Densities of Liquid $CH_{4-a}X_a$ (X=Br,I) and $CH_{4-(a+b+c+d)}F_aCl_bBr_cI_d$ Halomethanes

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The available density data for the air-saturated liquid and for the liquid at its saturation vapor pressure have been critically reviewed and the "best" data selected for the following halomethanes: CHF_2Cl, $CHFCl_2$, CF_3Cl, CF_3Br, CF_2Cl_2, $CFCl_3$, (data up to the critical point are available for these compounds); CH_3Br, CH_2Br_2, $CHBr_3$, CBr_4, CH_3I, CH_2I_2, CH_2FCl, (data up to the normal boiling point are available for these compounds); $CHCl_2Br$ (data over a small temperature range); CHI_3, CI_4, CH_2FI, CH_2ClBr, CH_2ClI, CHF_2Br, CHF_2I, $CHFBr_2$, $CHFI_2$, $CHCl_2I$, $CHClBr_2$, $CHClI_2$, CF_3I, CF_2Br_2, $CFBr_3$, CCl_3Br, CCl_3I, (few scattered data points are available for these compounds). The literature survey is complete up to June 1974. Selection of "best" data was arrived at by carefully evaluating each set of data for its accuracy, method of experimentation, sample purity, etc. The selection procedure is discussed. The uncertainty in the selected values is reported. For interpolation and limited extrapolation, the selected density data for each compound have been correlated through simple equations in temperature. The regression errors and the computed regression constants are reported in tables 2 and 4, respectively.

Key words: Halomethanes; liquid density; critically evaluated data.

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1. Introduction

1.1. Scope and Objectives

As part of our work on the physical and thermodynamic properties of haloalkanes (F, Cl, Br, I substituted), the densities of liquid halomethanes ($CH_{4-a}X_a$(X=Br,I) and $CH_{4-(a+b+c+d)}F_aCl_bBr_cI_d$) from the triple point to the critical point have been compiled, critically evaluated, and selected. The literature survey covered the period 1908 to June 1974, and the data were obtained, in a large majority of the cases, from the original literature. Only in the case of obscure and inaccessible publications was it necessary to depend wholly on the Chemical Abstracts.

Literature data are available for the air saturated liquid, denoted by symbol L (henceforth referred to as liquid), as well as for the liquid at its saturation vapor pressure, denoted by symbol SL (henceforth referred to as saturated liquid), both of which are compiled in the present work. A survey of sixty one halomethanes studied shows that density data for liquid and saturated liquid are available for thirty one compounds (table 1). Out of these, reliable experimental data up to the critical point are available for only six compounds. Reliable experimental data up to the normal boiling point are available for only seven compounds. For eighteen compounds only one or two data points are available and they are in general unreliable. The status of the density data is reported in table 1.

Many authors have measured densities at selected temperatures, usually around the ambient temperature, primarily in the process of synthesizing and identifying the compound. Others have carried out extensive and accurate density measurements over a wide temperature range on very pure samples in order to provide reliable data for process design calculations and for some theoretical studies.

Among the earlier workers involved in carrying out accurate density measurements, Timmermans and his co-workers, in the late 1920's and early 1930's, published reliable data for a number of these substances [1][1]. However, the data are reported over a limited temperature range. The subsequent quarter century has been a rather lean period. Only as late as 1960 did good experimental values again begin to appear in the literature. Most of the authors did not furnish "complete" experimental details such as sample purity, method of determination, or accuracy of measurements. This has considerably complicated proper selection of "good" data. In general, it is only recently that authors report pertinent information necessary for a critical evaluation of these data, thanks to the publication policies of many journals.

The Thermodynamics Research Center (henceforth referred to as TRC) [2] reports critically selected data at 20 and 25 °C for some of these compounds. The American Society of Heating, Refrigerating and Air Conditioning Engineers (ASHRAE) [3] also reports density data for some of these compounds.

1.2. Temperature Scale and Conversion Factors

It was observed during the compilation that the temperature scales used by different investigators were not quite identical. For all our reported temperatures we have strictly adhered to the International Practical Temperature Scale of 1968, IPTS-1968 [4]. Temperatures reported on the basis of IPTS-1948 and other temperature scales have accordingly been corrected to conform to the IPTS-1968 scale using the procedures of TRC [2]. Density values reported in other units have been converted to the c.g.s. units so that recorded densities are in g cm^{-3} and all temperatures in degrees Celsius. The conversion factors used are given below.

1 lb=453.59237 g
1 ml=1 cm^3 (exactly)
1 in=2.54 cm
1 g cm^{-3}=62.42795 lb ft^{-3}
1 g ml^{-1}=1 g cm^{-3}

1.3. Evaluation and Selection Procedures

Adequate accessory information was available for very few compounds to facilitate assessment and data selection. Table 1 presents literature information (or lack thereof) for 61 species. Complete literature references for many of these compounds are available in the TRC publications [2]. Those references from which data were selected for final analysis are boldface in table 1.

In the analysis, the work of each investigator was carefully scrutinized to ascertain the precision and accuracy of the actual measurements of density and temperature. The following points were considered in analyzing and evaluating the data.

1. Source, method of purification, and the purity of the sample.
2. Method of measurement and calibration of the measuring instruments.
3. Accuracy of parameter measurements, i.e., temperature, weight, liquid level, density of calibrating fluid, etc.
4. Computed accuracy in the final density value.
5. Reproducibility of results.
6. Purpose of measurement.
7. Number and closeness of data points.
8. Range of investigated temperature.
9. Reputation of the authors.

A general evaluation methodology for the method of purification is not possible since the merit of a purification process is a function of the particular substance and the nature of impurities present. In some cases however, commerical samples were used and no details for these were available. Besides the numerical purity values, the source and/or method of synthesis of the sample, and the number and kinds of purification procedures employed were also studied in order to assess their effect on the purity of the substance.

For the more common halomethanes, several sets of numerical values were invariably available. Initial screening on the basis of accuracy of measurement and purity of the sample usually eliminated some of the values. Thereafter, some of the other criteria above were used to eliminate some more

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[1] Figures in brackets indicate literature references at the end of this paper.

TABLE 1. Status of density data for halomethanes

Symbols: A=Adequate and good experimental data available up to the critical point.

A*=Adequate and good experimental data available up to the normal boiling point.

B=Inadequate and scanty data (need further experimentation).

C=Few data points (need further experimentation).

Note: Data from the boldface references are the selected data. References in parentheses contain data for the saturated liquid.

Serial No.	Compound	Status	References
1.	CH_3Br	A*	6, 7, **8**, **(9)**
2.	CH_2Br_2	A*	6, 10, ***11***, **12**, 13, 14, **15**, 16, 17, 18, **19**, 20
3.	$CHBr_3$	A*	6, 13, **16**, 18, **19**, 20, **21**, 22, 23, 24, 25, 26, 27, 28, 29, 30, **31**, 32, **33**, 34, 35
4.	CBr_4	A*	**19**
5.	CH_3I	A*	7, **8**, **36**, 37, **38**, 39, 40
6.	CH_2I_2	A*	6, 11, **12**, 13, 14, **15**, 18, 20, 29, 41, 42
7.	CHI_3	C	6, 43
8.	CI_4	C	6, 43, **44**
9.	CH_2FCl	A*	**45**
10.	CH_2FBr		
11.	CH_2FI	C	**13, 46**
12.	CH_2ClBr	C	**13, 47**
13.	CH_2ClI	C	**13**
14.	CH_2BrI		
15.	CHF_2Cl	A	(3), **(48)**
16.	CHF_2Br	C	**49**
17.	CHF_2I	C	**50**
18.	$CHFCl_2$	A	13, (3), **(48)**
19.	$CHFClBr$		
20.	$CHFClI$		
21.	$CHFBr_2$	C	**13**, 49
22.	$CHFBrI$		
23.	$CHFI_2$	C	**50**
24.	$CHCl_2Br$	B	13, **21, 51**
25.	$CHCl_2I$	C	**13, 52**
26.	$CHClBr_2$	C	**13**, 53
27.	$CHClBrI$		
28.	$CHClI_2$	C	54
29.	$CHBr_2I$		
30.	$CHBrI_2$		
31.	CF_3Cl	A	55, (56), **(57)**
32.	CF_3Br	A	**(7)**
33.	CF_3I	C	7, 58
34.	CF_2Cl_2	A	*59, (60), (61),* ***(62)***
35.	CF_2ClBr		
36.	CF_2ClI		
37.	CF_2Br_2	C	**63**
38.	CF_2BrI		
39.	CF_2I_2		
40.	$CFCl_3$	A	13, 26, 59, **(48)**, **(64)**
41.	$CFCl_2Br$		
42.	$CFCl_2I$		
43.	$CFClBr_2$		
44.	$CFClBrI$		
45.	$CFClI_2$		
46.	$CFBr_3$	C	13, 26, **63**
47.	$CFBr_2I$		
48.	$CFBrI_2$		
49.	CFI_3		
50.	CCl_3Br	C	**13, 65**
51.	CCl_3I	C	**13**
52.	CCl_2Br_2		
53.	CCl_2BrI		
54.	CCl_2I_2		
55.	$CClBr_3$		
56.	$CClBr_2I$		
57.	$CClBrI_2$		
58.	$CClI_3$		
59.	CBr_3I		
60.	CBr_2I_2		
61.	$CBrI_3$		

values and to assign additional weight to certain highly reliable data points. While quantitative information on the accuracy of measurement, sample purity, etc., was always sought, in some cases it was not available. It was then necessary to evaluate the data of different investigators on an empirical rating of the purification process and the method of measurement used. In order to develop a rating of the measurement techniques, we relied primarily on the information provided by Weissberger [5]. To some extent, an investigator's past reputation did influence the weight assigned to his data.

The emphasis has been to select density as a function of temperature for the liquid and the saturated liquid. Figure 1 depicts the typical temperature behavior of the saturated liquid density from the triple point to the critical point. After preliminary screening the available data were carefully analyzed to see the general trend over the entire temperature range and to check if each selected data point "fitted" with the other points in representing a systematic behavior. In certain cases where there was some doubt about the consistency of the data, density versus temperature plots were prepared to help the selection of "best" data. In order to smooth the data and to check gross inconsistencies among the data points, the following mathematical models were chosen to fit the data. Linear least squares regression on an IBM 7044 computer at IIT-Kanpur was used in each case.

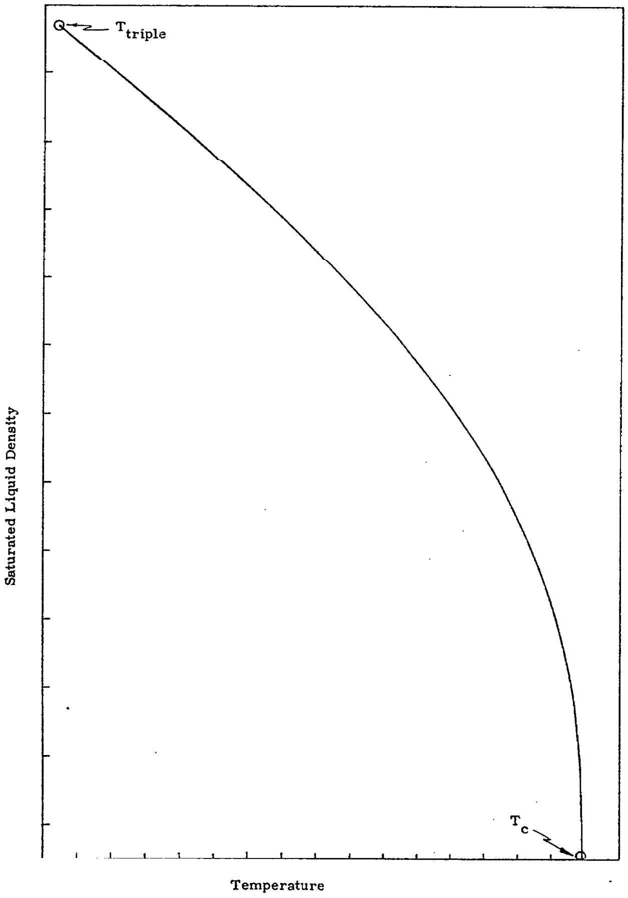

FIGURE 1. Saturated liquid density versus temperature (typical curve).

1. For data up to the boiling point,

$$d=a+bT+cT^2+dT^3+\ldots \quad (1)$$

2. For data up to the critical point,

$$d-d_c=A(1-Tr)^{1/3}+B(1-T_r)^{2/3}+C(1-T_r)^{3/3}+D(1-T_r)^{4/3}+\ldots (2)$$

Here $T_r=T/T_c$ and T_c is the critical temperature in K. Depending upon the accuracy of the experimental data, the average and maximum errors tolerable in regression were normally fixed at ±0.0001 g cm^{-3} and ±0.0005 g cm^{-3}. The final selected density values in the form of an equation were assigned an uncertainty based upon the "maximum error" in the final regression. Whenever available, the values of Timmermans and co-workers, and of Dreisbach and Martin are recommended as the "best" values for the stated temperature range.

An explanation of the errors referred to is necessary. In this report, two measures of uncertainty are discussed:

(a) The estimated errors in the propery measurements (experimental errors), based upon our assessment of the experimental techniques described in each paper.

(b) The statistical measures of error resulting from the regressions between observed and calculated densities.

Experimental errors in the density and temperature measurements are discussed whenever available under each compound. In the regressions we have used the standard error and average absolute error rather than the simple average error, since this eliminates the possibility of large positive and negative errors nullifying each other to give a misleading representation. The errors used in the regressions are defined below:

$$\text{Average absolute error} = \frac{\Sigma\text{Individual absolute errors}}{\text{No. of data points}} \quad (3)$$

$$\text{Standard error} = \left[\frac{\Sigma(\text{Individual errors})^2}{\text{No. of data points}}\right]^{1/2}. \quad (4)$$

The average absolute error served primarily to gauge the merit of the regression in its entirety.

The following regression procedure was used in arriving at the coefficients listed in table 4.

1. Average and maximum tolerable errors in data were fixed based upon uncertainties in the experimental values.
2. Each data point was assigned a weight based upon the experimental uncertainty.
3. A regression was peformed.
4. Data points giving deviations larger than the maximum error were suspect, checked again for their reliability, and in most cases eliminated.
5. Error distribution was checked. In some regressions errors were all positive though the regression was "good."
6. Steps 2 through 5 were repeated as necessary.

The coefficients reported in table 4 are based on the least squares fit.

In all cases only the experimental density values were used for the regression analysis. The TRC selected values and those reported in other compilations were not used in the regressions.

Table 2 gives the summary of regression errors for each compound. To illustrate the reliability of regression, the number of data points regressed is also included in each case.

The critical constants used in the computations are reported in table 3. The regression constants for each compound are reported in table 4.

In general the difference in density for the air saturated liquid and for the liquid at its saturation vapor pressure for these halomethanes is of the order of 5×10^{-5} g cm^{-3}. At lower temperatures the air solubility effect is predominant, but the vapor pressures are very small. This situation is reversed at higher temperatures. These two factors have opposing effect on the liquid density. Hence the overall effect is negligible especially when we are dealing with the density values good to ±0.001 to ±0.0001 g cm^{-3}.

The detailed discussion for each compound follows.

TABLE 2. *Summary of regression errors in densities of halomethanes*

Equation 1: $d=A+BT+CT^2$

1a: $d=A+BT+CT^2+DT^3$

2: $d-d_c=A(1-T_r)^{1/3}+B(1-T_r)^{2/3}+C(1-T_r)+D(1-T_r)^{4/3}$

Compound and state		Equation number	Average error × 10^4	Maximum error × 10^4	Standard error × 10^4	Number of points
CH_3Br	(SL)	1	12.0	25.2	3.38	17
CH_2Br_2	(L)	1	3.21	8.64	1.21	12
$CHBr_3$	(L)	1	4.08	−8.99	0.835	31
CBr_4	(L)	1a	2.33	6.3	3.04	6
CH_3I	(L)	1	1.51	−5.53	0.47	10
CH_3I	(L)	1	2.30	−5.38	0.785	12
CH_2I_2	(L)	1	1.88	−5.37	0.520	16
CH_2FCl	(SL)	1a	0.76	−2.52	0.386	8
CHF_2Cl	(SL)	2	4.48	−7.95	1.35	14
$CHFCl_2$	(SL)	2	2.69	−7.78	0.799	20
CF_3Cl	(SL)	2	10.9	−25.4	4.40	11
CF_3Br	(SL)	2	10.7	−48.0	15.6	25
CF_2Cl_2	(SL)	2	17.9	−48.7	5.21	18
$CFCl_3$	(SL)	2	2.10	5.20	0.617	16

TABLE 3. *Critical constants of mixed halomethanes* [66]

Compound	T_c, K	d_c, g cm^{-3}
CHF_2Cl	369.2	0.525
$CHFCl_2$	451.6	0.522
CF_3Cl	302.0	0.577[a]
CF_3Br	340.2	0.760
CF_2Cl_2	384.95	0.558
$CFCl_3$	471.2	0.554

[a] See text.

TABLE 4. Summary of regression constants for the densities of liquid halomethanes
Equation 1: $d=A+BT+CT^2$
1a: $d=A+BT+CT^2+DT^2$
2: $d-d_c=A(1-T_r)^{1/3}+B(1-T_r)^{2/3}+C(1-T_r)+D(1-T)^{4/3}$

Compound and state	Equation number	Empirical constants				Temperature range, °C
		A	$-B\times10^3$	$-C\times10^4$	$-D\times10^4$	
CH_3Br (SL)	1	2.3232	1.7037	0.01722		−95 to +50
CH_2Br_2 (L)	1	3.1840	2.0759	0.009142		0 to 100
$CHBr_3$ (L)	1	3.5603	1.9668	0.01080		10 to 125
CBr_4 (L)	1a	−1.79705	−34.5153	+0.78002	-0.5261×10^{-3}	100 to 190
CH_3I (L)	1	+2.9124	+1.4918	0.02279		0 to 40
CH_3I (L)	1	3.1196	2.9934	−0.00423		−70 to 0
CH_2I_2 (L)	1	4.2314	3.5538	−0.01531		+15 to 120
CH_2FCl (SL)	1a	2.0156	4.0559	−0.08306	0.136×10^{-3}	−80 to +40
CHF_2Cl (SL)	2	0.93746	−220.82	−3382.1	+967.09	−70 to +80
$CHFCl_2$ (SL)	2	0.77124	−1174.6	+14790.	−9977.9	−65 to +160
CF_3Cl (SL)	2	0.93484	−810.88	9063.28	−7357.8	−140 to +25
CF_3Br (SL)	2	1.0978	−1442.6	15083.	−9892.5	− 90 to +67
CF_2Cl_2 (SL)[a]	2					−122 to +110
$CFCl_3$ (SL)	2	0.92841	−629.42	+5233.5	−4814.0	−40 to +185

[a] $d-d_c=1.4016(1-T_r)^{1/3}-2.8977(1-T_r)^{2/3}+9.0828(1-T_r)-10.689(1-T_r)^{4/3}+4.701(1-T_r)^{5/3}$.

2. Bromomethane (CH_3Br)

The liquid density values at 0 °C and 4.5 °C are reported by Biltz et al. [6] and by Nodiff et al. [7]. Morgan and Lowry [8] reported dilatometric density data from −93.6 to +7.4 °C on a carefully purified sample.

Hsia [9] in 1931, reported saturated density data for the temperature range, −50 to +50 °C; he used a purified sample obtained by fractionation and vacuum distillation and reported an experimental accuracy of ±0.005 g cm^{-3} in the density measurements. The data, however, are given at 10 °C intervals, which suggests interpolation.

Both sets of data do not appear to be of high accuracy. However, these are selected as the "best" available for further analysis. The quadratic model (table 4) is recommended which gives an absolute average deviation of ±0.001 g cm^{-3}. The calculated values are assigned an uncertainty of ±0.002 g cm^{-3} and are compared satisfactorily with the TRC values below:

	d, g cm^{-3}		
t, °C	Calculated	TRC	Hsia
20	1.6758	1.6755	1.6769
25	1.6622	1.6617	
30	1.6485		1.6489

3. Dibromomethane (CH_2Br_2)

No data are available for the saturated liquid. For the liquid in air, a few data points per author are available from a number of authors [6, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20]. Some have provided supporting details. Mohanti and Dasgupta [11] report a density of 1.525 g cm^{-3} at 24.84 °C by pycnometric measurement on a Kahlbaum sample. Smyth and Rogers [18] used an Eastman Kodak sample purified by washing with dilute sodium carbonate solution and water, dried, and distilled at reduced pressure. The data measured by a pycnometer with an accuracy of ±0.0008 g cm^{-3} are reported below:

t, °C	d, g cm^{-3}
10	2.5438
20	2.4921
40	2.4442

A very low temperature (−196 °C) value of 2.49 g cm^{-3} is available from Biltz et al. [6]. In 1943, Friend and Hargreaves [19] reported data from 18.6 to 97.2 °C, using the specific gravity bottle method, for a distilled sample. Their data were selected in addition to the values of Smyth and Rogers, Timmermans and Hennaut-Roland [12], and Griffing et al. [15]. The latter's data came from the balance measurements on a 98 mol % pure sample covering the range 0 to 80 °C. Thus the selected data covered a range from 0 to 95 °C. Timmermans data at 0, 15, 30 °C are the "best" available and are recommended.

t, °C	d, g cm^{-3}
0	2.54852
15	2.50986
30	2.47133

Linear interpolation of these data yield 2.4970±0.0001 g cm^{-3} and 2.4842±0.0001 g cm^{-3} for 20 and 25 °C respectively. TRC has also recommended the above values at 20 and 25 °C.

In order to represent the data as a function of temperature up to 95 °C (normal boiling point is 96.95 °C) the above data along with those of Friend and Hargreaves, and Griffing et al. were regressed to polynomials. Because of their higher accuracy, Timmermans data were weighted five times. However, the results were not satisfactory (absolute av. dev. = ±0.0012 g cm^{-3}). Smyth and Rogers' value at 10 °C deviated considerably and was neglected in further calculations.

Subsequently the data set was subdivided into the Friend and Hargreaves subset and Griffing et al. subset. Because of their inherent reliability, Timmermans' values, still weighted, were retained in both subsets. However, no improvement in correlation resulted (absolute av. dev. = ±0.0012 g cm^{-3}). It appears that the regressed data are mutually inconsistent. Our final reported results are based on a non-weighted regression of all four sets of measurements. The absolute average error of 0.0003 g cm^{-3} is acceptable. At 15 °C, the calculated density of 2.5099 g cm^{-3} is in excellent agreement with Timmermans' value of 2.50986 g cm^{-3}. Likewise the values at 20 and 25 °C are in consonance with those reported by TRC:

	d, g cm^{-3}		
t, °C	Calculated	Timmermans	TRC
15	2.5099	2.50986	
20	2.4969		2.4970
25	2.4838		2.4842
30	2.4707	2.47133	

The figures quoted pertain to the quadratic equation (table 4) and the computed constants are recommended for predicting values from 0 to 95 °C with an uncertainty of ±0.0008 g cm^{-3} in the calculated value. As stated before, Timmermans' values are recommended from 0 to 30 °C with linear interpolation to calculate the values within this range with an uncertainty of ±0.00005 g cm^{-3}.

4. Tribromomethane ($CHBr_3$)

Saturated liquid densities are totally absent in the published literature. The liquid densities are available from the following references [6, 13, 16, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28 to 35]. Pascal [30] reported the following values from 0 to 90 °C without giving any details:

t, °C	d, g cm^{-3}
0	2.9315
17.5	2.8827
19	2.8779
50	2.7934
70	2.7364
90	2.6823

Smyth and Rogers [18] measured liquid density at 10, 40 and 70 °C using a pycnometer on an Eastman Kodak sample purified by washing with dilute sulfuric acid, followed by sodium hydroxide and water, dried, and distilled at reduced pressure. The data given below are reported accurate to ±0.0008 g cm^{-3}:

t, °C	d, g cm^{-3}
10	2.9189
40	2.8383
70	2.7597

Trew [34] used "purest form" of the sample purified by fractional distillation until a constant boiling fraction was obtained. A 2 cm^3-capacity pycnometer was used and density at 25 °C was reported to be 2.8794 g cm^{-3} which is an average of three readings. Timmermans and co-workers [21, 23] presented the following values:

t, °C	d, g cm^{-3}
15	2.9035
30	2.8646

Desreux [26] reported 2.8905 g cm^{-3} at 20 °C, but gave no details. Sherman and Sherman [31] published accurate data from 9 to 50 °C using a pycnometer and a sample purified by washing with sodium carbonate and water, drying, and distilling three times at 20 mmHg. They represented their data in the following manner:

$$v(\mathrm{cm^3 g^{-1}})=0.34204\,[1+0.90411\times10^{-3}(t-7.7)+0.6766\times10^{-6}(t-7.7)^2], \qquad (5)$$

t in °C.

Equation (5) resulted in the following density values:

t, °C	v, cm^{3}g^{-1}	d, g cm^{-3}
10	0.34275	2.9176
15	0.34431	2.9044
20	0.34588	2.8912
25	0.34746	2.8780
30	0.34905	2.8649
40	0.35227	2.8353
50	0.35554	2.8127

In the intervening period, until the work of Friend and Hargreaves [19], several single point values had been published, presumably with the main intention of sample identification. Not unexpectedly, therefore, most of these measurements have been at standard temperatures such as 20 °C and 30 °C. Friend and Hargreaves covered a higher temperature range, from 16.3 to 141.0 °C, through specific gravity measurements on a distilled sample. Lagemann et al. [16] using an Eastman Kodak white label sample, also reported accurate pycnometric data from 0 to 50 °C, but at 10° intervals.

The data of Timmermans were selected as the most accurate. In order to cover a wider range of temperature, the data of Sherman and Sherman, Friend and Hargreaves, and Lagemann were selected because of the overall agreement with the Timmermans values at the overlapping temperatures. The regression results for both quadratic and cubit fit were not satisfactory indicating non-compatability of these different sets of values. In spite of weighting Timmermans data five times, the deviations between calculated and Timmermans experimental values ranged from 0.0002 to 0.0004 g cm^{-3}. Various adjustments were made, but the results could not be improved to match Timmermans' values. Finally, the quadratic model (table 4) was chosen which gave an absolute average deviation of ±0.0004 g cm^{-3}. The calculated densities are compared below.

	d, g cm^{-3}		
t, °C	Present	Timmermans	TRC
15	2.9039	2.9035	
20	2.8909		2.8889
25	2.8779		2.8758
30	2.8648	2.8646	

The agreement with the TRC values is not satisfactory. The quadratic model is recommended and an uncertainty in the calculated value of ±0.0005 g cm^{-3} is assigned from 10 to 30 °C, and ±0.001 g cm^{-3} from 30 to about 135 °C.

5. Tetrabromomethane (CBr_4)

Friend and Hargreaves [19] reported liquid density from 100.7 to 190.0 °C (freezing point of CBr_4 is 91.0 °C (2)). Their values, given below, are based on the specific gravity measurements on a distilled sample:

t, °C	d, g cm^{-3}
100.7	2.9533
124.9	2.9004
139.2	2.8609
147.5	2.8355
155.2	2.8094
163.1	2.7831

These are the only reliable data available and are selected. These data were fitted satisfactorily to a cubic model with an absolute average deviation of ±0.0002 g cm^{-3}. The regression constants reported in table 4 are recommended for calculating the density data from 100 to 185 °C with an uncertainty of ±0.0006 g cm^{-3} in the value.

6. Iodomethane (CH_3I)

Morgan and Lowry [8] reported liquid densities from −70 to +40 °C obtained using a dilatometer. As in several earlier compounds, the reported values have been interpolated at 10° intervals. The most accurate data are from Timmermans and Delcourt [36] at 0, 15, and 30 °C:

t, °C	*d*, g cm^{-3} [36]
0	2.33504
15	2.29300
30	2.25102

Vogel [37] in 1943 reported two sets of density values for differently purified samples using the pycnometer method. The two sets differed from each other by about ±0.001 g cm^{-3}. This method has also been employed by Nodiff et al. [7] who reported 2.2174 g cm^{-3} at 42.50 °C, and by Moelwyn-Hughes and Missen [38] who reported 2.2645 g cm^{-3} at 25 °C. Bowden and Jones [39] report a density of 2.2146 g cm^{-3} at the normal boiling point, 42.43 °C. Zaalishvili and Kolysko [40] reported 2.2803 g cm^{-3} at 20 °C, but did not give any details.

Saturated liquid densities are not available for this compound.

For the temperature range, 0 to 30 °C, Timmermans values are selected as the best available. Linear interpolation of Timmermans data yield 2.2790 g cm^{-3} for 20 °C and 2.2650 g cm^{-3} for 25 °C. Zaalishvili and Kolysko value agrees within ±0.001 g cm^{-3} with the interpolated value at 20 °C and with the TRC [2] value of 2.2789 g cm^{-3}. The Moelwyn-Hughes and Missen value for 25 °C is selected.

For representing data over a slightly wider temperature range, i.e., 0 to 40 °C, the density data except those of Morgan and Lowry were regressed and the quadratic equation (table 4) with an absolute average deviation of ±0.0003 g cm^{-3} was selected. The calculated values are compared with selected experimental values and the TRC values below.

	d, g cm^{-3}			
t, °C	Calculated	Ref. [36]	Ref. [38]	TRC
15	2.2933	2.2930		
20	2.2792			2.2789
25	2.2650		2.2645	2.2649
30	2.2507	2.2510		

For the temperature range −65 to +40 °C covered by Morgan and Lowry and others, all these data were regressed to a quadratic model (table 4) with an absolute deviation of ±0.0008 g cm^{-3}.

Hence for the range −65 to +40 °C, the uncertainty in the calculated value is ±0.002 g cm^{-3}, and from 0 to 40 °C, it is ±0.0005 g cm^{-3}.

7. Diiodomethane (CH_2I_2)

No saturated liquid density data are available. For the liquid the data are reported in the following references (6, 11, 12, 13, 14, 15, 18, 20, 29, 41, 42). Smyth and Rogers [18] reported 3.3105 g cm^{-3} at 25 °C from the pycnometric measurements, while from the later work of Griffing et al. [15] a value of 3.3075 g cm^{-3} is available at the same temperature. Timmermans [12] has again provided accurate data as follows:

t, °C	*d*, g cm^{-3}
15	3.3345
20	3.3212
30	3.2944

Griffing et al. [15] used the balance method for 98 mol % pure sample and reported densities from 12.2 to 120 °C. Pycnometric data are also available from Grzeskowiak et al. [41] from 20 to 85 °C (four data points). The lower temperature values differ from Timmermans values, i.e., 3.3201 g cm^{-3} at 20 °C [41], and hence do not seem to be accurate enough for final selection. O'Connell [20] reported 3.3079 g cm^{-3} at 25 °C but did not give any details. Patterson and Thomson [42] reported the following data on a purified sample:

t, °C	*d*, g cm^{-3} [42]
19.41	3.32605
21.50	3.32138
26.75	3.3074
29.15	3.3010

The available data were plotted on large-scale plots and the data of Timmermans, and of Griffing et al., were selected for the final analysis. The data of Patterson and Thomson and of Grzeskowiak et al. are not quite compatible with the others. Since Griffing et al. data yielded larger errors and also because Timmermans' data are more reliable, certain values of Griffing et al. were rejected, primarily in the lower temperature range, which was still covered by Timmermans. This is a feature of all rejections for any compound. As far as possible it was ensured that rejection of a set of points did not introduce a large "temperature gap" where no data points were present. The quadratic model (table 4) is recommended with an uncertainty of ±0.0005 g cm^{-3} in the calculated value in the temperature range 15 to 60 °C, and ±0.001 between 60 to 120 °C. At 20 °C and 25 °C, the calculated densities of 3.3212 and 3.3079 g cm^{-3} agree very well with those of TRC [2].

8. Triiodomethane (CHI_3)

Very scanty data are available for this compound [6, 43]. Fruhwirth and Mayer - Pitsch [43] used an electrolytically prepared sample crystallized from alcohol. They report a liquid density of 1.6126 g cm^{-3} at 20 °C which appears to be incorrect. Hence the following TRC selected values are recommended for this compound (solid):

t, °C	d, g cm^{-3}
20	4.188
25	4.178

9. Tetraiodomethane (CI_4)

Very scanty data are available for this compound also [6, 43]. Lantenois [44] reported solid density of 4.50 g cm^{-3} at 0 °C and this is recommended with an uncertainty of ±0.05 g cm^{-3}.

10. Fluorochloromethane (CH_2FCl)

Only Phillips and Murphy [45] have reported density data that pertain to saturated liquid. For a 99.90 mol % pure sample, their nine data points span the range −80.95 to +41.40 °C. These are the only reliable data available and hence are selected. Polynomial regressions favor the cubic model (table 4), which represents density behavior over the regressed temperature range with an abosolute average error of 0.0001 g cm^{-3}. While this is admittedly indicative of a very good correlation, Phillips and Murphy's value of 1.3924 g cm^{-3} at −62.22 °C appears suspect in that it is away from the trend of the other values and was therefore removed (actual dev. of 0.0012 g cm^{-3} in the quadratic, and 0.0003 g cm^{-3} in the cubic models for this value). The cubic model (table 4) is recommended for calculation between −80 to 40 °C, and an uncertainty of ±0.0002 g cm^{-3} is assigned to the calculated value.

11, 12, 13. Fluoroiodomethane (CH_2FI), Chlorobromomethane (CH_2ClBr), Chloroiodomethane (CH_2ClI)

Altogether only five liquid density values are available for these compounds. For CH_2FI, 2.366 g cm^{-3} at 20 °C is reported by van Arkel and Janetzkey [46] and by Stevels [13].

For CH_2ClBr, Stevels [13] reports 1.944 g cm^{-3}, and Stevens et al. [47] 1.938 g cm^{-3}, at 20 °C. Hence at 20 °C, 1.941 g cm^{-3} is selected.

For CH_2ClI, Stevels [13] reports 2.422 g cm^{-3} at 20 °C.

The following values are recommended for these compounds:

Compound	t, °C	d, g cm^{-3}
CH_2FI	20	2.366±0.001
CH_2ClBr	20	1.941±0.001
CH_2ClI	20	2.422±0.001

14. Difluorochloromethane (CHF_2Cl)

Benning and McHarness [48] have reported density data for the saturated liquid on a very pure sample in the temperature range −69 to +87 °C, the measurements being made by a dilatometer. ASHRAE [3] have reported density values for the saturated liquid (99.90 mol % pure) from −72.49 to +25.97 °C and these appear to be experimental values. Neither of these two groups have mentioned the accuracy of the measurements. Data for liquid in air are not available.

Both sets of data for the saturated liquid were selected. These were regressed and fit equation (2) with an absolute average error of ±0.0004 g cm^{-3}. Equation (2) is recommended (table 4) for the temperature range −75 to +96.0 °C (t_c), and an uncertainty of ±0.0005 g cm^{-3} is assigned to the calculated value.

15. Difluorobromomethane (CHF_2Br)

Swarts [49] reported 4.55 g cm^{-3} at 15.7 °C. This is the only value available and is recommended with an uncertainty of ±0.01 g cm^{-3}.

16. Difluoroiodomethane (CHF_2I)

Ruff et al. [50] reported the following equation for density of liquid CHF_2I:

$$d(\text{g cm}^{-3})=3.401-0.00368\,T(\text{K}). \qquad (6)$$

No temperature range is reported. Density at 20 °C is calculated as 2.323 g cm^{-3} and is selected with an uncertainty of ±0.01 g cm^{-3}.

17. Fluorodichloromethane ($CHFCl_2$)

For the liquid, Stevels [13] reported 1.405 g cm^{-3} at 9 °C. This is the only value available.

For the saturated liquid two sets of data are available. Benning and McHarness [48] have measured, by dilatometric method, saturated liquid densities between −40 and +169 °C. Later data from ASHRAE [3] cover a smaller temperature region (−65.37 to +73.56 °C) for a 99.90 mol % pure sample, and these appear to be experimental values. These data were selected and processed further. Equation (2) represents both sets of data very well (absolute average error = ±0.0003 g cm^{-3}) and is recommended for the saturated liquid from −60 to +178.4 °C (t_c), assigning an uncertainty of ±0.0005 g cm^{-3} in the calculated value (table 4).

18. Fluorodibromomethane ($CHFBr_2$)

For $CHFBr_2$, Swarts [49] reports 2.4256 g cm^{-3} at 18.5 °C and Stevels [13], 2.421 at 20 °C. For 20 °C, 2.421±0.001 g cm^{-3} is recommended.

19. Fluorodiiodomethane ($CHFI_2$)

Ruff et al. [50] report the following liquid density values:

t, °C	d, g cm^{-3}
−19.4	3.3315
+14.4	3.2206
21.5	3.1969

They represented their data by the following model:

$$d(\text{g cm}^{-3})=4.165-0.003285\,T(\text{K}). \qquad (7)$$

This model is recommended from −20 to +30 °C with an uncertainty of ±0.001 g cm^{-3} in the calculated value.

20. Dichlorobromomethane ($CHCl_2Br$)

Timmermans and Martin [21, 51] reported three liquid density values at 0, 15 and 30 °C. An intermediate value of 1.980 g cm^{-3} at 20 °C have been reported by Stevels [13]. Because of their reliability, Timmermans and Martin's data are selected and recommended:

t, °C	d, g cm^{-3}
0	2.0385
15	2.0055
30	1.9724

Linear interpolation yields 1.9945 and 1.9835 g cm^{-3} at 20 °C and 25 °C. These values are recommended with an uncertainty of ±0.0002 g cm^{-3} in all values.

21. Dichloroiodomethane ($CHCl_2I$)

Stevels [13] reports 2.392 g cm^{-3} at 20 °C, and Hine and Dowell [52] report 2.3853 g cm^{-3} at 25 °C. Based upon these experimental values, the following values are recommended:

t, °C	d, g cm^{-3}
20	2.392±0.001
25	2.385±0.001

22. Chlorodibromomethane ($CHClBr_2$)

Stevels [13] has reported 2.451 g cm^{-3} at 20 °C. A value of 2.411 g cm^{-3} at 32 °C has been reported by Hine et al. [53]. Stevels' value is selected, assigning an uncertainty of ±0.001 g cm^{-3} in it, i.e., 2.451 ±0.001 g cm^{-3} at 20 °C.

23. Chlorodiiodomethane ($CHClI_2$)

Only a single value of 3.17 g cm^{-3} at 0 °C is reported by Auger [54]. Hence no selection is possible.

24. Trifluorochloromethane (CF_3Cl)

For the liquid in air, just two pycnometric values (at −130 °C and −82 °C) are available from Ruff and Keim [55]. Data for the saturated liquid have been reported by Reidel [56], and by Albright and Martin [57]. Reidel's values cover the range −140 to +25.10 °C, but lack experimental details. More detailed information has been furnished by Albright and Martin, whose float technique measurements on a 99.9 mol % pure sample yielded accurate and reliable data from −142.25 to +25.60 °C. They represented their data by the following equation:

$$d(\text{lb ft}^{-3})=36.07+0.01566(t_c-t)+1.110(t_c-t)^{1/2}$$
$$+6.665(t_c-t)^{1/3}+3.245\times10^{-5}(t_c-t)^{2} \qquad (8)$$

with an absolute average deviation of ±0.051 lb ft^{-3} (±0.0008 g cm^{-3}). Here d_c = 36.07 lb ft^{-3} and t, t_c are in °F. Converting density and temperature units to g cm^{-3} and K, respectively, eq (8) becomes:

$$d(\text{g cm}^{-3})=0.578+0.13636(1-T_r)+0.41456(1-T_r)^{1/2}$$
$$+0.87131(1-T_r)^{1/3}+0.15360(1-T_r)^{2}. \qquad (9)$$

The data of Albright and Martin have been selected as the most reliable. Regression to eq (2) with T_c = 302.0 K and d_c = 0.579 g cm^{-3} [66] yielded an absolute average error of ±0.002 g cm^{-3} which is believed to be larger than the experimental uncertainty. Adjustment of d_c to 0.578 g cm^{-3} reduced the absolute average error to ±0.0007 g cm^{-3}, which is almost identical to what Albright and Martin got by this equation.

Hence eq (2) (table 4) is recommended for the temperature range −140 °C to 28.8 °C (t_c) with an uncertainty of ±0.001 g cm^{-3} in the calculated value.

25. Trifluorobromomethane (CF_3Br)

Only saturated liquid density data are available for this compound. Nodiff et al [7] reported the following data:

t, °C	d, g cm^{-3}
−59	1.96
−54	1.94
−18	1.78

ASHRAE [3] published saturated liquid densities from −90 to +67 °C which are taken from "Freon-13 Bl, Tech, Bull. T-13 Bl, E.I. du Pont de Nemours & Co. (1963)". These data are recommended. Some selected data are reported below:

t, °C	T, K	d, g cm^{-3}
−90.0	183.15	2.1258
−80.0	193.15	2.0858
−70.0	203.15	2.0444
−60.0	213.15	2.0020
−57.75 (nbp)	215.40	1.9910
−50.0	223.15	1.9578
−40.0	233.15	1.9118
−30.0	243.15	1.8637
−20.0	253.15	1.8133
−10.0	263.15	1.7599
0.0	273.15	1.7031
+10.0	283.15	1.6417
20.0	293.15	1.5745
30.0	303.15	1.4990
40.0	313.15	1.4117
50.0	323.15	1.3034
55.0	328.15	1.2344
60.0	333.15	1.1464

Trifluorochloromethane data—Continued

t, °C	T, K	k, g cm^{-3}
61.0	334.15	1.1222
62.0	335.15	1.0979
63.0	336.15	1.0705
64.0	337.15	1.0321
65.0	338.15	0.9938
65.56	338.71	0.9725
(t_c)67.0	340.2	0.76 (d_c)

In the absense of raw data, these smoothed data were regressed to eq (2) with an average deviation of ±0.001 g cm^{-3}. Equation (2) is recommended assigning an uncertainty of ±0.003 g cm^{-3} in the calculated value.

26. Trifluoroiodomethane (CF_3I)

Only saturated liquid density data are available for this compound. The data are from Emeleus and Wood [58] at −22.5 °C and Nodiff et al. [7] at −78.5 °C and −32.5 °C. These are given below:

t, °C	d, g cm^{-3}	Reference
−22.5	2.316	[58]
−78.5	2.5485	[7]
−32.5	2.3608	[7]

No attempt is made at selecting the data because of insufficient information.

27. Difluorodichloromethane (CF_2Cl_2)

Only Ruff and Keim [59] have reported values of liquid densities, at −114.8, −84.8, and −72.8 °C. These, however, are too few to be analyzed and no selection for the liquid is made.

Saturated liquid data are available from Bichowsky and Gilkey [60], between −40 and +110 °C, from Vaziri [61] in the range of +55 to 110 °C, and from McHarness et al. [62] who covered the temperature range −121.91 to +112.0 °C for a 99.95 mol % pure sample. McHarness et al. employed both the float technique and the dilatometric method. McHarness et al. fitted their data to the following equation:

$$d(\text{lb ft}^{-3}) = 34.84+0.0269600(693.3-T)+0.834921(693.3-T)^{1/2}$$
$$+6.02683(693.3-T)^{1/3}-6.55549\times10^{-6}(693.3-T)^{2} \quad (10)$$

with an absolute average error of ±0.12 lb ft^{-3} (±0.002 g cm^{-3}). *In eq (10), T is in °R. Converting to density in g cm^{-3}* and temperature in K, eq (10) becomes:

$$d(\text{g cm}^{-3}) = 0.558 + 0.2994(1-T_r) + 0.3521(1-T_r)^{1/2} + 0.8544(1-T_r)^{1/3} - 0.05047(1-T_r)^2 \,. \quad (11)$$

The data source for Bichowsky and Gilkey, and McHarness et al. appears to be the same. The data of McHarness et al. are selected and regressed to eq (2) and the following expanded model is recommended:

$$d(\text{g cm}^{-3}) = 0.558 + 1.4016(1-T_r)^{1/3} - 2.8977(1-T_r)^{2/3} + 9.0828(1-T_r) - 10.6890(1-T_r)^{4/3} + 4.701(1-T_r)^{5/3} \,. \quad (12)$$

Absolute average error = ±0.001 g cm^{-3}.

Results at sample points are as follows:

t, °C	d_{calc}, g cm^{-3}	d_{expt}, g cm^{-3}
−121.91	1.7328	1.7342
0.0	1.3962	1.3946
46.9	1.2256	1.2260
106.7	0.81772	0.8141

Equation (12) is recommended with an uncertainty of ±0.001 g cm^{-3} from −122 to +90 °C, and ±0.004 g cm^{-3} from +91 to 111.8 °C (t_c), in the computed value.

28. Difluorodibromomethane (CF_2Br_2)

Only the values, 2.3492 g cm^{-3} at 0 °C and 2.2969 at 20 °C, are available from Desirant [63]. These values are recommended with ±0.001 g cm^{-3} uncertainty.

29. Fluorotrichloromethane ($CFCl_3$)

Data for the liquid density are very scarce. Ruff and Keim [59] reported a few values between −130.15 and −72.15 °C. Desreux [26] and Stevels [13] have reported single density values. None of these authors have either furnished experimental details or done measurements extensive and accurate enough to justify selection. The available liquid density values are given below:

t, °C	d, g cm^{-3}	Author	
−130.2	1.726	Ruff, Keim [59]	
−93.2	1.75	Ruff, Keim [59]	
−78.2	1.72	Ruff, Keim [59]	
−72.2	1.70	Ruff, Keim [59]	
+15.0	1.4995	Desreux	[26]
20.0	1.490	Stevels	[13]

For the saturated liquid, Benning and McHarness [48] have extensively covered the temperature region from −30 to +191 °C by dilatometric measurements. Pavlova [64] represented their experimental molar volumes from −50 to +50 °C by the following equation with a maximum deviation of 0.5% (±0.005 g cm^{-3}) from experimental data:

$$v(\text{cm}^3\text{g}^{-1}) = 0.6518 \times (1 + 1.5\times 10^{-3}t + 3.7\times 10^{-6}t^2 + 0.7\times 10^{-8}t^3) \quad (13)$$

t in °C.

		d, g cm^{-3}		
t, °C	v, cm^{3}g^{-1}	Ref. [64]	Ref. [48]	$\delta\times 10^4$
−29.2	0.6252	1.5995	1.5990	+5.
0.0	0.6518	1.5342	1.5342	0.
+26.10	0.6790	1.4728	1.4733	−5.
40.50	0.6957	1.4374	1.4379	−5.
48.80	0.7058	1.4168	1.4173	−5.

(δ = difference in densities between ref [48] and ref [64]).

Based upon the reported uncertainty, the data from Pavlova, and Benning and McHarness, were selected and regressed to eq (2) with an absolute average deviation ±0.0002 g cm^{-3}. Equation (2) is recommended (table 4) for the range −40 to +191 °C with an uncertainty of ±0.0008 g cm^{-3}, and for the range 191 to 198.0 °C (t_c) with an uncertainty of ±0.002 g cm^{-3} in the calculated value.

Sample results are shown below:

t, °C	d_{calc}, g cm^{-3}	d_{expt}, g cm^{-3}
−29.20	1.5988	1.5990
+26.10	1.4733	1.4733
98.89	1.2790	1.2791
190.9	0.8163	0.8170

30. Fluorotribromomethane ($CFBr_3$)

For the liquid, the following values are available:

t, °C	d, g cm^{-3}	Author	
20	2.7648	Desreux	[26]
20	2.757	Stevels	[13]
0	2.8211	Desirant	[63]
20	2.7648	Desirant	[63]

The values of Desirant truncated to three decimal places are recommended, assigning an uncertainty of ±0.001 g cm^{-3} in each value as given below:

t, °C	d, g cm^{-3}
0	2.821 ± 0.001
20	2.765 ± 0.001

31. Trichlorobromomethane (CCl_3Br)

The following values for the liquid are available:

t, °C	d, g cm^{-3}	Author	
20	2.012	Stevels	[13]
20	1.9942	Zokharkin	[65]

For 20 °C, 2.00 ± 0.01 g cm^{-3} is recommended.

32. Trichloroiodomethane (CCl_3I)

Stevels [13] reported a single value of 2.355 g cm^{-3} at 20 °C for the liquid. This is the only value available and is recommended, assigning an uncertainty of ±0.005 g cm^{-3}.

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