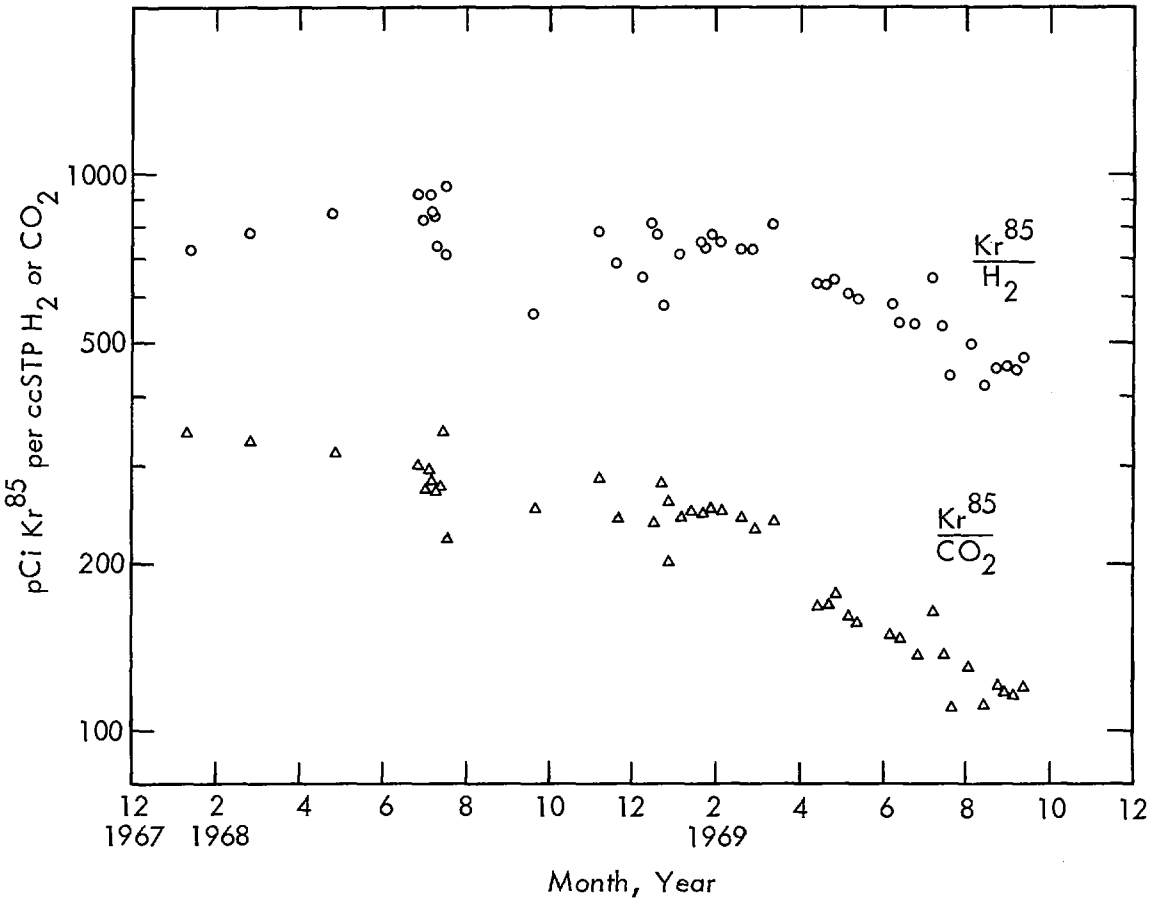

Fig. 12. Ratios of Kr^{85} to H_2 and CO_2.

The other major contaminant was Kr^{85} which was present at about one-fifth the level of CH_3T. Small quantities of C^{14} and Ar^{39} were also identified. The only other radionuclides identified in the gas were relatively short-lived rare gases.

During the production testing, about two and one-half chimney volumes of gas at formation pressure were removed. This removal, accompanied by dilution, has reduced the radionuclide concentrations to about 7% of their levels at reentry. The production characteristics of the Gasbuggy environment prevented an adequate test of the effectiveness of chimney flushing. However, the rapid drawdown concept is supported by the available data as an effective means of reducing contaminant levels. The changes in composition during production testing are seen to be consistent with a model involving a non-uniform gas influx rate and flow distribution over the chimney region.

Mixing times are estimated to be on the order of a few days, so that increasing concentrations following a sudden gas influx can be explained.

Much of the information gathered from this work, especially the gas production effects, may be unique to the Gasbuggy environment. Considerably more information must be available from experiments, both laboratory and in the field, before these data can be generalized to the benefit of nuclear stimulation. The great volume of data which have been summarized here have not yet been fully evaluated and correlated with information gained in other Gasbuggy programs. The interpretative efforts of the Gas Quality program are, therefore, continuing in an attempt to gain a better understanding of the phenomena involved.

ACKNOWLEDGMENTS

The Gas Quality Analysis and Evaluation Program for Project Gasbuggy has drawn upon the talents of a large number of people to produce the results tabulated here. In particular, the efforts of the following groups and individuals deserve recognition:

Mass spectrometry for both chemical analysis of samples and chemical purity of separated fractions was performed by Richard W. Crawford and the Analytical Chemistry Section of the LRL General Chemistry Division.

Sample procurement was a combined effort of many LRL Chemical Engineering, LRL Hazard Control, and Eberline Instrument Corporation personnel.

Radiochemistry was performed by the Gas Analysis Group of the LRL Radiochemistry Division under the immediate supervision of Dr. Floyd F. Momyer.

In addition, the continued interest of the project participants, El Paso Natural Gas, U.S. Bureau of Mines, and the Atomic Energy Commission, as well as valuable discussions with their representatives and with colleagues at LRL has been of great help to the continuing effort to obtain and evaluate these results.

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