

## The solubility of nitrogen, oxygen and argon in water and seawater

R. F. WEISS†

(Received 12 January 1970)

**Abstract**—Recent precise data on the solubilities of nitrogen, oxygen and argon in distilled water and seawater are fitted to thermodynamically consistent equations by the method of least squares. The temperature dependence of the Bunsen solubility coefficient is treated using the integrated van't Hoff equation. It is shown that the Setchénow relation gives an adequate representation of the salting-out effect. Equations expressing the solubilities of these gases, as a function of temperature and salinity, in units of the Bunsen coefficient, ml/l., and ml/kg are given along with solubility tables in each of these units.

### INTRODUCTION

ALTHOUGH a number of recent precise measurements of the solubilities of gases in distilled water and seawater by various workers have shown excellent agreement, there has been little agreement on methods of smoothing and representing their results. In addition, the lack of a uniform and thermodynamically sound treatment of the temperature and salinity dependence of these data has made comparison, interpolation, and extrapolation extremely difficult.

Solubilities have, in general, been reported in terms of the Henry's Law constant  $K$ , the Bunsen solubility coefficient  $\beta$ ,‡ or the air solubility  $C^*$  (ml/l.). The Bunsen coefficient is defined as the volume of gas (STP) absorbed per unit volume of liquid at the temperature of the measurement when the partial pressure of the gas is one atmosphere. The air solubility is the volume of the gas (STP) absorbed from water saturated air at a total pressure of one atmosphere, per unit volume of the liquid at the temperature of the measurement.

Previous treatments of the temperature dependence of solubility have used both graphical and numerical methods. DOUGLAS (1964, 1965) fit his values of  $\beta$  for  $N_2$ ,  $O_2$ , and Ar in distilled water and  $N_2$  and Ar in seawater by purely graphical means, as did KLOTS and BENSON (1963) for their values of  $K$  for these gases in distilled water. CARPENTER (1966) made an approximate polynomial fit of his  $C^*$  values for  $O_2$  in distilled water and seawater and smoothed the residuals graphically. A three term expression obtained by integration of the van't Hoff equation was used by MORRISON and JOHNSTONE (1954) to treat the solubilities of the noble gases per unit mass of distilled water. A four term expansion of this equation was used by GREEN and CARRITT (1967a) to fit values of  $\beta$  for  $O_2$  in distilled water and seawater. MURRAY, RILEY and WILSON (1969) and MURRAY and RILEY (1969) used a parabolic fit of  $\ln C^*$

†Scripps Institution of Oceanography, University of California at San Diego, La Jolla, California 92037.

‡The symbol  $\beta$  is used here for the Bunsen solubility coefficient instead of  $\alpha$ , which is easily confused with many other scientific parameters commonly given this symbol. The isotopic fractionation factor  $\alpha$ , in particular, is being used increasingly in dissolved gas studies (RICHARDS and BENSON, 1961; WEISS, 1970).

vs.  $1/\sqrt{T}$  (i.e. a three term power series in  $1/\sqrt{T}$ ) to fit their values for the solubilities of  $N_2$  and  $O_2$  in distilled water and seawater. HIMMELBLAU (1960) used a parabolic fit of  $\log K$  vs.  $1/T$  rotated by some small angle relative to the co-ordinates to represent the solubilities of various gases in distilled water over an extended temperature range.

The salinity dependence of solubility (salting-out) has also been treated by a number of different methods. A linear graphical fit of  $\beta$  vs. chlorinity over a limited (15‰ to 21‰) chlorinity range was used by Douglas for  $N_2$  and Ar. Carpenter fit values of  $C^*$  for  $O_2$  to a quadratic in chlorinity and graphically smoothed the residuals. MURRAY, RILEY and WILSON (1969) and MURRAY and RILEY (1969) fit their values of  $C^*$  for  $N_2$  and  $O_2$  to a three term power series in the square root of chlorinity. The empirical Setchénow relation, which states that at constant temperature the logarithm of the solubility coefficient is a linear function of salt concentration, and is commonly used to describe the salting out of non-electrolytes (HARNED and OWEN, 1958; MORRISON, 1952) was used by GREEN and CARRITT (1967b) to fit values of  $\beta$  for  $O_2$ .

#### SELECTION OF DATA

In order to test the goodness of fit of various solubility equations, high precision  $N_2$ ,  $O_2$  and Ar solubility data were selected from the literature. The choice of data was based upon the level of agreement with other workers as well as precision and susceptibility of the experimental method to systematic errors. Discussions of these factors are given by the individual authors in presenting their results and will receive only limited mention here. In addition, the choice of data was confined to those studies which include solubility measurements in distilled water as well as in seawater of various salinities and cover a temperature range commonly found in such natural waters (approximately  $0^\circ$  to  $35^\circ C$ ).

Nitrogen solubility measurements covering an adequate range of temperatures and salinities have been made by DOUGLAS (1964, 1965) and MURRAY, RILEY and WILSON (1969). The agreement between their work is extraordinary, showing an average difference of less than 0.5%. The distilled water  $N_2$  values of KLOTS and BENSON (1963) are in general about 1–1.5% higher than those of DOUGLAS (1964, 1965), and MURRAY, RILEY and WILSON (1969). Because of the close agreement between their solubilities, obtained by completely different techniques, it was decided to combine the results of DOUGLAS (1964, 1965), and of MURRAY, RILEY and WILSON (1969) in the present treatment.

Recently oxygen solubility measurements covering an adequate range of temperatures and salinities have been made by GREEN (1965), CARPENTER (1966), and MURRAY and RILEY (1969). The data of Carpenter, and of Murray and Riley, are in such good agreement as to be virtually indistinguishable. Over the range of temperatures and salinities covered, the results of Murray and Riley are on the average about 0.1% lower than those of Carpenter. Their data do not, however, agree well with Green's data, particularly in the low temperature range where Green's data are of the order of 1% higher than those of Carpenter and Murray and Riley. Carpenter has pointed out that Green failed to correct his work for oxygen added with reagents used in the determination and therefore should have obtained somewhat high results. Although the distilled water oxygen results of KLOTS and BENSON (1963) show a similar departure from the results of Carpenter, and of Murray and Riley at

low temperatures, it should be remembered that Klots and Benson's nitrogen values are also somewhat higher than those of other workers. As before, because of the extraordinary agreement between two independent sets of measurements it was decided to combine the results of Carpenter and Murray and Riley in the present treatment.

Argon solubility measurements by DOUGLAS (1964, 1965), the only high precision data in the literature covering an adequate range of temperatures and salinities, are used in the present treatment. The distilled water argon solubility values of KLOTS and BENSON (1963) average only about 0.2% higher than those of Douglas and are in good agreement.

Only raw unsmoothed solubility data given by the various authors were used in this work. In order to eliminate effects not directly related to the solubility of gases in water, all data were first converted to units of  $\beta$ . Values of  $C^*$  reported by Carpenter and Murray and Riley were converted using the vapor pressure of water and the composition of dry air. The vapor pressure was calculated using the formulation of GOFF and GRATCH (1946) for the vapor pressure of pure water and the equation of WITTING (1908) for the vapor pressure lowering by sea salt. The molecular percentages of  $N_2$ ,  $O_2$  and Ar in dry air were taken as 78.084, 20.946, and 0.934 respectively (GLUECKAUF, 1951). Values of chlorinity were converted to salinity, a unit more common in reporting oceanographic observations, using the equation  $S\% = 1.80655 \cdot Cl\%$  (WOOSTER, LEE and DIETRICH, 1969).

Neither Douglas nor Murray, Riley and Wilson corrected their nitrogen or argon solubility data for the effect of the dissolution of the gas on the volume of the aqueous phase. In the present treatment, partial molal volumes determined by ENNS, SCHOLANDER and BRADSTREET (1965) were used to correct the data for this effect. The corrected solubilities are increased by approximately 0.14%.

#### SOLUBILITY EQUATIONS

The temperature dependence of solubility at constant salinity is obtained from the integrated form of the van't Hoff equation (HILDEBRAND and SCOTT, 1950). If the thermal expansion of water and if  $\Delta C_p$ , the change in heat capacity of the gas in going from the ideal gaseous to the dissolved state, are expanded in powers of  $T$ , then  $\beta(T)$  is obtained in the form:

$$\ln \beta = a_1 + a_2/T + a_3 \ln T + a_4 T + a_5 T^2 + \dots \quad (1)$$

where the  $a$ 's are constants and  $T$  is the absolute temperature.

A digital computer program was written to fit the solubility data to equation (1) by the method of least squares. Each curve was fitted three times, using the first 3, 4 and 5 terms of the equation respectively. The standard deviation of the points about the regression line was calculated for each fit and compared. In no case did the use of 4 or 5 terms improve upon the spread of the data about the three term fit by more than 6% of the standard deviation. In most cases the standard deviation using 4 or 5 terms was actually several per cent greater than the standard deviation obtained by using the 3 term fit. This is due to the increased number of degrees of freedom removed by the introduction of extra terms. These results argue convincingly that the data are well represented by the three term fit.

In their treatment of the temperature dependence of solubility MURRAY, RILEY and WILSON (1969) concluded that  $\ln C^*$  should be linear in  $1/T$  and suggested that observed departures from linearity are associated with changes in the structure of water. Their treatment is, however, based on the incorrect assumption that  $\Delta H$ , the "heat of escape", is independent of temperature. Furthermore, in expressing solubility in terms of  $C^*$  rather than  $K$ , the thermal expansion of water and the temperature dependence of the vapor pressure of water are brought into the treatment. The observed deviations from linearity therefore reflect the variation with temperature of  $\Delta H$  and of the vapor pressure and density of water, and do not support the existence of changes in the structure of water.

Considerable discussion has appeared in the literature attempting to justify or discredit the use of the empirical Setchénow relation in representing the salinity dependence of solubility at constant temperature. In order to test this, points were

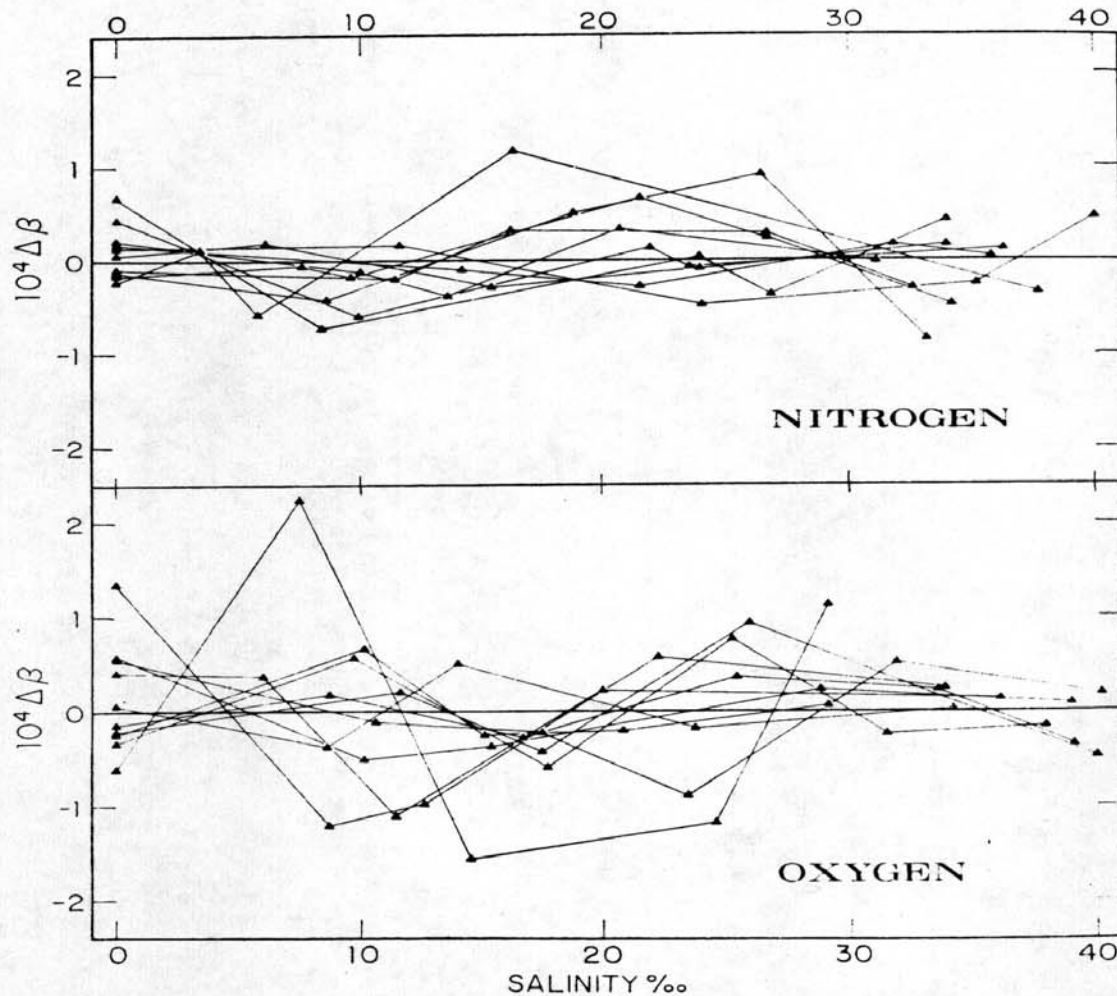


Fig. 1. Deviations of the Bunsen solubility coefficients of MURRAY, RILEY and WILSON (1969) and MURRAY and RILEY (1969) from fits of the Setchénow relation to the data, plotted against salinity. Lines connect the points measured at constant temperature.

taken from the least squares fits to the first three terms of equation (1) at 2°C intervals from -2° to +40°C. For each temperature the points determined at the various salinities were fit to the Setchénow relation, which may be written

$$\ln \beta = b_1 + b_2 S_{\text{‰}} \quad (2)$$

where the  $b$ 's are constants and  $S_{\text{‰}}$  is the salinity in per mil. Thus, for each set of experimental data for each gas, 23 separate isotherms were fitted to the data. The average and standard deviation of the  $\ln \beta$  vs.  $T$  curves about these isotherms for each value of salinity were investigated for systematic trends. In all cases these average and standard deviations for each salinity were notably less than the standard deviation of the original data about the  $\ln \beta$  vs.  $T$  fit, and therefore appear to support the validity of the Setchénow treatment.

Perhaps the best check of the Setchénow relation is afforded by the measurements of MURRAY, RILEY and WILSON (1969) and MURRAY and RILEY (1969). Unlike the other data used in this treatment, their  $N_2$  and  $O_2$  solubilities were determined at constant temperature and varying salinity and may therefore be used to test the Setchénow treatment directly rather than through an intermediate step of data fitting. The deviations of their  $N_2$  and  $O_2$  data from the Setchénow fits are plotted as a function of salinity in Fig. 1. As may be seen from the distribution of the points, and by following the lines which connect points of constant temperature, there are no systematic deviations from the Setchénow salinity dependence. Maximum deviation from the Setchénow fit for both gases is  $\sim 0.5\%$  [not  $1.5\%$  as stated by MURRAY, RILEY and WILSON (1969)]. Average deviation from the Setchénow fit for both gases was  $\sim 5 \times 10^{-5}$  in  $\beta$ . Although this average deviation is somewhat greater than the average deviations reported by Murray, Riley and Wilson for their power series treatment, it is quite close in magnitude to the average total correction required by their complete smoothing function.

Since none of the data investigated show a significant systematic error in the Setchénow treatment, it was adopted along with the first three terms of equation (1) in the final smoothing of  $\beta$ . Use of the Setchénow relation, rather than a more complex treatment of the salinity dependence, also permits the construction of relatively simple equations giving the solubility as a function of both temperature and salinity.

#### PROCEDURE

In summary, the procedure is as follows:

- (1) All solubility data are converted to units of  $\beta$ .
- (2) Data measured at constant temperature and varying salinity are fitted to the Setchénow relation (equation 2) by the method of least squares. Values at constant salinity and varying temperature are taken from these fits.
- (3) Data measured at constant salinity and varying temperature, or calculated in the previous step, are fitted to the first three terms of equation (1).
- (4) Points calculated from these fits are used to fit 23 evenly spaced isotherms from -2° to +40°C to the Setchénow equation.
- (5) The intercepts of these isotherms at zero salinity (i.e., the constants  $b_1$  in equation 2) are refitted to the first three terms of equation (1) to give a final equation for the temperature dependence of  $\beta$  in distilled water.

(6) The slopes of the isotherms (the constants  $b_2$  in equation 2) are fitted to a power series in  $T$ —three terms are needed for the required accuracy.

The final equation for  $\beta$  as a function of temperature and salinity which this treatment gives is of the form:

$$\ln \beta = A_1 + A_2 (100/T) + A_3 \ln (T/100) + S_{\text{‰}} [B_1 + B_2 (T/100) + B_3 (T/100)^2] \quad (3)$$

where the  $A$ 's and  $B$ 's are constants,  $T$  is absolute temperature, and  $S_{\text{‰}}$  is salinity in per mil.

It is also desirable to construct equations for gas solubilities from moist air as a function of temperature and salinity. Data on the composition of dry air and equations for the vapor pressure of water discussed earlier were used to calculate air solubilities in ml/l. from equation (3). Values calculated were for  $-2^\circ$  to  $+40^\circ\text{C}$  in steps of  $2^\circ\text{C}$  at salinities of 0, 30 and 40 per mil. These temperatures and salinities were chosen so that any bias introduced by the temperature dependence of the vapor pressure of water would have a minimum effect on the final equation when used for seawater and for most naturally occurring fresh water. The calculated air solubilities were then cycled through the procedure used to generate equation (3). The inclusion of the vapor pressure of water required only one additional parameter for the temperature dependence to give the same precision as observed in  $\beta$ .

Although it has been common to report some chemical concentrations in oceanography per unit volume of seawater, accurate work with such units often requires the use of tedious corrections for the compressibility and thermal expansion of water (CRAIG, WEISS and CLARKE, 1967; CRAIG and WEISS, 1968). As analytical precision is improved, the need to express concentrations per unit mass of seawater, as is done with salinity, increases. By so doing, most ambiguities caused by temperature and pressure effects are eliminated.

With this in mind, it was decided to construct equations for gas solubilities from moist air in units of milliliters per kilogram of seawater. KNUDSEN'S (1901) formula for  $\sigma_t$  was used to calculate densities which were applied to the air solubilities in ml/l. The procedure used to fit air solubilities in ml/l. was also found to be adequate for air solubilities in ml/kg and gave an equation of identical form:

$$\ln C^* = A_1 + A_2 (100/T) + A_3 \ln (T/100) + A_4 (T/100) + S_{\text{‰}} [B_1 + B_2 (T/100) + B_3 (T/100)^2] \quad (4)$$

where  $C^*$  may be either the solubility in ml (STP)/l. or in ml (STP)/kg from water saturated air at a total pressure of one atmosphere, the  $A$ 's and  $B$ 's are constants,  $T$  is the absolute temperature, and  $S_{\text{‰}}$  is the salinity in per mil.

## RESULTS

In combining the results of several workers, particular care must be paid to the weighting of the data. In the data reported here, differences in the numbers of measurements and in the degrees of scatter are statistically far less important than the magnitude of systematic differences between the methods. In general, once differences in the temperature and salinity distributions of the sets of data have been accounted for, an attempt should be made to weight the various methods as equally as possible.

## Nitrogen

The 51 solubility determinations of MURRAY, RILEY and WILSON (1969) were used to construct curves of  $\beta$  as a function of  $T$  at 0, 9, 18, 27 and 36 per mil salinities, reflecting the relatively even distribution of their data over the temperature and salinity range measured. These curves were combined with DOUGLAS' (1964, 1965) 101 solubility determinations measured at various temperatures at 0, 27.78, 33.61 and 37.91 per mil salinities. The nine curves thus obtained were given equal weight in fitting to equation (3), thereby insuring that the weighting of each set of data would approximate the distribution of the temperatures and salinities of the original data.

Constants from the fitting of the combined data to equations (3) and (4) for values of  $\beta$  and solubilities in ml/l. and ml/kg are given in Tables 1, 2 and 3 respectively.

Table 1. Constants for the calculation of the Bunsen solubility coefficient according to equation (3).

Gas	$A_1$	$A_2$	$A_3$	$B_1$	$B_2$	$B_3$
N <sub>2</sub>	- 59.6274	85.7661	24.3696	- 0.051580	0.026329	- 0.0037252
O <sub>2</sub>	- 58.3877	85.8079	23.8439	- 0.034892	0.015568	- 0.0019387
Ar	- 55.6578	82.0262	22.5929	- 0.036267	0.016241	- 0.0020114

Table 2. Volumetric solubility constants for the calculation of solubilities in ml/l. from moist air at one atmosphere total pressure, according to equation (4).

Gas	$A_1$	$A_2$	$A_3$	$A_4$	$B_1$	$B_2$	$B_3$
N <sub>2</sub>	- 172.4965	248.4262	143.0738	- 21.7120	- 0.049781	0.025018	- 0.0034861
O <sub>2</sub>	- 173.4292	249.6339	143.3483	- 21.8492	- 0.033096	0.014259	- 0.0017000
Ar	- 173.5146	245.4510	141.8222	- 21.8020	- 0.034474	0.014934	- 0.0017729

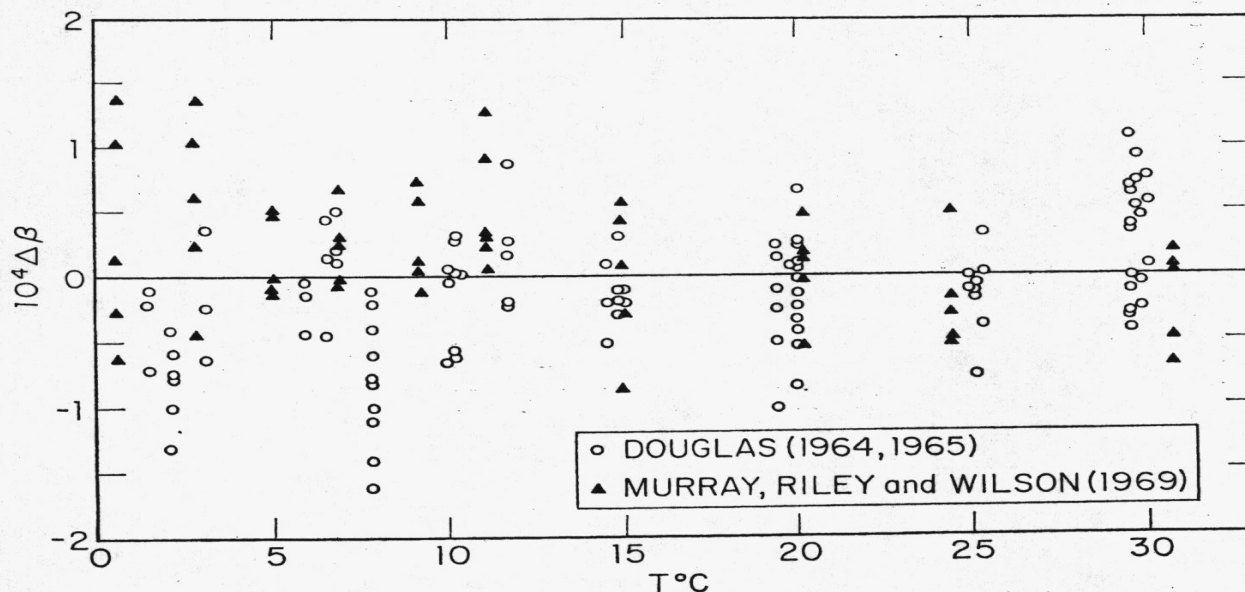


Fig. 2. Nitrogen solubilities—deviations of the original data in units of  $\beta$  from the combined fit, plotted against temperature.

Table 3. Gravimetric solubility constants for the calculation of solubilities in ml/kg from moist air at one atmosphere total pressure, according to equation (4).

Gas	$A_1$	$A_2$	$A_3$	$A_4$	$B_1$	$B_2$	$B_3$
N <sub>2</sub>	-177.0212	254.6078	146.3611	-22.0933	-0.054052	0.027266	-0.0038430
O <sub>2</sub>	-177.7888	255.5907	146.4813	-22.2040	-0.037362	0.016504	-0.0020564
Ar	-178.1725	251.8139	145.2337	-22.2046	-0.038729	0.017171	-0.0021281

Deviations of the original data from the combined fit to equation (3) are plotted in Fig. 2. The combined data, the data of MURRAY, RILEY and WILSON (1969) only, and the data of DOUGLAS (1964, 1965) only, all show a root mean square deviation from the combined fit of  $\sim 5.3 \times 10^{-5}$  in units of  $\beta$ . On the average, the data of Douglas are  $\sim 3 \times 10^{-5}$  lower, also in units of  $\beta$ , than the data of MURRAY, RILEY and WILSON (1969). Brief tables of the solubilities of nitrogen in units of  $\beta$ , ml/l. and ml/kg at various salinities and temperatures as calculated from equations (3) and (4) are given in Tables 4, 5 and 6 respectively.

Table 4. Bunsen coefficients  $\times 100$  for nitrogen.

T (°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	2.096	1.944	1.887	1.873	1.859	1.831	1.804
0	2.374	2.203	2.045	1.898	1.842	1.829	1.815	1.788	1.762
1	2.314	2.149	1.996	1.854	1.800	1.786	1.773	1.747	1.721
2	2.257	2.097	1.949	1.811	1.759	1.746	1.733	1.708	1.683
3	2.202	2.048	1.904	1.770	1.720	1.707	1.695	1.670	1.646
4	2.150	2.000	1.861	1.731	1.682	1.670	1.658	1.634	1.611
5	2.100	1.955	1.820	1.694	1.646	1.634	1.623	1.600	1.577
6	2.053	1.912	1.780	1.658	1.612	1.600	1.589	1.567	1.544
8	1.963	1.830	1.707	1.591	1.547	1.536	1.526	1.504	1.484
10	1.881	1.756	1.639	1.529	1.488	1.478	1.467	1.447	1.428
12	1.806	1.687	1.576	1.473	1.433	1.424	1.414	1.395	1.376
14	1.736	1.624	1.519	1.420	1.383	1.374	1.364	1.346	1.328
16	1.672	1.566	1.466	1.372	1.336	1.328	1.319	1.302	1.285
18	1.614	1.512	1.417	1.328	1.294	1.285	1.277	1.260	1.244
20	1.559	1.463	1.372	1.287	1.254	1.246	1.238	1.222	1.207
22	1.510	1.417	1.330	1.249	1.218	1.210	1.202	1.187	1.172
24	1.463	1.375	1.292	1.214	1.184	1.176	1.169	1.155	1.140
26	1.421	1.336	1.256	1.181	1.153	1.146	1.139	1.125	1.111
28	1.382	1.300	1.224	1.151	1.124	1.117	1.110	1.097	1.084
30	1.345	1.267	1.193	1.124	1.097	1.091	1.084	1.071	1.058
32	1.312	1.236	1.165	1.098	1.073	1.066	1.060	1.047	1.035
34	1.281	1.208	1.140	1.075	1.050	1.044	1.038	1.026	1.014
36	1.253	1.182	1.116	1.053	1.029	1.023	1.017	1.005	.994
38	1.226	1.158	1.094	1.033	1.010	1.004	.998	.987	.975
40	1.202	1.136	1.074	1.014	.992	.986	.981	.970	.959

### Oxygen

The data of CARPENTER (1966) were first adjusted, as was done by Carpenter, to give five curves of varying temperature at 0, 5-10, 10-30, 15-10, and 20-50 per mil chlorinities†.

†In Carpenter's Table 3 the seventh point down from the top at a chlorinity of 15.00‰ apparently has a typographical error in the temperature (CARPENTER, *personal communication*). The corrected value of 15.00°C, not 14.10°C, has been used in this work.



In Carpenter's Table 2 a number of distilled water solubility measurements made without analyzing the composition of the gas phase are reported. It was found that when these data were included in the distilled water solubility fits, they biased the low temperature fit by about 0.1% in the negative direction. Because of this effect it was decided not to include these measurements in the treatment reported here.

The 53 solubility determinations of MURRAY and RILEY (1969) were used to construct five curves of  $\beta$  as a function of  $T$  at 0, 9, 18, 27 and 36 per mil salinities. These curves were combined with the five similar curves of  $\beta$  obtained from the data of Carpenter, thereby giving equal weight to each method as well as reflecting the even distribution of both sets of data with respect to temperature and salinity.

Constants from the fitting of the combined data to equations (3) and (4) for values of  $\beta$  and solubilities in ml/l. and ml/kg are given in Tables 1, 2 and 3 respectively. Deviations of the original data in ml/l. from the combined fit to equation (4) are plotted in Fig. 3. The root mean square deviation of all the combined data points from

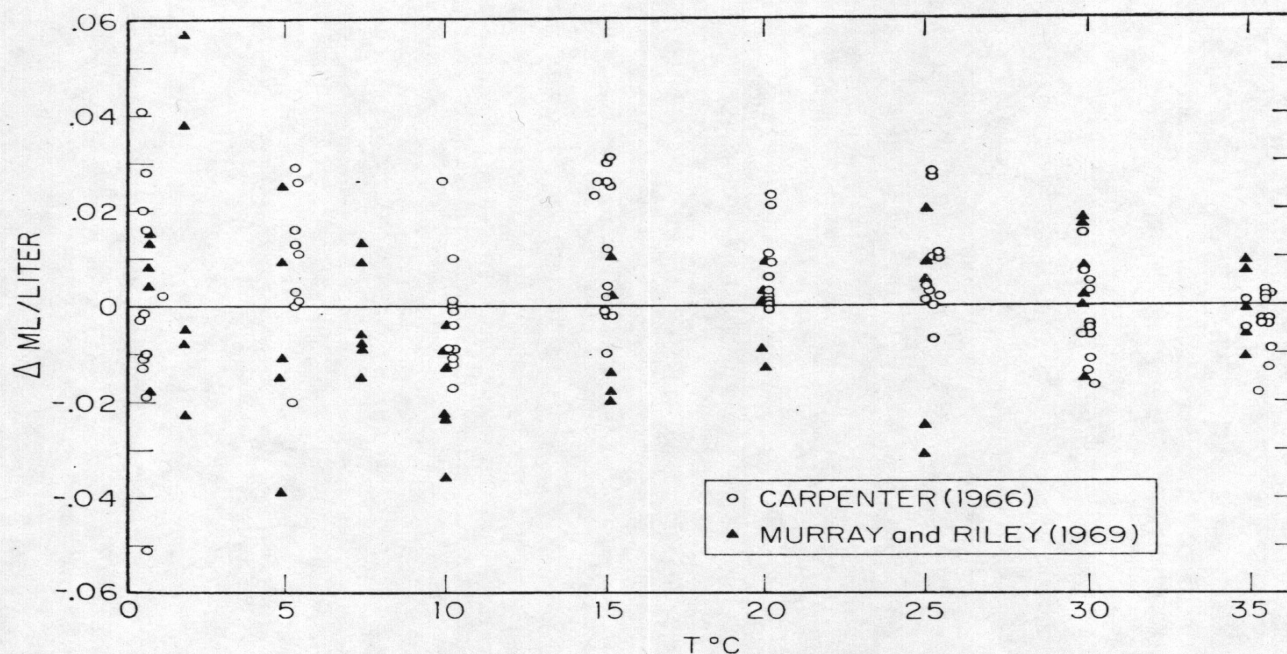


Fig. 3. Oxygen solubilities—deviations of the original data in ml/l. from the combined fit, plotted against temperature.

the combined fit is 0.016 ml/l. The root mean square deviation of the data of MURRAY and RILEY (1969) from the combined fit is 0.018 ml/l., while the value for the 90 measurements of CARPENTER (1966) used in this treatment is 0.015 ml/l. The data of Carpenter are on the average 0.005 ml/l. higher than the data of Murray and Riley. Brief tables for the solubility of oxygen in units of  $\beta$ , ml/l. and ml/kg at various temperatures and salinities as calculated from equations (3) and (4) are given in Tables 7, 8 and 9 respectively.

Table 5. Solubility of nitrogen from moist air at one atmosphere total pressure in (ml/l).

T(°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	16.28	15.10	14.65	14.54	14.44	14.22	14.01
0	18.42	17.10	15.87	14.73	14.30	14.19	14.09	13.88	13.67
1	17.95	16.67	15.48	14.38	13.96	13.86	13.75	13.55	13.35
2	17.50	16.26	15.11	14.04	13.64	13.54	13.44	13.24	13.05
3	17.07	15.87	14.75	13.72	13.32	13.23	13.13	12.94	12.76
4	16.65	15.49	14.41	13.41	13.03	12.93	12.84	12.66	12.47
5	16.26	15.13	14.09	13.11	12.74	12.65	12.56	12.38	12.21
6	15.88	14.79	13.77	12.83	12.47	12.38	12.29	12.12	11.95
8	15.16	14.14	13.18	12.29	11.95	11.87	11.79	11.62	11.46
10	14.51	13.54	12.64	11.80	11.48	11.40	11.32	11.17	11.01
12	13.90	12.99	12.14	11.34	11.04	10.96	10.89	10.74	10.60
14	13.34	12.48	11.67	10.92	10.63	10.56	10.49	10.35	10.21
16	12.83	12.01	11.24	10.53	10.25	10.19	10.12	9.99	9.86
18	12.35	11.57	10.84	10.16	9.90	9.84	9.77	9.65	9.52
20	11.90	11.16	10.47	9.82	9.57	9.51	9.45	9.33	9.21
22	11.48	10.78	10.12	9.50	9.26	9.21	9.15	9.03	8.92
24	11.09	10.42	9.79	9.20	8.98	8.92	8.87	8.76	8.65
26	10.73	10.09	9.49	8.92	8.71	8.65	8.60	8.50	8.39
28	10.38	9.77	9.20	8.66	8.45	8.40	8.35	8.25	8.15
30	10.06	9.48	8.93	8.41	8.21	8.16	8.12	8.02	7.92
32	9.76	9.20	8.67	8.18	7.99	7.94	7.89	7.80	7.71
34	9.48	8.94	8.43	7.96	7.77	7.73	7.68	7.59	7.51
36	9.21	8.69	8.20	7.75	7.57	7.53	7.48	7.40	7.31
38	8.95	8.46	7.99	7.55	7.38	7.33	7.29	7.21	7.13
40	8.71	8.23	7.78	7.36	7.19	7.15	7.11	7.03	6.95

Table 6. Solubility of nitrogen from moist air at one atmosphere total pressure in (ml/kg).

T(°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	16.03	14.75	14.26	14.15	14.03	13.80	13.57
0	18.43	16.97	15.62	14.39	13.92	13.81	13.69	13.47	13.25
1	17.95	16.54	15.24	14.04	13.59	13.48	13.37	13.15	12.94
2	17.50	16.13	14.87	13.71	13.28	13.17	13.06	12.85	12.64
3	17.07	15.74	14.52	13.40	12.97	12.87	12.77	12.56	12.36
4	16.65	15.37	14.19	13.10	12.68	12.58	12.48	12.28	12.09
5	16.26	15.01	13.87	12.81	12.41	12.31	12.21	12.02	11.83
6	15.88	14.67	13.56	12.53	12.14	12.05	11.95	11.77	11.58
8	15.17	14.03	12.98	12.01	11.64	11.55	11.46	11.29	11.11
10	14.51	13.44	12.45	11.53	11.18	11.10	11.01	10.85	10.68
12	13.91	12.90	11.96	11.09	10.76	10.68	10.60	10.44	10.28
14	13.35	12.40	11.51	10.68	10.37	10.29	10.21	10.06	9.91
16	12.84	11.93	11.09	10.30	10.00	9.93	9.86	9.71	9.57
18	12.36	11.50	10.70	9.95	9.66	9.59	9.52	9.39	9.25
20	11.92	11.10	10.33	9.62	9.35	9.28	9.21	9.08	8.96
22	11.51	10.72	9.99	9.31	9.05	8.99	8.93	8.80	8.68
24	11.12	10.37	9.68	9.02	8.78	8.72	8.65	8.54	8.42
26	10.76	10.05	9.38	8.75	8.52	8.46	8.40	8.29	8.17
28	10.42	9.74	9.10	8.50	8.27	8.22	8.16	8.05	7.94
30	10.11	9.45	8.84	8.26	8.04	7.99	7.94	7.83	7.73
32	9.81	9.18	8.59	8.04	7.83	7.78	7.73	7.62	7.52
34	9.53	8.92	8.36	7.83	7.62	7.57	7.52	7.43	7.33
36	9.26	8.68	8.14	7.63	7.43	7.38	7.33	7.24	7.15
38	9.01	8.45	7.93	7.43	7.25	7.20	7.15	7.06	6.97
40	8.78	8.24	7.73	7.25	7.07	7.03	6.98	6.89	6.81

Table 7. Bunsen coefficients  $\times 100$  for oxygen.

T(°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	4.400	4.107	3.996	3.968	3.941	3.887	3.834
0	4.910	4.586	4.283	4.000	3.892	3.865	3.839	3.787	3.736
1	4.777	4.464	4.171	3.897	3.793	3.767	3.742	3.691	3.642
2	4.650	4.347	4.064	3.799	3.698	3.674	3.649	3.600	3.552
3	4.529	4.236	3.962	3.706	3.608	3.584	3.560	3.513	3.466
4	4.413	4.129	3.864	3.616	3.521	3.498	3.475	3.429	3.384
5	4.302	4.028	3.771	3.530	3.438	3.416	3.393	3.349	3.305
6	4.196	3.930	3.681	3.448	3.359	3.337	3.316	3.272	3.230
8	3.998	3.748	3.514	3.295	3.211	3.190	3.170	3.129	3.089
10	3.816	3.581	3.360	3.153	3.074	3.055	3.035	2.997	2.959
12	3.649	3.427	3.219	3.024	2.949	2.930	2.912	2.876	2.840
14	3.495	3.286	3.089	2.904	2.834	2.816	2.799	2.764	2.730
16	3.354	3.156	2.970	2.795	2.727	2.711	2.694	2.662	2.630
18	3.224	3.037	2.860	2.693	2.629	2.614	2.598	2.567	2.536
20	3.105	2.926	2.758	2.600	2.539	2.524	2.509	2.480	2.451
22	2.994	2.825	2.665	2.514	2.456	2.441	2.427	2.399	2.371
24	2.892	2.731	2.578	2.434	2.379	2.365	2.351	2.325	2.298
26	2.798	2.644	2.498	2.360	2.307	2.294	2.281	2.256	2.230
28	2.711	2.564	2.424	2.292	2.242	2.229	2.217	2.192	2.168
30	2.630	2.489	2.356	2.229	2.181	2.169	2.157	2.133	2.110
32	2.556	2.421	2.293	2.171	2.124	2.113	2.101	2.079	2.056
34	2.487	2.357	2.234	2.117	2.072	2.061	2.050	2.028	2.007
36	2.423	2.298	2.180	2.068	2.024	2.014	2.003	1.982	1.961
38	2.364	2.244	2.130	2.021	1.980	1.969	1.959	1.939	1.919
40	2.310	2.194	2.084	1.979	1.939	1.929	1.919	1.899	1.880

Table 8. Solubility of oxygen from moist air at one atmosphere total pressure in (ml/l.).

T(°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	9.162	8.553	8.321	8.264	8.207	8.095	7.984
0	10.218	9.543	8.913	8.325	8.100	8.045	7.990	7.882	7.775
1	9.936	9.284	8.676	8.107	7.890	7.836	7.783	7.679	7.575
2	9.666	9.037	8.449	7.898	7.689	7.637	7.586	7.484	7.384
3	9.409	8.801	8.232	7.699	7.496	7.446	7.397	7.298	7.202
4	9.163	8.574	8.024	7.509	7.312	7.264	7.216	7.121	7.027
5	8.927	8.358	7.825	7.327	7.136	7.089	7.043	6.950	6.860
6	8.702	8.151	7.635	7.152	6.967	6.922	6.877	6.787	6.699
8	8.280	7.763	7.278	6.824	6.650	6.608	6.565	6.481	6.398
10	7.891	7.406	6.950	6.522	6.359	6.319	6.279	6.199	6.121
12	7.534	7.077	6.647	6.244	6.090	6.052	6.014	5.939	5.865
14	7.204	6.773	6.367	5.987	5.841	5.805	5.769	5.698	5.628
16	6.898	6.491	6.108	5.748	5.610	5.576	5.542	5.475	5.409
18	6.615	6.230	5.868	5.527	5.396	5.363	5.331	5.268	5.205
20	6.352	5.987	5.644	5.320	5.196	5.166	5.135	5.075	5.015
22	6.106	5.761	5.435	5.128	5.010	4.981	4.952	4.895	4.838
24	5.878	5.550	5.241	4.949	4.836	4.809	4.781	4.727	4.673
26	5.664	5.353	5.058	4.780	4.673	4.647	4.621	4.569	4.518
28	5.464	5.168	4.887	4.623	4.521	4.496	4.471	4.421	4.372
30	5.276	4.994	4.727	4.474	4.377	4.353	4.329	4.282	4.235
32	5.099	4.830	4.576	4.335	4.242	4.219	4.196	4.151	4.106
34	4.932	4.676	4.433	4.203	4.114	4.092	4.070	4.027	3.984
36	4.775	4.530	4.298	4.078	3.993	3.972	3.951	3.910	3.869
38	4.627	4.393	4.171	3.960	3.878	3.858	3.838	3.799	3.760
40	4.486	4.262	4.050	3.848	3.770	3.750	3.731	3.693	3.656

Table 9. Solubility of oxygen from moist air at one atmosphere total pressure in (ml/kg).

T(°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	9.020	8.354	8.101	8.039	7.978	7.856	7.736
0	10.221	9.470	8.775	8.131	7.887	7.827	7.767	7.650	7.534
1	9.938	9.213	8.541	7.918	7.682	7.624	7.567	7.453	7.341
2	9.668	8.967	8.318	7.715	7.487	7.431	7.375	7.265	7.156
3	9.410	8.733	8.104	7.521	7.300	7.246	7.192	7.085	6.980
4	9.164	8.509	7.901	7.336	7.121	7.069	7.016	6.913	6.811
5	8.929	8.294	7.705	7.158	6.950	6.899	6.849	6.749	6.650
6	8.703	8.089	7.519	6.988	6.787	6.737	6.688	6.591	6.495
8	8.282	7.705	7.169	6.670	6.480	6.433	6.387	6.296	6.205
10	7.895	7.352	6.847	6.377	6.198	6.154	6.110	6.024	5.938
12	7.538	7.027	6.551	6.106	5.937	5.896	5.855	5.773	5.692
14	7.210	6.727	6.277	5.857	5.697	5.657	5.618	5.541	5.465
16	6.906	6.450	6.024	5.626	5.474	5.437	5.400	5.327	5.254
18	6.625	6.193	5.789	5.411	5.268	5.232	5.197	5.127	5.059
20	6.364	5.954	5.571	5.212	5.075	5.042	5.008	4.942	4.877
22	6.121	5.732	5.368	5.027	4.896	4.864	4.832	4.769	4.707
24	5.894	5.525	5.178	4.853	4.729	4.699	4.668	4.608	4.549
26	5.683	5.331	5.001	4.691	4.573	4.543	4.514	4.457	4.401
28	5.485	5.150	4.835	4.539	4.426	4.398	4.370	4.316	4.261
30	5.299	4.979	4.679	4.396	4.288	4.261	4.235	4.183	4.131
32	5.125	4.819	4.532	4.262	4.158	4.133	4.107	4.057	4.008
34	4.961	4.669	4.394	4.135	4.036	4.011	3.987	3.939	3.891
36	4.806	4.527	4.263	4.015	3.920	3.897	3.873	3.827	3.782
38	4.660	4.392	4.140	3.902	3.810	3.788	3.766	3.721	3.678
40	4.521	4.265	4.022	3.794	3.706	3.685	3.663	3.621	3.579

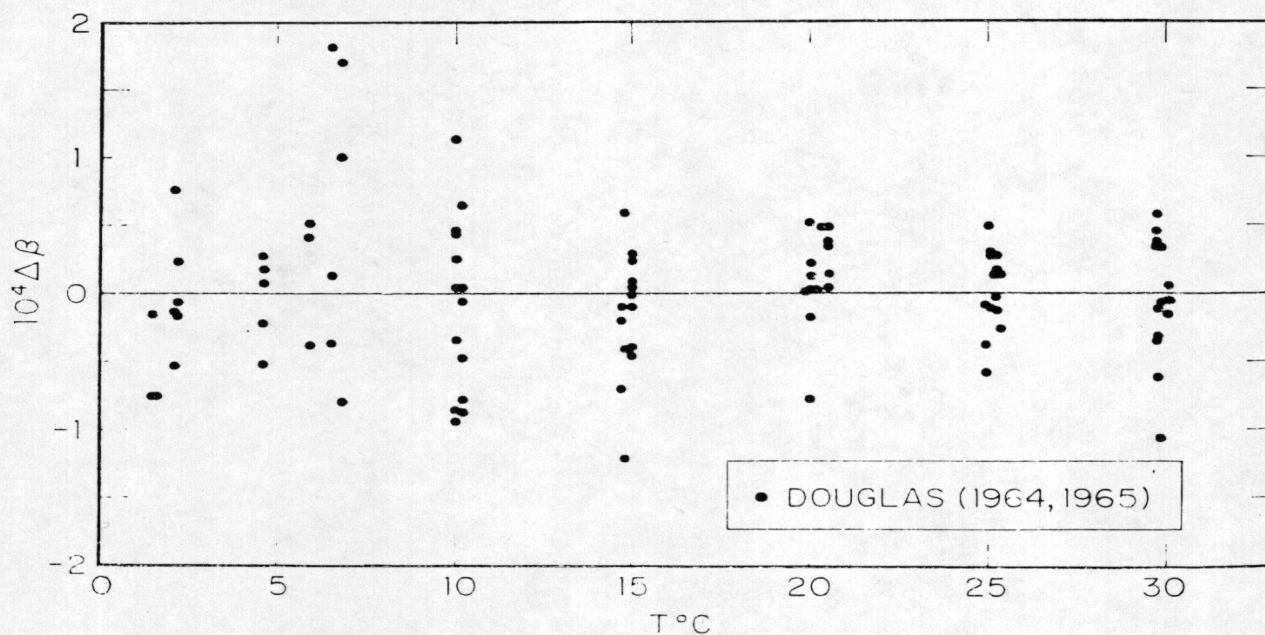


Fig. 4. Argon solubilities—deviations of the original data in units of  $\beta$  from the fitted values, plotted against temperature.

*Argon*

Constants from the fitting of DOUGLAS' (1964, 1965) 93 argon solubility measurements to equations (3) and (4) for values of  $\beta$  and solubilities in ml/l. and ml/kg are given in Tables 1, 2 and 3 respectively. Figure 4 shows the deviation of the original data from the fit to equation (3). The root mean square deviations of the data from the fit values is  $5.2 \times 10^{-5}$  in units of  $\beta$ . Brief tables for the solubility of argon in units of  $\beta$ , ml/l. and ml/kg at various temperatures and salinities as calculated from equations (3) and (4) are given in Tables 10, 11 and 12 respectively.

## SUMMARY AND CONCLUSIONS

Recent high precision measurements of  $N_2$ ,  $O_2$  and Ar solubilities in distilled water and seawater are extremely well represented by the equations in temperature and salinity described in this work. It is believed that these equations provide the most accurate solubility values available in the literature. For nitrogen the estimated accuracy is  $\pm 6 \times 10^{-5}$  in  $\beta$ , or  $\pm 0.05$  ml/l. or ml/kg in  $C^*$ . The estimated accuracy for oxygen is  $\pm 7 \times 10^{-5}$  in  $\beta$ , or  $\pm 0.015$  ml/l. or ml/kg in  $C^*$ . For argon the estimated accuracy is  $\pm 1 \times 10^{-4}$  in  $\beta$ , or  $\pm 0.0009$  ml/l. or ml/kg in  $C^*$ . Although rather difficult to estimate precisely, these accuracies are intended to represent the maximum uncertainty in the fitted values, including both statistical and systematic errors.

Table 10. Bunsen coefficients  $\times 100$  for argon.

T(°C)	SALINITY IN PER MIL									
	0	10	20	30	34	35	36	38	40	
-1	—	—	4.796	4.473	4.350	4.320	4.290	4.231	4.172	
0	5.363	5.005	4.671	4.359	4.240	4.211	4.182	4.124	4.068	
1	5.221	4.875	4.551	4.250	4.135	4.106	4.078	4.023	3.968	
2	5.084	4.750	4.437	4.145	4.034	4.006	3.979	3.925	3.872	
3	4.954	4.630	4.328	4.045	3.937	3.911	3.884	3.832	3.781	
4	4.829	4.516	4.223	3.949	3.845	3.819	3.793	3.743	3.693	
5	4.710	4.406	4.123	3.857	3.756	3.731	3.706	3.657	3.609	
6	4.595	4.302	4.027	3.769	3.671	3.647	3.623	3.575	3.529	
8	4.381	4.105	3.847	3.604	3.512	3.489	3.466	3.422	3.377	
10	4.184	3.925	3.681	3.453	3.365	3.344	3.323	3.280	3.238	
12	4.004	3.759	3.529	3.313	3.231	3.210	3.190	3.150	3.111	
14	3.837	3.606	3.389	3.184	3.106	3.087	3.068	3.030	2.993	
16	3.684	3.465	3.259	3.066	2.992	2.973	2.955	2.919	2.884	
18	3.543	3.335	3.140	2.956	2.886	2.868	2.851	2.817	2.783	
20	3.412	3.215	3.030	2.855	2.788	2.771	2.755	2.722	2.690	
22	3.291	3.104	2.928	2.761	2.697	2.682	2.666	2.635	2.604	
24	3.180	3.002	2.834	2.675	2.614	2.599	2.584	2.554	2.525	
26	3.077	2.907	2.746	2.594	2.536	2.522	2.508	2.479	2.451	
28	2.981	2.819	2.665	2.520	2.464	2.451	2.437	2.410	2.383	
30	2.893	2.737	2.590	2.451	2.398	2.385	2.372	2.346	2.320	
32	2.811	2.662	2.521	2.388	2.336	2.324	2.311	2.286	2.261	
34	2.734	2.592	2.457	2.328	2.279	2.267	2.255	2.231	2.207	
36	2.664	2.527	2.397	2.274	2.226	2.214	2.203	2.180	2.157	
38	2.599	2.467	2.342	2.223	2.177	2.166	2.154	2.132	2.110	
40	2.538	2.411	2.290	2.176	2.132	2.121	2.110	2.088	2.067	

Table 11. Solubility of argon from moist air at one atmosphere total pressure in (ml/l.).

T(°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	.4456	.4156	.4042	.4014	.3986	.3931	.3877
0	.4980	.4647	.4337	.4048	.3937	.3910	.3883	.3830	.3777
1	.4845	.4524	.4224	.3944	.3837	.3811	.3785	.3733	.3682
2	.4715	.4405	.4115	.3845	.3741	.3716	.3691	.3641	.3592
3	.4592	.4292	.4012	.3750	.3650	.3625	.3601	.3552	.3505
4	.4474	.4184	.3912	.3659	.3562	.3538	.3515	.3468	.3422
5	.4360	.4080	.3817	.3572	.3478	.3455	.3432	.3387	.3342
6	.4252	.3980	.3726	.3488	.3397	.3375	.3353	.3309	.3265
8	.4049	.3794	.3555	.3331	.3246	.3225	.3204	.3162	.3121
10	.3861	.3622	.3397	.3186	.3106	.3086	.3066	.3027	.2989
12	.3688	.3463	.3251	.3053	.2977	.2958	.2939	.2902	.2866
14	.3528	.3316	.3116	.2929	.2857	.2839	.2822	.2787	.2752
16	.3380	.3180	.2991	.2814	.2746	.2729	.2712	.2679	.2647
18	.3242	.3053	.2875	.2707	.2642	.2626	.2610	.2579	.2548
20	.3114	.2935	.2766	.2607	.2546	.2531	.2516	.2486	.2457
22	.2995	.2825	.2665	.2514	.2455	.2441	.2427	.2399	.2371
24	.2883	.2722	.2570	.2426	.2371	.2357	.2344	.2317	.2291
26	.2779	.2626	.2481	.2344	.2292	.2279	.2266	.2241	.2215
28	.2681	.2535	.2398	.2268	.2217	.2205	.2193	.2169	.2144
30	.2588	.2450	.2319	.2195	.2147	.2136	.2124	.2101	.2078
32	.2502	.2370	.2245	.2127	.2081	.2070	.2059	.2037	.2015
34	.2420	.2294	.2175	.2062	.2019	.2008	.1997	.1976	.1955
36	.2342	.2222	.2109	.2001	.1959	.1949	.1939	.1919	.1899
38	.2269	.2154	.2046	.1943	.1903	.1893	.1883	.1864	.1845
40	.2199	.2090	.1986	.1888	.1849	.1840	.1831	.1812	.1794

Table 12. Solubility of argon from moist air at one atmosphere total pressure in (ml/kg).

T(°C)	SALINITY IN PERMIL								
	0	10	20	30	34	35	36	38	40
-1	—	—	.4385	.4058	.3934	.3903	.3873	.3813	.3755
0	.4979	.4610	.4268	.3952	.3832	.3802	.3773	.3716	.3659
1	.4844	.4487	.4157	.3851	.3735	.3706	.3678	.3622	.3567
2	.4714	.4370	.4050	.3754	.3642	.3614	.3587	.3533	.3479
3	.4591	.4257	.3948	.3661	.3553	.3526	.3500	.3447	.3396
4	.4473	.4150	.3851	.3573	.3468	.3442	.3416	.3365	.3315
5	.4359	.4047	.3757	.3488	.3386	.3361	.3336	.3287	.3238
6	.4251	.3949	.3668	.3407	.3308	.3284	.3259	.3212	.3165
8	.4048	.3764	.3500	.3254	.3161	.3138	.3115	.3070	.3026
10	.3861	.3594	.3345	.3114	.3026	.3004	.2983	.2940	.2898
12	.3689	.3437	.3203	.2984	.2901	.2880	.2860	.2820	.2780
14	.3530	.3292	.3071	.2864	.2785	.2766	.2747	.2709	.2671
16	.3383	.3158	.2948	.2753	.2678	.2660	.2641	.2605	.2570
18	.3246	.3033	.2835	.2649	.2578	.2561	.2544	.2509	.2476
20	.3119	.2918	.2729	.2553	.2485	.2469	.2452	.2420	.2388
22	.3001	.2809	.2630	.2463	.2399	.2383	.2367	.2336	.2306
24	.2890	.2708	.2538	.2379	.2317	.2302	.2288	.2258	.2229
26	.2787	.2614	.2452	.2300	.2241	.2227	.2213	.2185	.2157
28	.2690	.2525	.2371	.2226	.2170	.2156	.2143	.2116	.2089
30	.2599	.2442	.2294	.2156	.2103	.2090	.2077	.2051	.2026
32	.2513	.2364	.2223	.2090	.2039	.2027	.2014	.1990	.1965
34	.2433	.2290	.2155	.2028	.1979	.1967	.1956	.1932	.1909
36	.2356	.2220	.2091	.1969	.1923	.1911	.1900	.1877	.1855
38	.2284	.2153	.2030	.1913	.1869	.1858	.1847	.1825	.1804
40	.2216	.2090	.1972	.1860	.1818	.1807	.1796	.1776	.1755

The procedures described here have also been used to treat the results of a series of helium and neon solubility measurements in distilled water and seawater made in this laboratory. The results of this work will be available shortly.

*Acknowledgements*—I thank Professor H. CRAIG for his interest, critical discussion, and support. Professor J. GIESKES also read the paper and provided valuable discussion. The work was supported by a grant from the National Science Foundation to the Isotope Laboratory, SIO. Calculations were performed at the UCSD Computer Center.

## REFERENCES

- CARPENTER J. H. (1966) New measurements of oxygen solubility in pure and natural water. *Limnol. Oceanogr.*, **11**, 264–277.
- CRAIG H. and R. F. WEISS (1968) Argon concentrations in the ocean: a discussion. *Earth Planet. Sci. Lett.*, **5**, 175–183.
- CRAIG H., R. F. WEISS and W. B. CLARKE (1967) Dissolved gases in the Equatorial and South Pacific Ocean. *J. geophys. Res.*, **72**, 6165–6181.
- DOUGLAS E. (1964) Solubilities of oxygen, argon, and nitrogen in distilled water. *J. phys. Chem.*, **68**, 169–174.
- DOUGLAS E. (1965) Solubilities of argon and nitrogen in sea water. *J. phys. Chem.*, **69**, 2608–2610.
- ENNS T., P. F. SCHOLANDER and E. D. BRADSTREET (1965) Effect of hydrostatic pressure on gases dissolved in water. *J. phys. Chem.*, **69**, 389–391.
- GLUECKAUF E. (1951) The composition of atmospheric air. *Compendium of Meteorology*, Am. Met. Soc., Boston, 3–11.
- GOFF J. A. and S. GRATCH (1946) Low pressure properties of water from  $-160^{\circ}$  to  $212^{\circ}$ F. *Trans. Am. Soc. Heat. Vent. Engrs.*, **52**, 95–122.
- GREEN E. J. (1965) A redetermination of the solubility of oxygen in sea water and some thermodynamic implications of the solubility relations. Ph.D. thesis, Mass. Inst. of Technol., Cambridge, Mass., 137 pp.
- GREEN E. J. and D. E. CARRITT (1967a) New tables for oxygen saturation of seawater. *J. mar. Res.*, **25**, 140–147.
- GREEN E. J. and D. E. CARRITT (1967b) Oxygen solubility in sea water: thermodynamic influence of sea salt. *Science*, **157**, 191–193.
- HARNED H. S. and B. B. OWEN (1958) *The Physical Chemistry of Electrolytic Solutions*, Reinhold, New York, 803 pp.
- HILDEBRAND J. H. and R. L. SCOTT (1950) *The Solubility of Nonelectrolytes*, Reinhold, New York, 488 pp.
- HIMMELBLAU D. M. (1960) Solubilities of inert gases in water. *J. chem. Engng Data*, **5**, 10–15.
- KLOTS C. E. and B. B. BENSON (1963) Solubilities of nitrogen, oxygen, and argon in distilled water. *J. mar. Res.*, **21**, 48–57.
- KNUDSEN M. (1901) *Hydrographical Tables*, G. E. Gad, Copenhagen, 63 pp.
- MORRISON T. J. (1952) The salting-out of non-electrolytes Part I. The effect of ionic size, ionic charge, and temperature. *J. Chem. Soc. (3)*, 3814–3822.
- MORRISON T. J. and N. B. JOHNSTONE (1954) Solubilities of the inert gases in water. *J. Chem. Soc. (3)*, 3441–3446.
- MURRAY C. N. and J. P. RILEY (1969) The solubility of gases in distilled water and sea water—II. Oxygen. *Deep-Sea Res.*, **16**, 311–320.
- MURRAY C. N., J. P. RILEY and T. R. S. WILSON (1969) The solubility of gases in distilled water and sea water—I. Nitrogen. *Deep-Sea Res.*, **16**, 297–310.
- RICHARDS F. A. and B. B. BENSON (1961) Nitrogen/argon and nitrogen isotope ratios in two anaerobic environments, the Cariaco Trench in the Caribbean Sea and Dramsfjord, Norway. *Deep-Sea Res.*, **7**, 254–264.
- WEISS R. F. (1970) Helium isotope effect in solution in water and seawater. *Science*, **168**, 247–248.
- WITTING R. (1908) Untersuchungen zur Kenntnis der Wasserbewegungen und der Wasserumsetzung in den Finland umgebenden Meeren. I. Finland. *Hydrogn. Biol. Untersuch.* (2), 173–190.
- WOOSTER W. S., A. J. LEE and G. DIETRICH (1969) Redefinition of salinity. *Deep-Sea Res.*, **16**, 321–322.

