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The concentration and isotopic fractionation of oxygen dissolved in freshwater and seawater in equilibrium with the atmosphere[1]

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Abstract

Previous work on the solubility of oxygen in freshwater has been extended to seawater. Measurements of the Henry coefficient in the ranges $0° < t < 45°C$ and $0 < S < 50$ fit the Setschenow relationship for the variation with salinity. The temperature dependence of the Setschenow coefficient for oxygen is found to be $\kappa = 0.0225034 - 13.6083/T + 2{,}565.68/T^2$. The equation for the Henry coefficient as a function of temperature and salinity is used to calculate values for unit standard atmospheric concentrations (USAC) in freshwater and seawater in equilibrium with air at a total pressure of 1 atmosphere. It is estimated that the possible error in the new USAC values is no greater than ±0.1% and probably less. Tables and equations are presented for obtaining accurate USAC values in the ranges $0° < t < 40°C$ and $0 < S < 40$. Simple procedures are given for calculating standard atmospheric concentrations at pressures different from 1 atm. The presence of sea salt has a negligible effect on the fractionation of the oxygen isotopes during solution.

Benson and Krause (1980*a*) presented new values for the concentration of oxygen in freshwater in equilibrium with the atmosphere in the temperature range 0° to 40°C. These were based upon very precise and accurate measurements (Benson et al. 1979) of the Henry coefficient, $k_{O,0}$, for oxygen dissolved in pure water from 0° to 60°C. The experimental values of $k_{O,0}$, fitted the function

$$\ln k_{O,0} = a_0 + a_1/T + a_2/T^2 \qquad (1)$$

(Benson and Krause 1976) with a precision better than ±0.02% for the full 60°C temperature range. The new concentration values have been recommended (Mortimer 1981) for use in studies involving freshwater.

The work to be reported here extends the results on oxygen to saline waters. New measurements of the Henry coefficient for oxygen in waters with varying salinity are used to examine the functional dependence of $k_{O,S}$ on salinity S, and to determine the way salting-out varies with temperature. Values are derived for the concentration of oxygen dissolved in freshwater and seawater in equilibrium with the atmosphere as a function of temperature, salinity, and atmospheric pressure and compared with the best previous work.

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Determination of the Henry coefficient

The equations of Benson et al. (1979) and Benson and Krause (1980*a*) must be generalized to take account of the presence of sea salt. For a system composed of pure oxygen gas and seawater, Henry's law is

$$f_O = k_{O,S}\frac{n_{O,d}}{n_w + n_s + n_{O,d}}, \qquad (2)$$

where f_O (atm) and $k_{O,S}$ (atm) are the vapor phase fugacity and the Henry coefficient for oxygen, and $n_{O,d}$, n_w, and n_s are the numbers of moles of dissolved oxygen, pure water, and sea salt in the volume V_s (dm^3) of the liquid phase sample bulb. As before, manometric measurements of the dissolved oxygen give

$$n_{O,d} = \frac{P_d V_d}{RT_d Z_d}. \qquad (3)$$

To obtain an expression for the denominator in Eq. 2 in terms of measurable quantities, we note first that

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$$V_s = V_{sw} + \bar{v}_O n_{O,d}, \qquad (4)$$

where V_{sw} (dm^3) is the volume occupied by the seawater sample when it is free of gas, and $\bar{v}_O$ is the partial molal volume of oxygen in the seawater (dm$^3 \cdot$mol^{-1}). Furthermore, because the number of grams of sea salt per kilogram of seawater is given by bS (Millero 1982), where b is in g$\cdot$kg$^{-1} \cdot S^{-1}$ and S is the salinity on the practical scale, the numbers of moles of water and sea salt in volume V_{sw} are given by

$$n_w = \left(\frac{1{,}000 - bS}{M_w}\right)\rho_s V_{sw}, \qquad (5)$$

and

$$n_s = \left(\frac{bS}{M_s}\right)\rho_s V_{sw}, \qquad (6)$$

where ρ_s (kg$\cdot$dm^{-3}) is the density of seawater, M_w (g$\cdot$mol^{-1}) is the gram molecular mass of water, and M_s (g$\cdot$mol^{-1}) is the mean gram molecular mass of sea salt (Millero 1982). When n_w and n_s from Eq. 5 and 6 are added to $n_{O,d}$, and V_{sw} is substituted from Eq. 4, algebraic manipulation leads to

$$n_w + n_s + n_{O,d} = \frac{\rho_s F V'_s}{M_w}, \qquad (7)$$

where

$$F \equiv 1{,}000 - \left(1 - \frac{M_w}{M_s}\right)bS \quad (\text{g}\cdot\text{kg}^{-1}), \qquad (8)$$

and

$$V'_s \equiv V_s\left[1 - \left(\bar{v}_O - \frac{M_w}{\rho_s F}\right)\frac{n_{O,d}}{V_s}\right]. \qquad (9)$$

An argument identical with that of Benson et al. (1979) leads to

$$f_O = \left(\frac{P_u V_u T}{T_u Z_u V_b}\right)(1 - \theta'_O P), \qquad (10)$$

where P_u, V_u, T_u, and Z_u are from the manometric measurements on the dried gas from the vapor phase sample bulb (volume V_b in dm^3). T (K) and P (atm) are the equilibrium temperature and total pressure, respectively, and θ'_O depends upon the second virial coefficients of oxygen and water. Substitution of Eq. 3, 7, and 10 into Eq. 2 yields

$$\left[\frac{RP_u V_u T_d Z_d V'_s}{M_w P_d V_d T_u Z_u V_b}\rho_s TF\right] = k_{O,S}(1 + \theta'_O P), \qquad (11)$$

where R is in atm$\cdot$dm$^3 \cdot$K$^{-1} \cdot$mol^{-1}.

The formal treatment above assumes that the Lewis and Randall rule is applicable (in obtaining Eq. 10) and that satisfactory values for the virial coefficients are available. Benson et al. (1979) pointed out, however, that neither assumption is required if the pressure dependence of the quantity in brackets on the left side of Eq. 11 is determined empirically, as we do. Furthermore, they showed that V'_s can be replaced by V_s with no significant error, especially when the empirical approach is used. Finally, then,

$$k_{O,S} = \left[\frac{RP_u V_u T_d Z_d V_s}{M_w P_d V_d T_u Z_u V_b}\rho_s T\right] \cdot\left[1{,}000 - \left(1 - \frac{M_w}{M_s}\right)bS\right] \cdot(1 - \lambda_O P). \qquad (12)$$

All the quantities in the two brackets are known or measurable, and λ_O can be calculated from eq. 21 of Benson et al. (1979). (The presence of sea salt in the water will have a negligible effect on the real gas corrections for the vapor phase.) When the salinity becomes zero, Eq. 12 reduces essentially to the previous expression for $k_{O,0}$, the Henry coefficient in pure water. (The factor 1,000 is an artifact from the choice of units.)

Equilibrium concentrations and the Henry coefficient

Consider a body of seawater with salinity S in thermodynamic equilibrium with an atmosphere of standard composition saturated with water vapor at a total pressure P (atm). For oxygen Henry's law becomes

$$f_O = k_{O,S}\frac{n_{O,d}}{n_w + n_s + n_{O,d} + n_{r,d}}, \qquad (13)$$

where f_O, $k_{O,S}$, $n_{O,d}$, n_w, and n_s have their previous meanings, and $n_{r,d}$ is the number of moles of all other (the "residual") gases, i.e. essentially nitrogen, dissolved in the volume V_s (dm^3). The standard atmospheric concentration (SAC), by mass, of dissolved oxygen in mol·kg^{-1} is

$$C^P{}_O \equiv \frac{n_{O,d}}{\rho_s V_s}, \qquad (14)$$

or, from Eq. 13,

$$C^P{}_O = \frac{f_O}{k_{O,s}} \frac{n_w + n_s + n_{O,d} + n_{r,d}}{\rho_s V_s}. \qquad (15)$$

For the present situation Eq. 4 is replaced by

$$V_s = V_{sw} + \bar{v}_O n_{O,d} + \bar{v}_r n_{r,d}, \qquad (16)$$

where V_{sw} and $\bar{v}_O$ have been defined above, and $\bar{v}_r$ (dm^3·mol^{-1}) is the effective partial molal volume of the residual gas. When n_w and n_s from Eq. 5 and 6 are added to $n_{O,d}$ plus $n_{r,d}$, and V_{sw} is substituted from Eq. 16, one obtains

$$n_w + n_s + n_{O,d} + n_{r,d} = \frac{\rho_s F V_s Y}{M_w}, \qquad (17)$$

where ρ_s, F, and M_w have their previous meanings, and Y is given by

$$Y \equiv \left[1 - \left(\bar{v}_O - \frac{M_w}{\rho_s F}\right)\frac{n_{O,d}}{V_s} - \left(\bar{v}_r - \frac{M_w}{\rho_s F}\right)\frac{n_{r,d}}{V_s}\right]. \qquad (18)$$

Furthermore, from Benson and Krause (1980*a*)

$$f_O = P_O(1 - \theta_O P), \qquad (19)$$

and

$$P_O = 0.20946(P - P_{wv}), \qquad (20)$$

where P_{wv} is the saturated vapor pressure of water, and θ_O depends upon the second virial coefficient of oxygen. Substituting Eq. 17, 19, and 20 into 15, and recognizing that Y differs from unity by <20 ppm, yields

$$C^P{}_O = 0.20946(P - P_{wv}) \cdot \frac{F}{k_{O,s} M_w}(1 - \theta_O P). \qquad (21)$$

This is the more general form of eq. 27 in Benson and Krause (1980*a*). It differs only in that the salinity factor, F, appears. (The density, ρ_s, is missing here because we have chosen to emphasize concentration per unit mass rather than per unit volume.) With the functions $k_{O,s}(T,S)$, $P_{wv}(T,S)$, $F(S)$, and $\theta_O(T)$ known, $C^P{}_O$ can be calculated from Eq. 21 for any T, P, and S.

It is convenient to separate $C^P{}_O$ into two parts, one dependent on T and S alone and the other primarily on P. The unit standard atmospheric concentration (USAC) by mass, $C^\dagger{}_O(T,S)$, was defined by Benson and Krause (1980*a*) as the concentration of dissolved oxygen per unit mass of seawater when the seawater is in equilibrium with an atmosphere of standard composition and saturated with water vapor at a total pressure (including that of the water vapor) of 1 atm. [Bieri (1974) referred to a similar quantity as the "normal atmospheric equilibrium concentration" (NAEC), although in writing down the related equations he did not take account of real gas behavior in the vapor phase. We prefer the acronyms SAC and USAC, and their associated phrases, because they distinguish between standard concentrations at an arbitrary pressure P and at unit pressure.] It is obtained by setting $P = 1$ atm in Eq. 21:

$$C^\dagger{}_O = 0.20946(1 - P_{wv}) \cdot \frac{F}{k_{O,s} M_w}(1 - \theta_O). \qquad (22)$$

The corresponding USAC by *volume*, $C^*{}_O(T,S)$, is referred to unit volume of seawater (at the equilibrium temperature). It can be determined from

$$C^*{}_O = \rho_s C^\dagger{}_O. \qquad (23)$$

With either USAC known as a function of T and S ($C^\dagger{}_O$ will be used as the example), the value of the corresponding $C^P{}_O$ at any pressure P is given by

$$C^P{}_O = C^\dagger{}_O P\left[\frac{(1 - P_{wv}/P)(1 - \theta_O P)}{(1 - P_{wv})(1 - \theta_O)}\right]. \qquad (24)$$

For use near sea level, the bracket differs little from unity, and a good approximation to $C^P{}_O$ is obtained by simply multiplying $C^\dagger{}_O$ by P (atm). For a more accurate result, especially at high temperature or salinity or

Table 1. Experimental values.

t (°C)	S	P (atm*)	$k_{O,0}$† (atm*)	$k_{O,S}$ (atm*)	% dev. of $k_{O,S}$ from fit
0.231	20.172	0.7754	25,427	29,332	0.054
0.228	31.634	0.7769	25,425	31,793	0.032
0.218	48.667	0.7842	25,418	35,806	−0.071
5.010	20.204	0.7950	28,863	33,097	0.069
5.023	31.703	0.7987	28,873	35,762	0.031
10.017	20.209	0.8131	32,536	37,095	0.091
10.029	31.639	0.8219	32,545	39,830	−0.186
15.009	20.098	0.8408	36,221	40,977	−0.077
15.009	20.459	0.8399	36,221	41,155	0.134
15.011	31.750	0.8468	36,222	44,082	0.026
15.008	48.514	0.8546	36,220	48,917	0.085
25.038	20.089	0.9104	43,488	48,758	−0.056
25.033	31.845	0.9171	43,484	52,139	−0.070
29.993	31.894	0.9402	46,922	55,957	−0.035
35.082	20.228	0.9710	50,288	56,054	0.016
35.081	31.856	0.9749	50,287	59,633	−0.026
35.006	49.478	0.9733	50,239	65,518	0.026
45.007	31.912	1.0464	56,258	66,167	0.019
					0.075 rms

* 1 atmosphere = 101.325 kilopascals.

† Smooth values at zero salinity: Benson et al. 1979.

at a pressure quite different from 1 atm, the bracket should be included.

Experimental measurements with seawater

The experimental method was similar to that used for the solubility of oxygen in distilled water (Benson et al. 1979), except that the apparatus was modified to permit a 250-cm^3 sample of seawater to be withdrawn for measurement of salinity. The salinity sample was taken after the vapor phase and liquid phase samples had been isolated.

The seawater used in these measurements was taken from Buzzards Bay, off Massachusetts, filtered through a 0.45-micron Millipore type HA filter, and stored in the dark before use. Evaporation and dilution with distilled water were used to vary the salinity, which was measured with a Beckman RS7-C salinometer standardized against IAPSO standard seawater and checked at other salinities by quantitative dilution tests. The gas used was Airco research grade oxygen with a total impurity stated to be <30 ppm. Because tests had shown that CO_2 was effectively removed by the liquid nitrogen traps used to dry the oxygen samples, it was unnecessary to eliminate all the CO_2 during degassing of the seawater before an equilibration was begun. Consequently, we could carry out the equilibrations with seawater having essentially normal bicarbonate content. The pH was determined with a Corning model 10 meter and was constant within 8.2 ± 0.1.

The results for $k_{O,S}$ from 18 experiments on seawater samples with salinities up to 50 and at temperatures between 0° and 45°C are summarized in Table 1. The corresponding smooth values for $k_{O,0}$ at each temperature have been calculated from eq. 12 of Benson and Krause (1980*a*). The total pressure P is included for reference, but we did not carry out pressure runs for real gas vapor phase corrections, because as noted earlier, the function $\lambda_O(t)$ determined with pure water (Benson et al. 1979) was applicable here as well. The various quantities needed to calculate $k_{O,S}$ from Eq. 12 are given in Table 2.

We determine $k_{O,S}$ first, because of its direct thermodynamic relationship to the change in free energy. This means that for $k_{O,S}$ the functional dependence upon T and S is both more fundamental and probably simpler than for $C^{\dagger}_O$ or C^{*}_O. Equations 22 and 23 show that the functions $P_{wv}(T,S)$,

Table 2. Parameters for determination of $k_{O,S}$, $C^{\dagger}_O$, C^*_O, and C^P_O (t in °C, T in K).

Source	
Taylor et al. 1969	$R = 82.0562 \times 10^{-3}\ \mathrm{atm \cdot dm^3 \cdot K^{-1} \cdot mol^{-1}}$
Millero 1982	$b = 1.004880\ \mathrm{g \cdot kg^{-1}} \cdot S^{-1}$ $M_w = 18.0153\ \mathrm{g \cdot mol^{-1}}$
Millero 1982	$M_s = 62.7933\ \mathrm{g \cdot mol^{-1}}$ $F = 1{,}000 - \left(1 - \frac{M_w}{M_s} bS\right) = 1{,}000 - 0.716582S$
Benson et al. 1979	$\lambda_O = 0.002805 - 0.0000375t$
Benson et al. 1979	$(1 - \theta_O) = 0.999025 + 1.426 \times 10^{-5}\, t - 6.436 \times 10^{-8}\, t^2$
Benson et al. 1979	$\ln k_{O,0} = 3.71814 + 5{,}596.17/T - 1{,}049{,}668/T^2$
Green and Carritt 1967*a*	$P_{wv} = (1 - 5.370 \times 10^{-4}\, S)\exp\left(18.1973\left(1 - \frac{373.16}{T}\right) + 3.1813 \times 10^{-7}\left\{1 - \exp\left[26.1205\left(1 - \frac{T}{373.16}\right)\right]\right\} - 1.8726 \times 10^{-2}\left\{1 - \exp\left[8.03945\left(1 - \frac{373.16}{T}\right)\right]\right\} + 5.02802 \ln\left(\frac{373.16}{T}\right)\right)$
Millero and Poisson 1981	$\rho_s = \rho_0 + AS + BS^{3/2} + CS^2\ (\mathrm{kg \cdot m^{-3}})$
Millero and Poisson 1981	$\rho_0 = 999.842594 + 6.793952 \times 10^{-2}\, t - 9.095290 \times 10^{-3}\, t^2 + 1.001685 \times 10^{-4}\, t^3 - 1.120083 \times 10^{-6}\, t^4 + 6.536332 \times 10^{-9}\, t^5$
Millero and Poisson 1981	$A = 8.24493 \times 10^{-1} - 4.0899 \times 10^{-3}\, t + 7.6438 \times 10^{-5}\, t^2 - 8.2467 \times 10^{-7}\, t^3 + 5.3875 \times 10^{-9}\, t^4$
Millero and Poisson 1981	$B = -5.72466 \times 10^{-3} + 1.0227 \times 10^{-4}\, t - 1.6546 \times 10^{-6}\, t^2$
Millero and Poisson 1981	$C = 4.8314 \times 10^{-4}$

$\theta_O(T)$, and $\rho_s(T,S)$ play roles in the ways $C^{\dagger}_O$ and C^*_O vary with T and S.

The errors involved in the measurements and calculations have been discussed in detail by Benson et al. (1979) and Benson and Krause (1980*a*). Only a few remarks will be added here. First, it should be pointed out again that the experimental manometric factors in Eq. 12 occur as ratios of pairs of similar quantities. Furthermore, the salinity factor F will contribute no significant error in calculating $k_{O,S}$. (Even an error as large as 0.15 in the salinity would change the calculated value for $k_{O,S}$ by only 0.01%.) Consequently, with M_w, R, ρ_s, T, V_s, V_b, and $(1 - \lambda_O P)$ known very accurately, the values calculated for $k_{O,S}$ are essentially free of uncertainty due to systematic analytical errors. Substitution of Eq. 12 into 22 yields

$$C^{\dagger}_O = 0.20946(1 - P_{wv}) \cdot \frac{(1 - \theta_O)}{(1 - \lambda_O P)}\left[\frac{P_d V_d T_u Z_u V_b}{P_u V_u T_d Z_d V_s}\right] \cdot \frac{1}{R \rho_s T} \qquad (25)$$

for $C^{\dagger}_O$ in terms of the measured quantities. Equation 25 shows that F and M_w have canceled out. Otherwise, the remarks above about systematic analytical errors in $k_{O,S}$ are valid for values of C_O also. In determining values for C_O in freshwater, the most significant uncertainty was associated with the use of $(1 - \theta_O)$ for the real gas correction for oxygen in the mixture of gases making up the atmosphere (Benson and Krause 1980*a*). This remains true here, even though the measurements of the Henry coefficient were less precise for seawater than for distilled water. We estimate that the maximum possible uncertainties in our values for $C^{\dagger}_{O}$ and C^{*}_{O} are no greater than (+0.09, −0.11)% at 0°C and (+0.06, −0.08)% at 45°C.

Variation of the Henry coefficient with temperature and salinity

Equation 1 gives the variation of $k_{O,0}$ with temperature. The remaining problem is to establish the functional dependence of the Henry coefficient on salinity. Following Fox (1909) and Truesdale et al. (1955), Montgomery et al. (1964) assumed that at fixed temperature a variant of C^{*}_{O} (*see* Benson and Krause 1980*a*: p. 669) changed linearly with salinity. Consequently, they measured solubilities only in pure water and seawater of 35.7 salinity. Carpenter (1966) showed that C^{*}_{O} is clearly not linear with S. After trying fits of his data to exponential and quadratic equations in chlorinity, he concluded that the latter yielded better results. Green (1965) and Green and Carritt (1967*b*) reported experiments designed to minimize systematic errors in the determination of the *ratio* of the Bunsen coefficient for oxygen in seawater to that in pure water. Seawater and distilled water were simultaneously equilibrated with air and analyzed under identical conditions. They, too, found that the dependence on S was not linear, but their data for the ratio were well represented by the empirical exponential function commonly used in chemical studies (*see* Pitzer and Brewer 1961) and referred to as the Setschenow (1889) relationship. For nitrogen, Murray et al. (1969) found that the Setschenow relationship fitted their C^{*}_{N} data only moderately well, but the fit was "very much better" with a three-term power series in the square root of chlorinity. In reporting measurements of C^{*}_{O}, Murray and Riley (1969) found again that the quadratic in $(Cl)^{1/2}$ fitted the data closely except at high chlorinities (above 18). In an analytical reexamination of the Carpenter and Murray and Riley data, Weiss (1970) concluded that the data exhibited no significant systematic deviation from the Setschenow relationship, and he adopted it in constructing the equations for the Bunsen coefficient, $C^{\dagger}$, and C^{*} for oxygen (as well as for nitrogen and argon), which have been considered the standards for the past decade.

Table 3. Dependence of the Henry coefficient on salinity. Comparison of linear and logarithmic fits to the present data.

t (°C)	S	$(k_{O,S} - k_{O,0})/S$	$(\ln k_{O,S} - \ln k_{O,0})/S$
0.231	20.172	193.6	0.007082
0.228	31.634	201.3	0.007065
0.218	48.667	213.5	0.007041
15.009	20.278*	239.0*	0.006191*
15.011	31.750	247.6	0.006185
15.008	48.514	259.7	0.006194
35.082	20.228	285.1	0.005367
35.081	31.856	293.4	0.005351
35.006	49.478	308.8	0.005367

* Average of two measurements.

If the Henry coefficient, $k_{O,S}$, were linear with salinity, the quantity $(k_{O,S} - k_{O,0})/S$ would be constant at fixed temperature. The third column in Table 3 shows that at each of the three temperatures our data agree with those of Carpenter, Green, and Murray and Riley in demonstrating that $(k_{O,S} - k_{O,0})/S$ is *not* constant. In fact the three points at each temperature fit the quadratic in S,

$$(k_{O,S} - k_{O,0})/S = g_1 + g_2 S, \quad (26)$$

very well—within the precision of the data. On the other hand, the striking constancy of $(\ln k_{O,S} - \ln k_{O,0})/S$ at each of the temperatures (Table 3, column 4) indicates that the Setschenow relationship,

$$\ln \frac{k_{O,S}}{k_{O,0}} = \kappa S, \quad (27)$$

fits the data just as well. The proportionality of $\ln (k_{O,S}/k_{O,0})$ to S is also shown in Fig. 1. It is not surprising that previous workers had difficulty in distinguishing between the two relationships. Even with the higher precision of the present data, a clearcut choice between the two is not possible. With very precise data at fixed temperatures, and with many different salinities at each temperature, it may be possible in the future to establish one function as preferable to the other and to determine over what range of salinities it is applicable. For the present—and for any practical purpose—the Setschenow relationship is the obvious choice

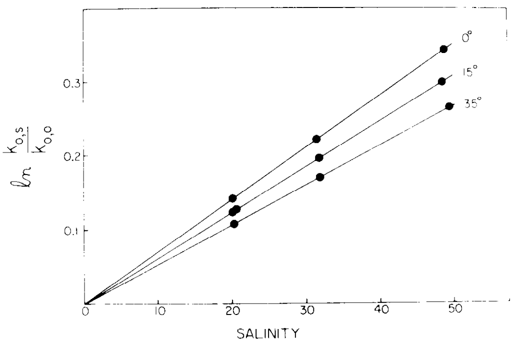

Fig. 1. The dependence of the Henry coefficient on salinity. Fits of the present data at 0°, 15°, and 35°C to the Setschenow relationship.

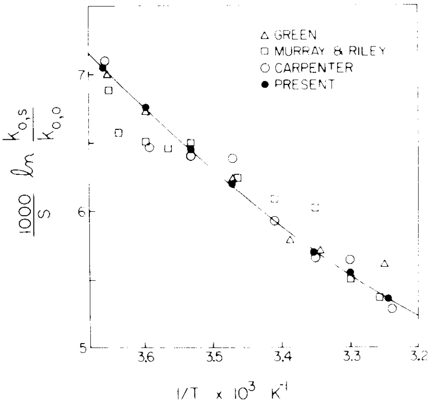

Fig. 2. Variation of the Setschenow coefficient with temperature.

on the basis of simplicity. Not only does it involve only one constant, κ, rather than the two, g_1 and g_2, but with each of the constants being a function of temperature, the temperature dependence of the salting-out would become very complex with the quadratic.

To determine the variation of the salting-out coefficient κ with temperature, we have fitted values of $\ln (k_{O,S}/k_{O,O})$, calculated from Table 1, to S multiplied by a power series in $1/T$. The three-term series yields

$$\kappa = 0.0225034 - 13.6083/T + 2{,}565.68/T^2, \qquad (28)$$

which fits our data very well (Fig. 2).

If we neglect the very small effect of salinity on the vapor pressure of water, it can be shown from Eq. 23 that

$$\kappa \equiv \frac{1}{S} \ln \frac{k_{O,S}}{k_{O,0}} = \frac{1}{S} \ln \frac{C^*_{O,0}}{C^*_{O,S}} + \frac{1}{S} \cdot \ln \left\{ \frac{\rho_s}{\rho_0} \left[1 - \left(1 - \frac{M_w}{M_s} \right) \frac{bS}{1{,}000} \right] \right\}. \qquad (29)$$

[Note that if a Setschenow coefficient were defined in terms of the ratio of the values of C^*, or what is essentially equivalent, the ratio of the Bunsen coefficients—*see* eq. 22: Benson and Krause (1980*a*)—it would differ from the Setschenow coefficient defined here in terms of the ratio of the k values. The dependence of the difference upon salinity and temperature is not simple, but the difference is small and it would introduce no apparent deviation from linearity in a graph like Fig. 1. It would, however, change the slopes in Fig. 1 and slightly rotate the curve in Fig. 2.] We have used the C^*_O data from Green (1965), Carpenter (1966), and Murray and Riley (1969) to calculate from Eq. 29 values for κ which are directly comparable with ours. The smoothed values for $C^*_{O,0}$ of each worker(s) were used with the respective experimental values for $C^*_{O,S}$, so that each of the four sets of numbers is completely independent. Each of the points in Fig. 2 represents the mean value for κ determined from the corresponding data at that temperature. Figure 2 shows that the Setschenow coefficients from the other three sources are in good agreement with our results, especially for the Green and Carpenter data. For each of the three sources, the scatter of points in Fig. 2 relative to our curve is consistent with the precision of deter-

Table 4. Precision of the experimental determination of the Setschenow salting-out coefficient, κ, calculated from various sets of data. (Mean of the percentage standard deviations at different temperatures.)

Source	Mean % σ of κ
Green 1965	2.10
Carpenter 1966	1.81
Murray and Riley 1969	3.10
Present	0.44

Table 5. Values for $C^{\dagger}_{O}$ in μmol·kg^{-1}, at integral temperatures and intervals of five in salinity. To convert from μmol to μg-atoms, cm^3 (STP ideal gas), or mg, one multiplies by 2, 2.2414 × 10^{-2}, or 3.19988 × 10^{-2}.

Temp (°C)	Salinity								
	0	5	10	15	20	25	30	35	40
0.0	457.00	439.55	422.77	406.61	391.07	376.12	361.74	347.90	334.59
1.0	444.31	427.50	411.31	395.73	380.74	366.31	352.42	339.06	326.20
2.0	432.21	415.99	400.38	385.35	370.87	356.94	343.52	330.60	318.17
3.0	420.66	405.01	389.94	375.42	361.44	347.97	335.01	322.52	310.49
4.0	409.63	394.52	379.96	365.93	352.42	339.40	326.86	314.77	303.13
5.0	399.09	384.49	370.42	356.86	343.79	331.19	319.06	307.36	296.09
6.0	389.02	374.90	361.30	348.18	335.53	323.34	311.58	300.25	289.33
7.0	379.38	365.73	352.56	339.86	327.62	315.81	304.42	293.44	282.85
8.0	370.16	356.94	344.19	331.90	320.03	308.59	297.55	286.91	276.64
9.0	361.33	348.53	336.18	324.26	312.76	301.67	290.96	280.63	270.67
10.0	352.86	340.46	328.49	316.94	305.79	295.02	284.64	274.61	264.93
11.0	344.75	332.72	321.12	309.91	299.09	288.64	278.56	268.82	259.42
12.0	336.96	325.30	314.04	303.16	292.66	282.51	272.72	263.26	254.12
13.0	329.49	318.17	307.24	296.68	286.48	276.62	267.10	257.91	249.02
14.0	322.31	311.32	300.70	290.44	280.53	270.95	261.70	252.76	244.12
15.0	315.42	304.74	294.42	284.45	274.81	265.50	256.50	247.80	239.39
16.0	308.79	298.41	288.38	278.68	269.31	260.25	251.49	243.02	234.83
17.0	302.41	292.32	282.56	273.13	264.01	255.19	246.66	238.41	230.43
18.0	296.27	286.45	276.96	267.78	258.90	250.31	242.00	233.97	226.20
19.0	290.35	280.80	271.56	262.62	253.97	245.60	237.51	229.68	222.10
20.0	284.65	275.35	266.35	257.64	249.22	241.06	233.17	225.54	218.15
21.0	279.15	270.10	261.33	252.84	244.63	236.68	228.98	221.53	214.33
22.0	273.85	265.02	256.48	248.20	240.19	232.44	224.93	217.67	210.63
23.0	268.73	260.12	251.79	243.72	235.91	228.34	221.02	213.92	207.05
24.0	263.78	255.39	247.26	239.39	231.76	224.38	217.23	210.30	203.59
25.0	258.99	250.81	242.88	235.19	227.75	220.54	213.55	206.79	200.23
26.0	254.36	246.38	238.63	231.13	223.86	216.82	210.00	203.38	196.98
27.0	249.88	242.08	234.52	227.20	220.10	213.22	206.55	200.08	193.82
28.0	245.54	237.93	230.54	223.38	216.44	209.72	203.20	196.88	190.75
29.0	241.33	233.89	226.68	219.68	212.90	206.32	199.95	193.77	187.77
30.0	237.25	229.98	222.93	216.09	209.46	203.02	196.79	190.74	184.88
31.0	233.29	226.18	219.28	212.59	206.11	199.82	193.72	187.80	182.06
32.0	229.43	222.48	215.74	209.20	202.85	196.70	190.72	184.93	179.31
33.0	225.69	218.89	212.29	205.89	199.68	193.66	187.81	182.14	176.64
34.0	222.04	215.39	208.93	202.67	196.59	190.69	184.97	179.41	174.02
35.0	218.49	211.98	205.66	199.53	193.58	187.80	182.20	176.76	171.48
36.0	215.02	208.65	202.47	196.47	190.64	184.98	179.49	174.16	168.98
37.0	211.65	205.41	199.35	193.48	187.77	182.23	176.84	171.62	166.55
38.0	208.34	202.24	196.31	190.55	184.96	179.53	174.25	169.13	164.16
39.0	205.12	199.14	193.33	187.69	182.21	176.89	171.72	166.70	161.82
40.0	201.96	196.10	190.41	184.88	179.52	174.30	169.23	164.31	159.53

mination of κ by that source as indicated in Table 4.

In summary, our new measurements in seawater, together with those of Benson et al. (1979) in distilled water, show that the Henry coefficient for oxygen varies with temperature and salinity according to

$$\ln k_{O,S} = 3.71814 + 5{,}596.17/T - 1{,}049{,}668/T^2 + S(0.0225034 - 13.6083/T + 2{,}565.68/T^2). \qquad (30)$$

The precision of fit of our k values to this expression is 0.017% for the distilled water values and 0.075% for the seawater values (*see Table 1*).

Dependence of $C^{\dagger}_{O}$ *and* C^{*}_{O} *on temperature and salinity*

Values for the two USACs may be found by substituting values for $k_{O,S}$ from Eq. 30, and parameters from Table 2, into Eq. 22 and 23. As noted earlier, we believe that the uncertainties in the resulting smooth values

Table 6. Values for C^*_O in μmol·dm^{-3} at integral temperatures and intervals of five in salinity. For conversion to other units, *see Table 5.*

Temp (°C)	Salinity 0	5	10	15	20	25	30	35	40
0.0	456.93	441.27	426.13	411.49	397.34	383.66	370.45	357.68	345.34
1.0	444.27	429.19	414.59	400.48	386.83	373.64	360.89	348.57	336.66
2.0	432.19	417.65	403.58	389.96	376.80	364.06	351.75	339.85	328.35
3.0	420.65	406.62	393.05	379.91	367.20	354.90	343.01	331.51	320.39
4.0	409.62	396.09	382.98	370.29	358.02	346.13	334.64	323.52	312.76
5.0	399.08	386.01	373.35	361.09	349.22	337.74	326.62	315.86	305.46
6.0	389.00	376.37	364.13	352.28	340.81	329.69	318.94	308.53	298.45
7.0	379.35	367.14	355.31	343.84	332.74	321.98	311.57	301.49	291.72
8.0	370.10	358.30	346.85	335.75	325.00	314.59	304.50	294.73	285.27
9.0	361.25	349.82	338.73	327.99	317.58	307.49	297.71	288.24	279.07
10.0	352.76	341.69	330.95	320.54	310.45	300.67	291.20	282.01	273.11
11.0	344.61	333.89	323.49	313.40	303.61	294.13	284.93	276.02	267.38
12.0	336.79	326.40	316.31	306.53	297.04	287.84	278.91	270.26	261.87
13.0	329.28	319.21	309.42	299.93	290.72	281.78	273.12	264.71	256.56
14.0	322.07	312.29	302.79	293.58	284.64	275.96	267.54	259.37	251.45
15.0	315.13	305.64	296.42	287.47	278.78	270.35	262.17	254.23	246.53
16.0	308.46	299.24	290.29	281.59	273.15	264.95	256.99	249.27	241.78
17.0	302.04	293.08	284.38	275.92	267.71	259.74	252.00	244.49	237.20
18.0	295.85	287.14	278.68	270.46	262.48	254.72	247.19	239.88	232.78
19.0	289.89	281.42	273.19	265.19	257.42	249.88	242.54	235.42	228.51
20.0	284.14	275.90	267.89	260.11	252.55	245.20	238.06	231.12	224.38
21.0	278.59	270.58	262.78	255.20	247.84	240.68	233.72	226.96	220.39
22.0	273.24	265.44	257.84	250.46	243.28	236.31	229.53	222.94	216.53
23.0	268.07	260.46	253.07	245.87	238.88	232.08	225.47	219.04	212.79
24.0	263.07	255.66	248.45	241.44	234.62	227.99	221.54	215.27	209.17
25.0	258.23	251.01	243.98	237.15	230.49	224.02	217.73	211.61	205.66
26.0	253.55	246.51	239.66	232.99	226.50	220.18	214.04	208.07	202.26
27.0	249.01	242.15	235.46	228.95	222.62	216.46	210.46	204.63	198.96
28.0	244.62	237.92	231.40	225.04	218.86	212.84	206.99	201.29	195.74
29.0	240.36	233.82	227.45	221.25	215.21	209.33	203.61	198.04	192.62
30.0	236.22	229.84	223.62	217.56	211.66	205.92	200.33	194.89	189.59
31.0	232.20	225.97	219.89	213.97	208.21	202.60	197.13	191.81	186.63
32.0	228.29	222.20	216.27	210.49	204.85	199.37	194.02	188.82	183.75
33.0	224.49	218.54	212.74	207.09	201.58	196.22	190.99	185.90	180.95
34.0	220.79	214.98	209.31	203.78	198.39	193.15	188.04	183.06	178.21
35.0	217.18	211.50	205.96	200.55	195.29	190.15	185.15	180.28	175.53
36.0	213.67	208.11	202.69	197.40	192.25	187.23	182.33	177.56	172.92
37.0	210.23	204.80	199.50	194.32	189.28	184.37	179.58	174.91	170.36
38.0	206.88	201.56	196.37	191.31	186.38	181.57	176.88	172.31	167.85
39.0	203.60	198.40	193.32	188.37	183.54	178.83	174.24	169.76	165.40
40.0	200.39	195.30	190.33	185.48	180.76	176.15	171.65	167.26	162.99

for C_O are no greater (and probably less) than (+0.09, −0.11)% at 0°C and (+0.06, −0.08)% at 45°C over the 0–50 salinity range of the measurements.

For convenience, values for $C^\dagger_O$ and C^*_O are given in Tables 5, 6, and 7 at integral temperatures from 0° to 40°C and at intervals of five from 0 to 40 in salinity. Linear interpolation in either temperature or salinity will introduce errors <0.02%. Combined interpolations can lead to errors as large as 0.035%. In the three tables we have emphasized μmol·kg^{-1}, μmol·dm^{-3}, and mg·liter^{-1}, but factors for conversion to other units are provided. (For the present purposes, the liter and the cubic decimeter are identical. We use mg·liter^{-1} in Table 7 to conform with standard notation in sanitation engineering.)

For computer and programmable calculator applications, without the complexity associated with direct use of the basic Eq. 22 (or 23) with Eq. 30 and Table 2, we have fitted the values in Tables 5 and 6 to equa-

Table 7. Values for C^*_O in mg·liter^{-1} at integral temperatures and intervals of five in salinity.

Temp (°C)	Salinity								
	0	5	10	15	20	25	30	35	40
0.0	14.621	14.120	13.636	13.167	12.714	12.277	11.854	11.445	11.051
1.0	14.216	13.733	13.266	12.815	12.378	11.956	11.548	11.154	10.773
2.0	13.829	13.364	12.914	12.478	12.057	11.650	11.256	10.875	10.507
3.0	13.460	13.012	12.577	12.157	11.750	11.356	10.976	10.608	10.252
4.0	13.107	12.674	12.255	11.849	11.456	11.076	10.708	10.352	10.008
5.0	12.770	12.352	11.947	11.554	11.175	10.807	10.452	10.107	9.774
6.0	12.448	12.043	11.652	11.273	10.905	10.550	10.206	9.872	9.550
7.0	12.139	11.748	11.369	11.003	10.647	10.303	9.970	9.647	9.335
8.0	11.843	11.465	11.099	10.744	10.400	10.066	9.744	9.431	9.128
9.0	11.560	11.194	10.839	10.495	10.162	9.839	9.526	9.223	8.930
10.0	11.288	10.934	10.590	10.257	9.934	9.621	9.318	9.024	8.739
11.0	11.027	10.684	10.351	10.028	9.715	9.412	9.118	8.832	8.556
12.0	10.777	10.444	10.122	9.809	9.505	9.210	8.925	8.648	8.379
13.0	10.537	10.214	9.901	9.597	9.303	9.017	8.739	8.471	8.210
14.0	10.306	9.993	9.689	9.394	9.108	8.830	8.561	8.300	8.046
15.0	10.084	9.780	9.485	9.199	8.921	8.651	8.389	8.135	7.889
16.0	9.870	9.575	9.289	9.010	8.740	8.478	8.224	7.976	7.737
17.0	9.665	9.378	9.100	8.829	8.567	8.311	8.064	7.823	7.590
18.0	9.467	9.188	8.917	8.654	8.399	8.151	7.910	7.676	7.449
19.0	9.276	9.005	8.742	8.486	8.237	7.996	7.761	7.533	7.312
20.0	9.092	8.829	8.572	8.323	8.081	7.846	7.618	7.396	7.180
21.0	8.915	8.658	8.409	8.166	7.930	7.701	7.479	7.262	7.052
22.0	8.743	8.494	8.251	8.014	7.785	7.562	7.345	7.134	6.929
23.0	8.578	8.335	8.098	7.868	7.644	7.426	7.215	7.009	6.809
24.0	8.418	8.181	7.950	7.726	7.508	7.295	7.089	6.888	6.693
25.0	8.263	8.032	7.807	7.588	7.376	7.169	6.967	6.771	6.581
26.0	8.113	7.888	7.669	7.455	7.248	7.046	6.849	6.658	6.472
27.0	7.968	7.748	7.535	7.326	7.124	6.926	6.735	6.548	6.366
28.0	7.828	7.613	7.404	7.201	7.003	6.811	6.623	6.441	6.264
29.0	7.691	7.482	7.278	7.080	6.886	6.698	6.515	6.337	6.164
30.0	7.559	7.354	7.155	6.962	6.773	6.589	6.410	6.236	6.067
31.0	7.430	7.231	7.036	6.847	6.662	6.483	6.308	6.138	5.972
32.0	7.305	7.110	6.920	6.735	6.555	6.379	6.209	6.042	5.880
33.0	7.183	6.993	6.807	6.627	6.450	6.279	6.112	5.949	5.790
34.0	7.065	6.879	6.698	6.521	6.348	6.181	6.017	5.858	5.702
35.0	6.950	6.768	6.590	6.417	6.249	6.085	5.925	5.769	5.617
36.0	6.837	6.659	6.486	6.317	6.152	5.991	5.834	5.682	5.533
37.0	6.727	6.553	6.384	6.218	6.057	5.900	5.746	5.597	5.451
38.0	6.620	6.450	6.284	6.122	5.964	5.810	5.660	5.514	5.371
39.0	6.515	6.349	6.186	6.028	5.873	5.722	5.575	5.432	5.293
40.0	6.412	6.249	6.090	5.935	5.784	5.636	5.493	5.352	5.215

tions comparable to eq. 26 and 25 of Benson and Krause (1980*a*), with additional terms of the Setschenow form in salinity. For $C^\dagger_O$ in μmol·kg^{-1},

$$\ln C^\dagger_O = -135.29996 + 1.572288 \times 10^5/T - 6.637149 \times 10^7/T^2 + 1.243678 \times 10^{10}/T^3 - 8.621061 \times 10^{11}/T^4 - S(0.020573 - 12.142/T + 2{,}363.1/T^2). \quad (31)$$

To obtain $C^\dagger_O$ in cm^3 (STP ideal gas)·kg^{-1} or mg·kg^{-1}, we replace the first constant by (−139.09803) or (−138.74202). Similarly, for C^*_O in μmol·dm^{-3},

$$\ln C^*_O = -135.90205 + 1.575701 \times 10^5/T - 6.642308 \times 10^7/T^2 + 1.243800 \times 10^{10}/T^3 - 8.621949 \times 10^{11}/T^4 - S(0.017674 - 10.754/T + 2{,}140.7/T^2). \quad (32)$$

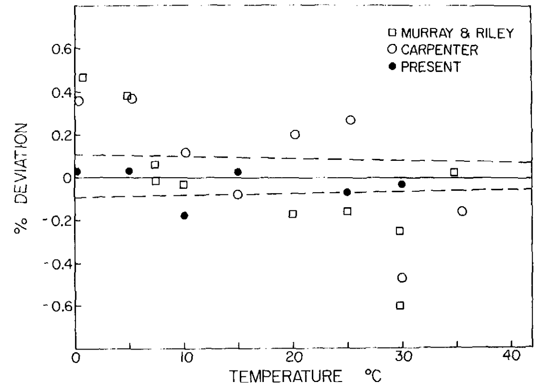

Fig. 3. Percentage deviations from the present smooth values of experimental values for $C^{\dagger}_{O}$ or C^{*}_{O} in the salinity range 30–40.

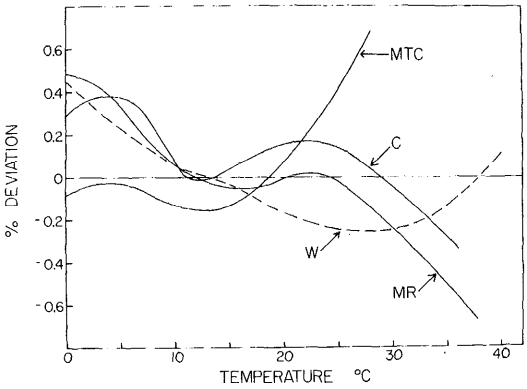

Fig. 4. Comparison of the temperature dependence of the smooth values for $C^{\dagger}_{O}$ or C^{*}_{O} from Montgomery et al. 1964 (MTC), Carpenter 1966 (C), Murray and Riley 1969 (MR), and Weiss 1970 (W), with the present smooth values at approximately 35 salinity.

When C^{*}_{O} is desired in cm^3 (STP ideal gas)·dm^{-3} we substitute (−139.70012) for the first constant in Eq. 32. For C^{*}_{O} in mg·liter^{-1} (equivalent to Table 7) we substitute (−139.34411) for the first constant.

The fit of these equations to the three tables is very good. The numbers obtained never disagree with those in the tables by more than 2 in the last place. It is important, however, that the equations *not* be used except when $0° < t < 40°\text{C}$ and $0 < S < 40$. Beyond changes associated with different units, the first constants in Eq. 31 and 32 differ a little from the corresponding ones in eq. 26 and 25 of Benson and Krause (1980*a*). Although we recommend the new values because the fit is improved very slightly, use of the previous constants will have introduced no significant errors.

Figure 3 shows the deviations from our smooth results of three sets of experimental values for $C^{\dagger}_{O}$ or C^{*}_{O} in the salinity range $30 < S < 40$. The Carpenter (1966) and Murray and Riley (1969) values have been converted to correspond with ours by the procedures described by Benson and Krause (1980*a*: p. 669). (The Montgomery et al. 1964 results at 35.7 salinity could not be included here, because they did not report their experimental data.) The deviations from our smooth results of the four Carpenter and Murray and Riley points at 0°C and 5°C seem to be systematic, but—in fact—they are consistent with the random scatter in those data. This is suggested in Fig. 3 by their points from 25°C to 35°C, but becomes more evident from examination of fig. 1 of Benson and Krause (1980*a*) and our present Fig. 2. Our estimate of the maximum possible range of uncertainty in our smooth function is indicated by the dashed lines. Another comparison of the experimental data with the smooth fit is given in Table 8.

Table 8. Comparison of deviations of experimental data for $C^{\dagger}_{O}$ or C^{*}_{O} for S between 30 and 40, from present smooth values.

Source	$\frac{\Sigma\ \%\ \text{dev.}}{n}$	$\left[\frac{\Sigma\ (\%\ \text{dev.})^2}{n}\right]^{1/2}$
Carpenter 1966	0.08	0.28
Murray and Riley 1969	−0.03	0.29
Present	−0.026	0.074

The deviations from our smooth results of smooth values for $C^{\dagger}_{O}$ or C^{*}_{O} from several other sources at approximately 35 salinity are shown in Fig. 4 as a function of temperature. (Again, all C_{O} values have been made equivalent; cf. Benson and Krause 1980*a*.) In this region of greatest interest for work in the open oceans, the Weiss fit of the Carpenter and Murray and Riley data is 0.4% high at 0°C and about 0.2% low at 25°C. The current UNESCO values (Postma et al. 1976) are based on Weiss' results, but

Table 9. Values for the quantity bracketed in Eq. 24 vs. P and t. No corrections have been made for the effect of salinity on P_{wv}; for salinities <40 this causes errors <0.03% in the numbers not in parentheses and <0.05% in those in parentheses. Linear interpolation in P and t (in either order) will introduce errors <0.02% with the numbers not in parentheses and <0.05% with those in parentheses.

t (°C)	P (atm) 1.1	1.0	0.9	0.8	0.7	0.6	0.5
0.0	1.0005	1.0000	0.9994	0.9987	0.9977	0.9963	0.9944
5.0	1.0007	1.0000	0.9991	0.9980	0.9966	0.9946	0.9918
10.0	1.0010	1.0000	0.9987	0.9971	0.9950	0.9922	0.9882
15.0	1.0015	1.0000	0.9982	0.9959	0.9929	(0.9889)	(0.9833)
20.0	1.0021	1.0000	0.9974	0.9942	(0.9901)	(0.9845)	
25.0	1.0029	1.0000	0.9965	0.9921	(0.9864)	(0.9787)	
30.0	1.0039	1.0000	0.9952	(0.9892)	(0.9814)		
35.0	1.0053	1.0000	(0.9935)	(0.9854)			
40.0	1.0071	1.0000	(0.9913)	(0.9805)			

are systematically lower than his by 0.1% for reasons discussed earlier (Benson and Krause 1980*a*).

Calculation of standard atmospheric concentrations at arbitrary pressure

Equation 24 provides a simple method for finding $C^P{}_O$. Calculations show that the bracket in Eq. 24 depends very little upon salinity. (Both $C^P{}_O$ and $C^{\dagger}{}_O$ vary with salinity, of course, but their *ratio* is nearly independent of it.) Consequently, except for unusual situations, the values for the bracket given by Benson and Krause (1980*a*) can be used for seawater as well as freshwater. Table 9 is a simplified version of their table. For the common oceanographic regimes ($1.1 > P > 0.9$ atm, $0° < t < 30°C$, and $S < 40$), the maximum correction (at $P = 0.9$, $t = 30°C$) provided by the bracket is 0.48%. This means that for oceanographic purposes, simply multiplying $C^{\dagger}{}_O$ or $C^*{}_O$ by P to find the corresponding $C^P{}_O$ may lead to an error of about 0.5%, but when the bracket is included the error in the calculated value for $C^P{}_O$ will be <0.03%, even when the effect of salinity on the bracket is neglected. In regions where low atmospheric pressures are encountered, the bracket correction becomes larger, but the water usually will be relatively fresh, and the lack of correction for S in Table 9 will be insignificant. Only for regimes in which the pressure is quite different from 1 atmosphere and the temperature, salinity, or both are high, is it necessary to calculate the values for the bracket directly from Table 2.

Isotopic fractionation of atmospheric oxygen during solution

We have examined the variation of the isotopic fractionation of helium with salinity (Benson and Krause 1980*b*). With very high salt concentrations the fractionation is increased over that in freshwater by 1.5 to 2.5 per mil, depending on temperature, but in the range of oceanographic salinities, the fractionation of helium is increased by only 0.2 to 0.3 per mil. This is measurable (under some circumstances) for helium, but because the isotopic fractionation of oxygen is smaller than that of helium by a factor of about 20, the effect of salinity on the oxygen fractionation should be undetectable in the range $0 < S < 40$. Consequently, the equation given by Benson and Krause (1980*a*) should be valid for seawater as well:

$$\delta = -0.730 + (427/T), \qquad (33)$$

where T is in kelvin, and δ is the per mil difference between the $^{34}O_2$:$^{32}O_2$ abundance ratio in the dissolved gas and that abundance ratio in atmospheric oxygen, when the water is in *equilibrium* with air.

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