Rockwell Hanford Operations

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☐	N/A Peer Review			
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☒	P. J. Reder <i>P J Reder</i> Configuration Management	1/19/87		
☒	W. L. Heilman <i>W L Heilman</i> Systems Integration Dept. Mgr.	3/21/86		
☒	M. D. Veatch <i>M D Veatch</i> Acting Mgr. Site Dept.	3/14/86		
☒	S. M. Baker <i>S M Baker</i> Manager Site Analysis	3/14/86		
☒	M. J. Smith <i>M J Smith</i> Research Manager	3/25/86		
☒	R. C. Arthur <i>R C Arthur</i> Informal Reviewer	3/19/86		
☒	S. H. Hall <i>S H Hall</i> Informal Reviewer	3/19/86		
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☐	Engineering and Design			
☒	G. T. Harper <i>G T Harper</i>	7/11/86		
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1.0 INTRODUCTION

The Basalt Waste Isolation Project (BWIP) of Rockwell Hanford Operations is investigating the potential of using selected flows within the Columbia River Basalt Group underlying the Hanford Site in southeastern Washington State as the location of a deep repository for the permanent disposal of high-level radioactive wastes. As part of the site characterization activities at the Hanford Site, quantitative information relative to the concentration and distribution of methane dissolved in groundwater is needed:

- To estimate the potential economic importance of methane
- To estimate the effect on mine safety
- As a constraint on conceptual groundwater flow models.

For about 3 yr, the BWIP has collected dissolved-gas data from which absolute concentrations of methane and other components can be computed. Some gas concentration data have been reported previously by the BWIP (Wintczak 1984; Mack 1983). In both BWIP publications, average values for field data (not always specified by the author) were used in the calculations so that reported gas concentrations are given as mean values rather than as a range of values that more accurately reflect measurement uncertainties.

The objective of this document is to consolidate all available data (from BWIP and non-BWIP sources) for the Hanford Site* and to present a summary table of gas concentration results. In addition, details of the computational procedures required to reduce data obtained from the field measurements made by the BWIP are provided in the appendix. Because an approved procedure for measurement of gas flow was not in place for the data gathering activities of the BWIP reported in this document, the resultant data are preliminary.

2.0 MEASUREMENT TECHNIQUES

The field technique used by the BWIP for determining the absolute concentrations of gaseous components dissolved in groundwater makes use of a 114 L (30 gal) stainless steel gas separation barrel (fig. 1). A descriptive summary of the operation of this apparatus is provided by Weiss (1985).

"The gas barrel assembly is attached to the wellhead discharge line with a hose large enough to accommodate all flow from the borehole. A length of tygon [latex was used] tubing is connected to the gas outlet at the top of the barrel and submerged in a bucket of water to prevent

*The BWIP is unaware of any published data for dissolved gases in groundwaters from the Columbia River basalts beyond the Hanford Site boundaries.

Figure 1. Gas Separation Barrel.

air from being drawn into the gas separation barrel. Atmospheric gases are purged from the barrel by covering the water outlet and allowing it to completely fill with water. As the barrel fills, the atmospheric gases are expelled through the ball valve on the top. After the pressure in the barrel has been allowed to equilibrate with the atmospheric pressure, the flowmeter is connected to the tygon tubing leading from the gas valve. The flowmeter consists of a calibrated glass tube in which floats a ball (either glass, carbon, or stainless steel, depending on the flow rate). As gas flows through the tubing, the ball floats upward, its height being proportional to the gas flow rate."

Two different gas flowmeters have been used by the BWIP. Fischer Porter (Series 10A1460-LK) and Manostat flowmeters were operated according to directions appearing in the respective instruction manuals. Each of the flowmeters comes with a selection of flow tubes to accommodate different gas flow rates. In the field notebook (Rockwell 1981, p. 44), reference is made to a Gilmont tube (size 10) which was used for one sampling event (Cohasset flow bottom at McGee Well). It is not known which flowmeter stand was used with this tube; this omission does not affect the quality of measured results.

The relative amounts (in mole percent) of each component of the dissolved-gas samples collected by the BWIP are determined by mass spectrometry. Samples are collected of either the gas phase discharging from the top of the gas separation barrel (into a glass sample container) or water pumped from the borehole (into a stainless steel collection bottle) from which dissolved gases are extracted in vacuo.

In addition to the data collected by the BWIP, quantitative dissolved-gas data for groundwaters from four Hanford Site boreholes is reported in Apps et al. (1979). Field sampling and laboratory procedures for gas analyses reported by Apps et al. (1979) are fully described in that document.

3.0 PRESENTATION OF RESULTS

Figure 2 is a location map of the Hanford Site identifying all boreholes for which dissolved-gas concentration data are available. Table 1 is a listing of the concentration results by borehole and zone. Table 2 contains gas composition data for these samples as reported in the BWIP hydrochemical data base (Early et al. 1985) or as calculated from data appearing in Apps et al. (1979). The method by which gas concentrations were computed from BWIP data for table 1 is detailed in the appendix.

Most of the gas concentrations in table 1 are listed as ranges of values. This reflects the fact that both the observed water and gas flow rates typically fluctuate over the time of measurement. Flow integrators for measuring gas and water discharge rates were not used. For most of the data reported, the minimum and maximum concentrations based on observed gas and water flow rates are given. The possibility exists that observed

Figure 2. Location Map of the Hanford Site Showing Boreholes for Which Dissolved Gas Concentration Data are Available.

Table 1. Dissolved Gas Concentrations in Groundwaters from Hanford Site Boreholes. (sheet 1 of 2)

Borehole	Stratigraphic zone ^a	Depth interval ^a (ft)	Pumping technique ^b	Calculated gas concentration ^c (mg/l)	Methane percent saturation at depth ^d	Source of data
DB 7	Mabton interbed	778-814	Windmill pump	CH ₄ 0.623 (if CH ₄) ^e (10) N ₂ 0.824 (if N ₂) ^e (10)		BWIP
DC 2	Grande Ronde Formation	3,243-3,273	Swab	CH ₄ 43 N ₂ 299 ^f		Apps et al. (1979)
DC 6	Grande Ronde Formation	2,260-4,333	Flowing	N ₂ 17 (7-21-78) 28 (8-16-78)		Apps et al. (1979)
DC 7	Grande Ronde Formation	2,780-3,948	Submersible pump	N ₂ 5.5		BWIP
DC 16A	Frenchman Springs Member	2,476-2,559	Submersible pump	CH ₄ 225-358 ⁽¹⁰⁾ 358 ^(c)	21-34 34	BWIP
	Vantage interbed	2,671-2,730	Windmill pump	CH ₄ 345-663 ⁽¹⁰⁾ 424-584 ^(c)	31-60 39-53	BWIP
	Rocky Coulee flow	2,836-2,946	Windmill pump	CH ₄ 102-487 ⁽¹⁰⁾ 256-358 ^(c)	9-43 22-31	BWIP
DC 19C	Frenchman Springs Member	1,826-1,952	Submersible pump	CH ₄ 16-40 ⁽¹⁰⁾ N ₂ 28-70 ⁽¹⁰⁾		BWIP
	Frenchman Springs Member	2,421-2,468	Submersible pump	CH ₄ 55-61 ⁽¹⁰⁾ N ₂ 42-47 ⁽¹⁰⁾		BWIP
	Cohasset flow top	3,008-3,105	Submersible pump	CH ₄ 26-35 ⁽¹⁰⁾ N ₂ 44-59 ⁽¹⁰⁾ CH ₄ 33 ^(c) N ₂ 56 ^(c)		BWIP
	Umtanum flow top	3,567-3,586	Submersible pump	CH ₄ 84 ^(b) N ₂ 99 ^(b)		BWIP
DC 20C	Wanapum-Grande Ronde composite	1,581-3,781	Pump	CH ₄ 27-30 ⁽¹⁰⁾ N ₂ 34-38 ⁽¹⁰⁾		BWIP
RRI 2	Cohasset flow bottom	3,247-3,344	Windmill pump	CH ₄ 392-1,135 ⁽¹⁰⁾ (9-17-82) 378 ± 135 593 ± 211 ⁽¹⁰⁾ (10-24-83)	32-93 31-48	BWIP
	Umtanum flow top	3,568-3,781	Windmill pump	CH ₄ 610-801 ⁽¹⁰⁾	47-62	BWIP
	Umtanum fracture zone	3,781-3,827	Windmill pump	CH ₄ 468-1,222 ⁽¹⁰⁾	36-93	BWIP
	Very high Mg flow	3,837-3,889	Windmill pump	CH ₄ 512-860 ⁽¹⁰⁾	39-65	BWIP

Table 1. Dissolved Gas Concentrations in Groundwaters from Hanford Site Boreholes. (sheet 2 of 2)

Borehole	Stratigraphic zone ^a	Depth interval ^a (ft)	Pumping technique ^b	Calculated gas concentration ^c (mg/l)	Methane percent saturation at depth ^d	Source of data
RRL-6B	Umtanum flow top	3,708-3,823	Windmill pump	CH ₄ 277-742 ⁽¹¹⁰⁾	21-57	BWIP
RRL-14	Cohasset flow top	3,017-3,147	Windmill pump	CH ₄ 363-645 ⁽¹¹⁰⁾	31-55	BWIP
	Cohasset flow top	3,077-3,140	Windmill pump	CH ₄ 127 ± 25-804 ± 160 ⁽¹¹⁰⁾ 381 ± 76-569 ± 114 ⁽⁴⁾	11-68 32-48	BWIP
	Cohasset flow bottom	3,294-3,403	Windmill pump	CH ₄ 409-566 ⁽¹¹⁰⁾	33-46	BWIP
	Umtanum flow top	3,715-3,814	Windmill pump	CH ₄ 390	30	BWIP
McGee	Priest Rapids Member	691-978	Flowing	CH ₄ 6 N ₂ 19 ⁽¹⁾		Apps et al. (1979)
	Cohasset flow bottom	2,393-2,524	Windmill pump	CH ₄ 1.3-3.7 N ₂ 9-25		BWIP
	Umtanum flow bottom	2,854-3,123	Windmill pump (?)	CH ₄ 0.37 ⁽¹¹⁰⁾ N ₂ 0.141 ⁽¹¹⁰⁾		BWIP
Ford	Priest Rapids Member	6,207-777	Flowing	CH ₄ 6 N ₂ 19		Apps et al. (1979)

NOTE: m = ft x 0.3048

TR = Total range of computed gas concentration values

S = Stable value or stable range of gas concentration values

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^aStratigraphic zone designations and depth intervals (except for DB-7) are from Early et al. (1985) and may differ slightly from that recorded in Rockwell (1981).

^bThe term pump refers to a line shaft turbine pump. Use of a pump or a submersible pump for sampling may lead to some escape of gas from the borehole annulus. The measured gas concentrations associated with these techniques are minimum values. For the other techniques listed, the entire flow of water (and gas) goes into the sampling device.

Dissolved gas concentrations made by the BWIP are based only on gases that exsolve from groundwater in the separation barrel. No measurement of the residual dissolved gas component is made. Concentrations from Apps et al. (1979) include the total dissolved-gas component.

^dPercent saturation values for methane are listed only for the samples with high methane concentrations in boreholes, DC-16A, RRL-2, RRL-6B, and RRL-14. See appendix for calculation details.

^cGas composition was not determined. Concentrations are based upon assuming a gas composition of pure CH₄ and N₂, respectively.

^eThe DC-2 borehole was flushed with N₂ during swabbing and sampling.

^fGas concentration of N₂ corrected for presumed air contamination.

^gCalculations reported in Mack (1983) are analogous but use different gas compositions. In addition, based on records in Rockwell (1981, p. 49) for the Cohasset flow top in DC-19C, gas flow rate reported in the referenced letter was incorrectly determined because the wrong column in the flowmeter calibration table was used.

The plus-minus uncertainty is based on reported uncertainty in water flow rate.

Table 2. Dissolved Gas Compositions (Mole Percent) for Groundwaters from Hanford Site Boreholes. (sheet 1 of 2)

Borehole	Stratigraphic zone ^a	Depth interval ^a (packer setting) (ft)	Sample numbers	Composition (mole percent) ^b							
				N ₂	O ₂	CO ₂	Ar	He	H ₂	CH ₄	CO
DB-7	Mabton interbed	778-813									
DC-2	Grande Ronde Formation	3,243-3,273		79.3	<0.01	(c)	0.58	(c)	0.31	19.8	NR
DC-6	Grande Ronde Formation	2,260-4,333		96.5	0.12	<0.14	2.0	(c)	0.11	1.1	NR
			96.0	0.08	2.3	0.91	(c)	(c)	0.75	7.21/8 NR 8.16/8	
DC-7	Grande Ronde Formation	2,780-3,948	82-23	91.30	0.59	<0.01	1.19	0.21	0.08	4.37	2.30
			82-56	92.00	0.37	<0.01	1.13	0.21	0.09	4.39	1.80
			82-10A	94.60	<0.01	0.02	1.01	0.19	<0.01	4.14	<0.10
			82-10B	91.00	0.54	<0.01	1.10	0.20	0.03	5.16	2.00
			82-33	89.70	0.64	<0.01	1.10	0.20	0.03	5.19	3.20
DC-16A	Frenchman Springs Member	2,476-2,559	82-322	3.28	<0.01	0.06	0.06	<0.010	0.01	96.60	<0.10
			82-334	1.86	<0.01	<0.01	0.04	0.14	<0.01	98.00	<0.10
			82-355	1.91	<0.01	<0.01	0.04	0.01	<0.01	98.00	<0.10
			82-394	1.91	<0.01	<0.01	0.06	<0.010	<0.01	98.00	<0.10
	Vantage interbed	2,671-2,730	82-430	1.78	0.09	0.07	0.03	<0.010	0.04	98.00	<0.10
			82-464	1.73	0.01	<0.01	0.03	<0.010	<0.01	98.20	<0.10
	Rocky Coulee flow	2,836-2,946	83-20	19.00	4.80	0.04	0.24	<0.010	<0.01	75.90	<0.10
			83-29	1.59	<0.01	<0.01	0.03	<0.010	<0.01	98.40	<0.10
			83-54	1.49	<0.01	0.01	0.03	<0.010	<0.01	98.50	<0.10
	DC-19C	Frenchman Springs Member	1,826-1,952	84-53	69.10	14.40	0.05	1.04	<0.010	<0.01	15.40
Frenchman Springs Member		2,421-2,468	84-40A	29.60	0.10	1.30	0.90	(c)	<0.01	68.10	<0.10
			84-40B	40.60	<0.01	0.30	1.20	<0.003	<0.01	57.90	<0.10
			84-44	58.50	<0.01	0.01	0.94	0.004	<0.01	40.60	<0.10
Columbia flow top		3,008-3,105	84-73	45.70	<0.01	0.01	0.97	0.007	<0.01	50.50	2.80
			84-75	48.90	0.04	0.07	0.79	<0.010	0.03	47.80	2.40
Umatanum flow top		3,567-3,586	84-46	37.40	<0.01	0.01	0.68	<0.010	<0.01	60.00	1.90
			84-86	41.20	0.98	0.08	0.66	<0.010	0.10	56.00	0.90
DC-20C	Wanapum-Grande Ronde composite	1,581-3,781	84-76	30.40	0.04	0.08	0.73	0.006	<0.01	68.80	<0.10
			84-79	53.00	<0.01	0.01	0.80	<0.003	<0.01	46.30	<0.10

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Table 2. Dissolved Gas Compositions (Mole Percent) for Groundwaters from Hanford Site Boreholes. (sheet 2 of 2)

Borehole	Stratigraphic zone ^a	Depth interval ^b (packer setting) (ft)	Sample numbers	Composition (mole percent) ^b							
				N ₂	O ₂	CO ₂	Ar	He	H ₂	CH ₄	CO
RRR-2	Cohasset flow bottom	3,247-3,344	82-401	2.36	<0.01	0.04	0.04	<0.010	<0.01	97.60	<0.10
			82-414	62.70	16.70	0.07	0.76	<0.010	<0.01	19.90	<0.10
			82-479	1.45	<0.01	<0.01	0.02		0.17	98.20	<0.10
			84-47	1.26	<0.01	<0.01	0.02	<0.010	0.02	98.70	<0.10
	Umtanum flow top	3,568-3,781	82-364	1.69	0.14	0.04	0.03	<0.010	0.20	97.90	(c)
			82-370	3.11	0.03	<0.01	0.05	<0.010	0.11	96.70	<0.10
			82-391A	1.97	<0.01	0.02	0.03		0.16	97.80	<0.50
			82-391B	1.87	<0.01	0.01	0.03	0.01	0.15	97.90	<0.50
			82-391C	2.21	0.31	0.02	0.04	0.01	0.17	97.20	<0.50
	Umtanum fracture zone	3,781-3,827	82-383	3.56	0.07	<0.01	0.06	<0.010	0.13	96.20	<0.10
	Very high Mg flow	3,837-3,889	82-411	1.08	<0.01	<0.01	0.02	<0.010	<0.01	98.90	<0.10
			82-445	1.64	<0.01	<0.01	0.02	<0.010	<0.01	98.30	<0.10
			82-456	2.43	0.03	<0.01	0.03	0.02	0.03	97.50	<0.10
RRR-6	Umtanum flow top	3,708-3,823	83-25	2.08	<0.01	0.04	0.04	<0.010	<0.01	97.80	<0.10
			83-69	2.72	0.44	0.02	0.06	0.290	<0.01	96.50	<0.10
RRR-14	Cohasset flow top	3,017-3,147	82-418	1.65	0.02	<0.01	0.02	<0.010	<0.01	98.30	<0.10
			82-448	1.61	<0.01	<0.01	0.02	<0.010	<0.01	98.40	<0.10
	Cohasset flow top	3,077-3,140	84-11	2.02	<0.01	0.01	0.04	0.010	<0.01	97.90	<0.10
	Cohasset flow bottom	3,294-3,403	83-151	2.17	<0.01	0.05	0.26	<0.010	<0.01	97.50	<0.10
	Umtanum flow top	3,715-3,814	83-49	1.90	0.03	0.02	0.04	<0.010	<0.01	98.00	<0.10
			83-96	1.91	0.03	0.01	0.04	<0.010	<0.01	98.00	<0.10
McGee	Priest Rapids Member	691-978		72.5	15.3	2.0	0.98	(c)	(c)	9.3	NR
	Cohasset flow bottom	2,393-2,524	83-513	75.10	0.03	0.04	0.95	0.040	<0.01	19.10	4.70
	Umtanum flow bottom	2,854-3,123	84-24	67.90	0.01	0.09	0.89	0.060	<0.01	31.00	<0.10
Ford	Priest Rapids Member	620-777		57.2	0.52	7.1	1.3	(c)	0.02	33.8	NR

NR = not reported

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^aStratigraphic zone designations and depth intervals (except for DB-7) are from Early et al. (1985) and may differ slightly from that recorded in Rockwell (1981).

^bSee table 1 for source of data. Data for BWIP samples are reported in Early et al. (1985). Data from Apps et al. (1979) have been recalculated on a mole percent basis.

(c) Amount was below detection level.

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fluctuations in gas flow rates are related to fluctuations in water flow rates. However, data to support this hypothesis are not available. Therefore, the total range of computed gas concentrations reported in table 1 includes contributions from both observed gas and water flow rate variations. If the gas flow was observed to stabilize during measurement, both the total range of gas concentrations and a more restricted range of values representing the stable flow conditions are reported. Occasionally, water flow rates are reported in a field notebook (Rockwell 1981) with a plus or minus uncertainty. This uncertainty is transferred explicitly into the calculated concentrations.

In addition to uncertainties in gas and water flow rates that are explicitly included in the concentration ranges listed in table 1, other potential sources of error exist:

- Use of pumping devices in boreholes that allow a portion of the exsolved gases to escape through the annulus
- Diversion of only a portion of the flow from the borehole through the gas separation barrel
- Condensation of water within the flowmeter
- Incomplete separation of gas from water in the gas separation barrel
- Contamination of formation gases with air
- Uncertainty of temperature and pressure of the separated gas phase in the barrel.

In theory, accurate gas flow measurements can be obtained only when the sampled zone is isolated by packers and the entire flow of water from the borehole is pumped through the gas separation barrel. In table 1, samples collected with the windmill-type pump or from zones flowing under artesian heads conform to this requirement. When a submersible pump or a line shaft turbine pump (pump) is utilized and high gas concentrations are encountered, a fraction of the dissolved gas may separate within the borehole and escape through the annulus rather than enter the gas separation barrel. Consequently, observed gas flow rates measured under these conditions will be lower limits. This possible source of error exists only when the intake of the pump is above the water depth at which gas exsolution occurs.

Moreover, if only a fraction of the total fluid flow from the borehole is diverted to the gas separation barrel and if some gas separation occurs prior to the point of diversion, gas flow measurements may be in error. There is no way to know if the amount of gas observed is too high or too low. Measurements made at boreholes DC-19C and DC-20C are examples of sampling events where this problem was encountered.

During gas flow measurements, the gas discharging from the top of the gas separation barrel passed through ~3 m (10 ft) of latex tubing before entering the flowmeter. The gas stream cooled as it passed through the tubing so that excess water vapor entrained in the gas could condense. Had condensation occurred on the floating bead, it could have resulted in its sticking and could possibly have led to erroneously low readings.

The fluid in the gas separation barrel is at atmospheric pressure. All gas measured by the flowmeter has exsolved from the water either in the pipe coming from the sampled zone in the borehole or during its time of residence in the gas separation barrel. A fraction of the total gas components of the groundwater will remain in solution due to its solubility and leave with the water exiting the barrel. This effect will result in underestimates in the calculated dissolved gas concentration but will be important only for those few samples listed in table 1, with very low reported gas concentrations.

In a closely related problem, if the water flow rate to the gas separation barrel is large, its residence time is short. Since the amount of gas measured is directly proportional to the efficiency of exsolution, large water flow rates tend to result in underestimates of dissolved-gas concentrations. For some sampling events reported in table 1, the water residence time was as low as two minutes. Incomplete gas exsolution is a possibility for short water residence times, but no objective criterion for correction of the data is available.

For several sampling events noted in table 2, observed gas compositions included significant proportions of oxygen. For the concentrations presented in table 1, it is assumed that this oxygen resulted from air contamination because the basalt geochemical environment is believed to be highly reducing (DOE-RL 1982). A discussion of the correction procedure is provided in the appendix.

The calculations summarized in the appendix require corrections for gas temperature and pressure. For most samples, the water temperature in the gas separation barrel was reported and assumed to be equivalent to the gas temperature which may be incorrect. For several samples, a temperature was not recorded but was assumed to be 25 °C. Both of these assumptions probably led to the introduction of small errors. Atmospheric pressure was not reported for many gas sampling events. For these occurrences, pressure values for the date and approximate time of gas sampling were obtained from the meteorology station operated by Pacific Northwest Laboratory (PNL), near the 200 West Area. No correction for elevation was applied to the pressure values. For all other sampling events, the reported atmospheric pressure values were also obtained from PNL but elevation corrections were applied. In any event, the magnitude of corrections to dissolved gas concentrations due to temperature and pressure are small (less than ~5% for temperature and 1% for pressure for the worst case) and any uncertainties in computed gas concentrations resulting from errors in measurement are insignificant, relative to uncertainties in observed flowmeter readings reported in table 1.

4.0 DISCUSSION

The uncertainties of gas concentrations reported in table 1 provide a rather wide range of values for most sampling events. Furthermore, the additional potential uncertainties discussed in section 3.0, while not quantifiable, should not be ignored. Most of these possible sources of error tend to underestimate the true dissolved-gas concentrations. Regardless of the acknowledged uncertainties, several key observations are possible.

- The reference repository location is a region of high dissolved-methane concentrations in both the Wanapum and Grande Ronde Formations (~30 to 60% saturated at in situ conditions).
- Gas concentrations appear to drop off rapidly with increasing lateral distance from the reference repository location.
- Several boreholes near to or on the periphery of the reference repository location appear to have unusually high concentrations of nitrogen.

According to table 1, boreholes lying within the reference repository location (DC-16A, RRL-2, RRL-6, and RRL-14) have methane concentrations that range from ~30 to 60% of saturation (at formation conditions). As a result of the large uncertainties associated with two measurements (both from RRL-2), inferred concentrations as high as 90 to 95% of saturation may exist. However, currently there is no evidence to suggest that any of the measured groundwaters are saturated with methane which would imply a two-phase fluid system (i.e., gas + water). If these observations are confirmed by more precise, future dissolved-gas analyses, they will certainly be important considerations in refining our understanding of the groundwater flow system in the vicinity of the reference repository location and assessing the possible economic importance of methane.

The possible impact of relatively high dissolved-methane concentrations in the reference repository location on the safety of workers in a deep repository also requires consideration. In view of the uncertainties concerning methane concentrations, until more precise gas measurements are made, it seems most prudent to assume that, at a maximum, groundwaters in the vicinity of the proposed repository are saturated with methane under ambient conditions. The curve in figure A-1 permits one to estimate the saturation concentration of methane for Hanford Site groundwater. For example, at borehole RRL-2 at the depth of the Cohasset flow top (~915 m (3,002 ft)), the solubility of methane is ~1,170 mg/L. By assuming a dissolved methane concentration of this magnitude, a conservative estimate for the likely inflow of methane associated with groundwater to the exploratory shaft and repository will result. This assumption also includes an implicit provision that the fluid system is single phase (i.e., water).

With the exception of boreholes in the vicinity of the reference repository location, total dissolved-gas concentrations (irrespective of the relative amount of methane and nitrogen) >100 mg/L are rare. Indeed, data presented in table 1 illustrate that boreholes DC-6, DC-7, McGee, and Ford have particularly low total gas concentrations.

Boreholes DC-19C and DC-20C, which lie near the edge of the reference repository location, appear to have rather high concentrations of dissolved nitrogen. The amounts observed exceed that which can be attributed to the equilibrium between the atmosphere and water at the time of recharge. These boreholes were drilled with an aerated mist technique. However, gas compositions obtained for these sampling events do not indicate measurable amounts of oxygen (see table 2). Therefore, contamination of the sampled zones with compressed air during drilling is an unlikely explanation for the high nitrogen concentrations. As noted in section 3.0, gas flow measurements made in these boreholes may be in error due to the fact that only a fraction of the water pumped from sampled zones passed through the gas separation barrel. The quality of these analyses requires reevaluation. It is recommended that these boreholes be sampled more carefully in the future.

5.0 SUMMARY AND CONCLUSIONS

Table 1 presents all available dissolved-gas data for Hanford Site boreholes. The original field data have been evaluated rigorously so that a range of concentration values appropriate to the uncertainties associated with the original measurements are reported. The quality of the concentration data collected by the BWIP is difficult to assess. However, it is observed that duplicate analyses for the Cohasset flow bottom in borehole RRL-2 and a zone within the Grande Ronde Formation in DC-6 appear to be similar. Furthermore, the continuity of results for boreholes in which a sequence of samples have been obtained from increasing depths (e.g., DC-16A, DC-19C, RRL-2, and RRL-14) suggests that calculated methane concentrations may be approximately correct.

In summary, it is believed that the data are qualitatively useful and can be incorporated into studies on the development of conceptual groundwater flow models. Most measured values for methane concentration from reference repository boreholes are in the range of from 30 to 60% saturation (~350 to 700 mg/L for the Cohasset flowtop at RRL-2). In the past, the BWIP has published a concentration value of 700 mg/L for methane for the Cohasset flowtop at RRL-2 (Wintczak 1984) which has been used for designing the exploratory shaft ventilation systems (FitzPatrick 1985) and conducting hydrothermal tests of waste package components (McGrail 1985). Clearly, this value is consistent with the results of this study. However, in recognition of significant sampling uncertainties as outlined above,

it is recommended tentatively that a conservative methane concentration of 1,200 mg/L (methane saturated) in groundwater be assumed as an upper-bounding value. Refinement of this methane concentration estimate is necessary and will be provided by future characterization studies.

The uncertainties associated with the measured concentrations of methane and nitrogen presented in table 1 suggest that new sampling and analytical procedures be adopted. The BWIP recently procured several downhole sampling devices capable of collecting dissolved gases in situ. Addition of a more reliable gas extraction process and analysis both by gas chromatographic and mass spectrometric techniques should greatly improve future measurements.

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APPENDIX A

Procedures for Computing Dissolved Gas Concentrations in Groundwater

Table A-1 contains those measurements from which gas concentrations can be computed for the BWIP data. The units for each parameter are standardized for convenience although the field notebook reports data in several different forms (Rockwell 1981). Conversion factors used are provided in table A-1. Following is an example of the calculations necessary to reduce data obtained with the gas separation barrel to absolute concentrations of each component. It follows the format of Mack (1983) and is consistent with the instruction manuals for the flowmeters used.

Sample Calculation

Borehole:	DC-19C
Stratigraphic zone:	Cohasset flow top
Depth interval:	3,008-3,105 ft
Temperature:	27 °C
Pressure:	734.6 mm of Hg
Vapor pressure of H ₂ O:	26.739 mm of Hg (at 27 °C)
Water flow rate:	63.2 L/min
Observed gas flow rate:	4,806-6,555 cm ³ /min

The observed gas flow rates for the above zone are read from calibration curves or tables in the instruction manuals for the flowmeters and assume a composition of air. The observed flow rate is multiplied by a correction factor to compensate for gas compositions other than that of air.

$$\text{Correction factor} = \sqrt{\frac{\text{density of air at STP}}{\text{density of gas at STP}}}$$

where the standard temperature and pressure (STP) is defined as

STP = 70 °F (21 °C) and 1 atm (760 mm of Hg) of pressure

Table A-1. Field Measurements Associated with Use of the Gas Separation Barrel in Hanford Site Boreholes. (sheet 1 of 2)

Borehole	Stratigraphic zone ^a	Depth interval ^a (ft)	Temperature ^b (°C)	Atmospheric pressure ^c (mm of Hg)	Observed water flow rate (L/min)	Observed gas flow rate (cm ³ /min)
DB 7	Mabton interbed	778-814	23.0	741.9	3.18	0-2,360 ⁽¹¹⁾
DC 7	Grande Ronde Formation	2,780-3,948	26 (25.3, 26.0)	743.5*	18.9	94
DC 16A	Frenchman Springs Member	2,476-2,559	34.5	744.6	32.2	9,202-14,629 ⁽¹¹⁾ 14,629 ⁽¹²⁾
	Vantage interbed	2,671-2,730	23.4	752.7	7.57	3,067-5,899 ⁽¹¹⁾ 3,775-5,191 ⁽¹²⁾
	Rocky Coulee flow	2,836-2,946	19	746.3*	7.95	944-4,483 ⁽¹¹⁾ 2,360-3,303 ⁽¹²⁾
DC 19C	Frenchman Springs Member	1,826-1,952	28.1	749.4*	16.2-28.4 37.47	4,746-6,538 ⁽¹¹⁾ 6,161-7,564 ⁽¹²⁾
	Frenchman Springs Member	2,421-2,468	24.7	739.7*	65.4	7,115-7,934 ⁽¹¹⁾
	Cohasset flow top	3,008-3,105	27.0	734.6	63.2	4,806-6,555 ⁽¹¹⁾ 6,070-6,194 ⁽¹²⁾
	Umtanum flow top	3,567-3,586	22.7	740.7	22.7	4,453
DC 20C	Wanapum-Grande Ronde composite	1,581-3,781	32.5	742.7*	17.1	1,141-1,264 ⁽¹¹⁾
RRI 2	Cohasset flow bottom	3,247-3,344	22.9	750.6	5.69 5.43 5.18	2,594-4,955 ⁽¹¹⁾ 2,831-5,191 ⁽¹²⁾ 5,899-6,843 ⁽¹¹⁾ 3,067-5,191 ⁽¹²⁾ (9.17-8.2)
			19.5	750.8* ^d	5.30 ± 1.89	2,315-3,630 ⁽¹¹⁾ (10.24-8.3)
	Umtanum flow top	3,568-3,781	29.4	744.9	6	4,483-5,899 ⁽¹¹⁾
	Umtanum fracture zone	3,781-3,827	25.9	745.9	5.45-6.67	3,775-8,022 ⁽¹¹⁾
	Very high Mg flow	3,837-3,889	22.3	746.3	4.43 5.06 4.28	3,067-4,483 ⁽¹¹⁾ 3,067-4,955 ⁽¹²⁾ 3,067-4,247 ⁽¹¹⁾

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Table A-1. Field Measurements Associated with Use of the Gas Separation Barrel in Hanford Site Boreholes. (sheet 2 of 2)

Borehole	Stratigraphic zone ^a	Depth interval ^a (ft)	Temperature ^b (°C)	Atmospheric pressure (mm of Hg)	Observed water flow rate (L/min)	Observed gas flow rate (cm ³ /min)
RRL 6B	Umtanum flow top	3,708-3,823	25*	733.0	3.54 3.53 3.38	1,180-3,067 ⁽¹¹⁾ 1,416-3,067 ⁽¹¹⁾ 1,416-3,067 ⁽¹¹⁾
RRL 14	Cohassett flow top	3,017-3,147	21.3	741.4	3.73-4.92	2,124-2,831 ⁽¹¹⁾
	Cohassett flow top	3,077-3,140	20.0	744.1*	4.92 ± 0.98	732-4,629 ⁽¹¹⁾ 2,194-3,275 ⁽¹¹⁾
	Cohassett flow bottom	3,294-3,403	25*	738.1*	7.59	3,775-5,191 ⁽¹¹⁾
	Umtanum flow top	3,715-3,814	25*	742.0	8	3,775
McGee	Cohassett flow bottom	2,393-2,524	27.9	743.2*	2.35	25-70 ⁽¹¹⁾
	Umtanum flow bottom	2,854-3,123	25.7	743.8*	4.92	0-864 ⁽¹¹⁾

NOTE: m = ft x 0.3048

TR = total range of observed gas flow values

S = stable value or stable range of gas flow values

Gas flow SCFH x 471.9 = cm³/min

Pressure in Hg x 25.4 = mm of Hg

Water flow = gal/min x 3.7853 = L/min

60/(s/L) = L/min

L/s x 60 = L/min

mL/s x 0.06 = L/min

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^aStratigraphic zone designations and depth intervals (except for DB-7) are from Early et al. (1985) and may differ slightly from that recorded in Rockwell (1981).

^bWater temperature is as reported in Rockwell (1981). When not available in the field notebook, data were taken from Early et al. (1985) for the sampling event. Temperatures not available in either source were assumed to be 25 °C and noted with an asterisk.

^cAtmospheric pressure readings are as reported in Rockwell (1981). When not available in the field notebook, data were obtained from the meteorological station near the 200 West Area operated by Pacific Northwest Laboratory. These data are noted with an asterisk.

^dIncorrect pressure reported in Rockwell (1981).

and the gas density is computed as follows. (Table A-2 contains the mole percent of each gas component and table A-3 contains gas densities for this example.)

$$\begin{aligned}\text{Density of gas} &= \sum_{\text{all } i} \frac{\text{mol\% gas } i}{1000} \times (\text{density of gas } i \text{ at STP}) \\ &= (0.473)(0.00116) + (0.0088)(0.00166) \\ &\quad + (0.492)(0.0006653) + (0.026)(0.001163) \\ &= 0.000921 \text{ g/cm}^3\end{aligned}$$

$$\text{Correction factor} = \sqrt{\frac{0.0012}{0.000921}} = 1.141$$

$$\text{Corrected gas flow rate} = 5,484 - 7,479 \text{ cm}^3/\text{min}.$$

Assuming that the gas mixture behaves as an ideal gas, one can compute the total number of moles of gas discharging through the flowmeter during one minute by use of the ideal gas law:

$$PV = nRT$$

where

$$\begin{aligned}P &= \frac{\text{gas pressure (mm of Hg)}}{760 \text{ (mm of Hg/1 atm)}} \\ &= \frac{\text{atmospheric pressure (mm of Hg)} - \text{vapor pressure of H}_2\text{O (mm of Hg)}}{760 \text{ (mm of Hg/1 atm)}}\end{aligned}$$

$$V = \text{gas volume (cm}^3/\text{min)}$$

$$n = \text{number of moles of gas/min}$$

$$R = \text{universal gas constant} = 82.057 \text{ cm}^3 \cdot \text{atm/mol K}$$

$$T = \text{temperature (K)}$$

and

$$n = \frac{PV}{(760) RT} = \frac{(734.6 - 26.793)(5,484)}{(760)(82.057)(273 + 27)} = 0.207473 \text{ mol of gas/min}.$$

Table A-2. Gas Compositions
Measured for the Cohasset
Flow Top at Borehole DC-19C
from Table 2.

Gas	Mole percent		Mean
	Sample numbers		
	84-73	84-75	
N ₂	45.7	48.9	47.3
Ar	0.97	0.79	0.88
CH ₄	50.5	47.8	49.2
CO	2.8	2.4	2.6

Table A-3. Densities of Selected
Gases at Standard Temperature and
Pressure (70 °F (21 °C) and
1 atm of pressure).

Gas	Density at STP (g/cm ³)
Air	0.001200
Argon	0.00166
Carbon dioxide	0.001835
Carbon monoxide	0.001163
Methane	0.0006653
Nitrogen	0.001160
Oxygen	0.001326

Source: Manostat (n.d.).

NOTE: STP = Standard temperature and pressure.

Therefore, the following relationship holds:

$$\begin{aligned}\text{Gas flow rate} &= 0.207473 \text{ mol of gas}/63.2 \text{ L H}_2\text{O} \\ &= 0.003293 \text{ mol of gas/L H}_2\text{O}.\end{aligned}$$

To convert the number of moles to a concentration of each gas component:

$$n_i = \frac{(n)(\text{mol\% of gas } i)}{100}.$$

Therefore,

$$\text{N}_2 \text{ concentration} = (0.003293)(0.473) = 0.001553 \text{ mol/L}$$

$$\text{CH}_4 \text{ concentration} = (0.003293)(0.492) = 0.001615 \text{ mol/L}^*$$

*At this point in the calculation scheme if the sample is believed to be contaminated with air (i.e., more than several mole percent oxygen is present in the analysis) a correction is made. Atmospheric air has a composition as follows:

<u>Constituent</u>	<u>Volume (mole percent)</u>
N ₂	78.084
O ₂	20.948
CO ₂	0.0315
Ar	0.934

Source: Ozima and Podosek (1983).

Therefore, on a mole basis, 1 mol of O₂ will carry with it

$$\frac{78.084}{20.946} = 3.728 \text{ mol of N}_2.$$

By a simple proportional relationship the number of mol/L of O₂ measured in the sample can be used to compute the number of mol/L of N₂ associated with it. By subtraction, the effects of atmospheric contamination can be removed from the original gas analysis. The same procedure can be used to correct for the effects of contamination to the other gas components.

but since

Weight concentration = mole concentration x mole weight

$$= \frac{\text{mol}}{\text{L}} \times \frac{\text{g}}{\text{mol}}$$

then

$$\text{N}_2 \text{ concentration} = (0.001553)(28) = 0.0435 \text{ g/L} = 44 \text{ mg/L}$$

$$\text{CH}_4 \text{ concentration} = (0.001615)(16) = 0.0258 \text{ g/L} = 26 \text{ mg/L}.$$

The solubility of methane at elevated temperatures and pressures is required to compute the level of saturation for groundwaters listed in table 1. Culberson and McKetta, Jr. (1951) provide experimental methane solubility data that can be utilized. They report solubility results for 77 °F (25 °C), 100 °F (38 °C), and 160 °F (71 °C) among others. In Early et al. (1983, p. I-184), the following temperature-depth relationship for Hanford Site boreholes is given:

$$T(^{\circ}\text{C}) = 15 + 0.038 \text{ depth (m)} .$$

For the temperatures listed above, one can compute the appropriate depths below the surface. Incorporating these depth values into the pressure/depth equation for Hanford Site boreholes in Early et al. (1983, p. I-185):

$$P(\text{MPa}) = 0.063 + 0.0096 \text{ depth (m)} ,$$

values of pressure appropriate to the previously determined depths (and temperatures) result. By linear interpolation of the data presented in Culberson and McKetta, Jr. (1951) for these pressure and temperature values, an estimate of methane solubility in water, customized to the subsurface pressure/temperature conditions at the Hanford Site, can be made. Figure A-1 is a graph showing these relationships. The curve is eye-approximated as the uncertainties associated with the pressure and temperature equations given above probably are on the order of several percent and do not justify a more sophisticated curve-fitting procedure.

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Figure A-1. Methane Concentrations in Groundwaters from Selected Boreholes (Data taken from table 1).

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APPENDIX B

Letter from G. S. Mack to R. W. Bryce

Internal Letter

Rockwell International

Date December 2, 1983

No. 10120-83-346

TO Name (Last, First, Middle Initial) Address

R. W. Bryce

FROM Name Organization Internal Address Phone

G. S. Mack

Subject: Gas Measurements for DC-19C Umtanum Flow Top and
Cohasset Flow Top

Gas flow data were collected at borehole DC-19C within both the Cohasset flow top (3,008 to 3,105 feet) and the Umtanum flow top (3,586 to 3,567 feet). The samples were collected on November 10, 1983, and November 6, 1983, respectively.

Gas flow is measured with a monostat Calcuflow flowmeter using air at STP as a standard. The flow measurement must be corrected for the difference in density between the gas found at DC-19C and air. To do this, the viscosity and conditions were assumed to be similar to standard conditions.

$$V_{\text{gas}} = V_{\text{air}} \left(\frac{0.00120}{\rho_{\text{gas}}} \right)^{1/2}$$

V_{gas} = volume rate for gas

V_{air} = volume rate for air obtained from standard tables and measurement

ρ_{gas} = density of the gas

0.00120 = density of air at STP in g/cm³

COHASSETT FLOW TOP:

Mole Fraction	g/mole	g	Weight %
48% CH ₄	16.043	7.70	35% CH ₄
49% N ₂	28.013	13.73	62% N ₂
2% CO	28.011	0.56	3% CO
		21.99	

$$\begin{aligned} \rho_{\text{gas}} &= \rho_{\text{CH}_4} (\% \text{ CH}_4) + \rho_{\text{N}_2} (\% \text{ N}_2) + \rho_{\text{CO}} (\% \text{ CO}) \\ &= (0.000653) (0.35) + (0.001160) (0.62) + (0.001163) (0.03) \end{aligned}$$

$$\rho_{\text{gas}} = 9.83 \times 10^{-4} \text{ g/cm}^3$$

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$$V_{\text{gas}} = V_{\text{air}} \left(\frac{0.00120}{\rho_{\text{gas}}} \right)^{1/2} = 2678 \frac{\text{ml}}{\text{min}} \text{ air} \left(\frac{0.00120}{9.83 \times 10^{-4}} \right)^{1/2}$$

$$V_{\text{gas}} = 3000 \frac{\text{ml}}{\text{min}} \text{ gas}$$

For CH_4 :

$$V_{\text{CH}_4} = (\% \text{CH}_4)(V_{\text{gas}}) = (0.48)(3000 \frac{\text{ml}}{\text{min}} \text{ gas}) = 1440 \text{ ml/min CH}_4$$

P = pressure of the gas

P = Patm - PH₂O

$$= (28.923 \text{ in Hg}) \left(\frac{25.4 \text{ mm}}{1 \text{ in}} \right) - 26.739 \text{ mm Hg}$$

P = 707.9 mm Hg

Assume Ideal Gas Conditions:

For one minute

$$n_{\text{CH}_4} = \frac{PV}{RT} = \frac{(707.9 \text{ mm Hg}) (1.44 \text{ L})}{\left(62.4 \frac{\text{mm Hg L}}{\text{mole}} \right) (300.16 \text{ K})} = 0.054 \text{ moles CH}_4 \left(\frac{1.6 \times 10^3 \text{ mg}}{\text{mole}} \right) = 873 \text{ mg CH}_4$$

For the Liquid Flow $(16.7 \frac{\text{gal}}{\text{min}})$ in one minute:

$$(16.7 \text{ gal}) \left(\frac{3.785 \text{ L}}{1 \text{ gal}} \right) = 63.21 \text{ L}$$

Concentration of CH_4 :

$$\frac{873 \text{ mg CH}_4}{63.21 \text{ L soln}} = \frac{13.8 \text{ mg CH}_4}{\text{L soln}} \approx 14 \text{ ppm CH}_4$$

For N_2 :

$$V_{\text{N}_2} = (\% \text{N}_2) V_{\text{gas}} = (0.49) (3000 \frac{\text{ml}}{\text{min}} \text{ gas}) = 1.47 \text{ L}$$

For One Minute:

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$$n_{N_2} = \frac{PV}{RT} = \frac{(707.9 \text{ mm Hg}) (1.47 \text{ L})}{(62.4 \frac{\text{mm Hg L}}{\text{O}_k \text{ mole}}) (300.16 \text{ O}_k)} = 0.056 \text{ mole } N_2 \left(\frac{28.013 \times 10^3 \text{ mg}}{1 \text{ mole}} \right) = 1560 \text{ mg } N_2$$

Concentration of N_2 :

$$\frac{n_{N_2}}{\text{flow soln}} = \frac{1560 \text{ mg } N_2}{63.21 \text{ L}} = 24.6 \frac{\text{mg } N_2}{\text{L}} \text{ or } 25 \text{ ppm } N_2$$

Umtanum Flow Top:

Mole Fraction	g/Mole	g	Weight Percent
56% CH_4	16.043	8.98	43% CH_4
41% N_2	28.013	11.49	55% N_2
0.98% O_2	31.999	0.31	1% O_2
0.9% CO	28.011	0.25	1% CO
99%		21.03 gm	100%

$$\begin{aligned} \rho_{\text{gas}} &= (\%CH_4)\rho_{CH_4} + (\%N_2)\rho_{N_2} + (\%O_2)\rho_{O_2} + (\%CO)\rho_{CO} \\ &= (0.43)(0.0006653) + (0.55)(0.001160) + (0.01)(0.001326) + (0.01)(0.001163) \\ \rho_{\text{gas}} &= 9.49 \times 10^{-4} \text{ g/cm}^3 \text{ @ STP} \end{aligned}$$

$$V_{\text{gas}} = V_{\text{air}} \left(\frac{\rho_{\text{air}}}{\rho_{\text{gas}}} \right)^{1/2} = (4453 \frac{\text{ml air}}{\text{min}}) \left(\frac{0.00120}{9.49 \times 10^{-4}} \right)^{1/2} = 5007 \frac{\text{ml gas}}{\text{min}}$$

Pressure:

$$P_{\text{gas}} = P_{\text{atm}} - P_{H_2O} = [(29.161 \text{ in Hg}) \left(\frac{25.4 \text{ mm}}{\text{in}} \right) - 20.690 \text{ mm Hg}] = 720.00 \text{ mm Hg}$$

For CH_4 :

$$V_{CH_4} = (\%CH_4)V_{\text{gas}} = (0.56)(5007 \frac{\text{ml gas}}{\text{min}}) = 2800 \frac{\text{ml } CH_4}{\text{min}}$$

For One Minute:

$$n_{CH_4} = \frac{PV}{RT} = \frac{(720.0 \text{ mm Hg}) (2.800 \text{ L})}{(62.4 \frac{\text{mm Hg L}}{\text{O}_k \text{ mole}}) (295.86 \text{ O}_k)} = 0.109 \text{ mole} \left(\frac{16.043 \times 10^3 \text{ mg}}{1 \text{ mole}} \right) = 1750 \text{ mg}$$

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Volume of Solution:

$$(6.0 \frac{\text{gal-soln}}{\text{min}}) (\frac{1-\text{min}}{1}) (\frac{3.785 \text{ l}}{1 \text{ gal}}) = 22.7 \text{ l-soln}$$

Concentration:

$$\frac{n_{\text{CH}_4}}{\text{Vol of soln}} = \frac{1750 \text{ mg}}{22.7 \text{ l-soln}} = 77.2 \frac{\text{mg}}{\text{l}} \text{ CH}_4 \approx 77 \text{ ppm CH}_4$$

For N₂:

$$V_{\text{N}_2} = (\% \text{N}_2) V_{\text{gas}} = (0.41)(5007 \frac{\text{ml-gas}}{\text{min}}) = 2053 \frac{\text{ml-N}_2}{\text{min}}$$

For One Minute:

$$n = \frac{PV}{RT} = \frac{(720.0 \text{ mm Hg})(2053 \text{ l})}{(62.4 \frac{\text{mm Hg l}}{\text{O}_k \text{ mole}})(295.86 \text{ O}_k)} = 0.0801 \text{ mole} (\frac{28.0134 \times 10^3 \text{ mg}}{1 \text{ mole}}) = 2240 \text{ mg}$$

Concentration:

$$\frac{n_{\text{N}_2}}{\text{Vol of soln}} = \frac{2240 \text{ mg}}{22.7 \text{ l}} = 98.8 \frac{\text{mg}}{\text{l}} \text{ N}_2 \approx 99 \text{ ppm N}_2$$

Summary of Table:

Cohassett Flow Top

gas	Measured Mole Fraction	Calculated Concentration ppm
CH ₄	0.48	14
N ₂	0.49	25
CO	.02	

Umtanum Flow Top

gas	Measured Mole Fraction	Calculated Concentration ppm
CH ₄	.56	77
N ₂	.41	99
CO	.009	
O ₂	.0098	

Gregory Mack
Gregory Mack
Hydrologist

cc: S. M. Baker D. L. Graham W. H. Price S. R. Strait *MS*