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**On the pH-buffering effects of the
CO₂-CO₃²⁻-system in deep
groundwaters**

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1982-12-10

SVENSK KÄRNBRÄNSLEFÖRSÖRJNING AB / AVDELNING KBS

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 $\text{CO}_2\text{-CO}_3^{2-}$ SYSTEM IN DEEP GROUNDWATERS

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SUMMARY

Equilibria in the system $H_2O-CO_2-CaCO_3(s)$ are discussed and data from some deep granitic groundwaters are summarized. For most waters pH would be in the range 7-9 and the total carbonate concentration in the range 30-400 mg/l (more often 90-275 mg/l). The observed field data represent reasonably well closed water systems with fixed total carbonate concentrations.

The carbonate concentration (CO_3^{2-}) can be estimated from $\log [CO_3^{2-}] = 0.76pH - 10.83 \pm 0.08$, which would represent about 50% of all the observed concentration/pH-data (for the waters with the highest carbonate concentrations).

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1. CARBONATE EQUILIBRIA IN NATURAL WATERS

In this study the system $H_2O-CO_2-CaCO_3(s)$ is briefly described in order to illustrate how pH is affected by reactions in this system and to define what pH and carbonate concentrations would be expected in deep granitic groundwaters.

The carbonate system has been thoroughly described in the literature. The subsequent discussion and calculated cases are largely taken from ref. 1 and 2.

1.1. Reactions and thermodynamic constants

The presence of carbonate due to equilibria in the system $CO_2-H_2CO_3-HCO_3^- -CO_3^{2-}$ and the precipitation of $CaCO_3(s)$ are the major pH-buffering mechanisms in most natural waters. The following reactions are of importance:



By combining reaction (1) and (2) and neglecting (5) and (6), the following equations will be obtained, defining the CO_3^{2-} -system in most natural waters:

$$[H_2CO_3]/P(CO_2) = K' \quad (8)$$

$$[H^+][CO_3^{2-}]/[H_2CO_3] = K_1 \quad (9)$$

$$[H^+][CO_3^{2-}]/[HCO_3^-] = K_2 \quad (10)$$

$$[H^+][OH^-] = K_w \quad (11)$$

$$[Ca^{2+}][CO_3^{2-}] = K_s \quad (12)$$

Values of the constants are given in Table 1.

Table 1 Stability constants in the carbonate system

	I = 0, 25 ⁰ C	I = 4x10 ⁻³ , 10 ⁰ C
log K'	-1.47	-1.27
log K ₁	-6.35	-6.43
log K ₂	-10.33	-10.38
log K _s	-8.42	-7.95

1.2. The carbonate system in granitic groundwaters

1.2.1. Total carbonate concentrations

The following concentrations of CO₂ and total CO₃²⁻ would be expected in nature (typical ranges):

Air	(2.9-3.3)x10 ⁻⁴ atm
Rain water	0.6-3 mg/l
Surface water	6-300 mg/l
Groundwaters	30-485 mg/l

In granitic groundwaters the total carbonate is usually in the concentration range 60-250 mg/ml. In a few cases concentrations as low as 20 mg/l, and up to 400 mg/l have been found (3-6).

In the subsequent calculations the following total carbonate concentrations (C_T) have been assumed as possible minimum and maximum concentrations in granitic groundwaters:

$$(C_T)_{\min} = 19 \text{ mg/l } (= 10^{-3.5} \text{ M})$$

$$(C_T)_{\max} = 485 \text{ mg/l } (= 10^{-2.1} \text{ M})$$

1.2.2 Total calcium concentrations

Usually the calcium concentration is between 5 and 50 mg/l,

and seldom above 100 mg/l, in granitic groundwaters. In a few cases, however, very high calcium concentrations (over 1000 mg/l; ca 1800 mg/l in one case), far above the calcium concentration in sea water (ca 390 mg/l) have been observed.

In the subsequent analysis, the following calcium concentrations (C_M) will be used, representing likely minimum and maximum levels:

$$\begin{aligned}(C_M)_{\min} &= 5.0 \text{ mg/l} \quad (= 10^{-3.9} \text{ M}) \\ (C_M)_{\max} &= 400 \text{ mg/l} \quad (= 10^{-2.0} \text{ M})\end{aligned}$$

1.2.3 pH-range

For most deep groundwaters pH would be in the interval 7.2-8.5, and seldom below 7 or above 9. In a few cases, however, pH as low as 6.3 has been recorded, as well as above 10 in others.

The following minimum and maximum pH-values are probable:

$$\begin{aligned}\text{pH}_{\min} &= 6.0 \\ \text{pH}_{\max} &= 10.5\end{aligned}$$

2. CALCULATED pH- AND CONCENTRATION RANGES

The pH that would be obtained by dissolution of $\text{CaCO}_3(\text{s})$ is calculated and discussed below for some cases of environmental interest.

2.1 Dissolution of $\text{CaCO}_3(\text{s})$ in pure water

No initial CO_2 -pressure is assumed, and the gas phase after equilibrium is negligible. Thus, pH, $[\text{Ca}^{2+}]$ and total $[\text{CO}_3^{2-}]$ are entirely determined by the dissolution of $\text{CaCO}_3(\text{s})$.

This case will simply be characterized by eqn (9-12) and the mass balances

$$\begin{aligned} [\text{Ca}^{2+}] &= [\text{CO}_3^{2-}] + [\text{HCO}_3^-] + [\text{H}_2\text{CO}_3^*] \\ 2[\text{Ca}^{2+}] + [\text{H}^+] &= 2[\text{CO}_3^{2-}] + [\text{HCO}_3^-] + [\text{OH}^-] \end{aligned}$$

The following equilibrium concentrations are obtained at $I = 0$, 25°C (c.f. Table 1):

$$\begin{aligned} [\text{Ca}^{2+}] &= 10^{-3.9} && (5.1 \text{ mg/l}) \\ [\text{HCO}_3^-] &= 10^{-4.05} \\ [\text{CO}_3^{2-}] &= 10^{-4.4} && (C_T = 7.8 \text{ mg/l}) \\ \text{pH} &= 9.9 \end{aligned}$$

This system is normally not representative of conditions in natural groundwaters.

2.2 Dissolution of $\text{CaCO}_3(\text{s})$ in water with a constant external CO_2 -pressure

An external constant CO_2 -pressure is assumed, e.g. $P(\text{CO}_2) = 10^{-3.5}$ atm (equilibrium with the atmosphere).

This case is characterized by eqn (9-12) and the mass balance

$$2[\text{Ca}^{2+}] + [\text{H}^+] = 2[\text{CO}_3^{2-}] + [\text{HCO}_3^-] + [\text{OH}^-]$$

The following equilibrium concentrations are obtained at $I = 0$, 25°C :

$$\begin{aligned} [\text{Ca}^{2+}] &= 10^{-3.4} && (16.0 \text{ mg/l}) \\ [\text{HCO}_3^-] &= 10^{-3.0} \\ [\text{CO}_3^{2-}] &= 10^{-4.9} && (C_T = 61.8 \text{ mg/l}) \\ \text{pH} &= 8.4 \end{aligned}$$

This system is representative of an aqueous system open to the atmosphere and saturated with respect to $\text{CaCO}_3(\text{s})$.

2.3 Dissolution of $\text{CaCO}_3(\text{s})$ in a closed water system with a fixed total carbonate concentration

This case is representative of a closed groundwater system, where total carbonate and pH are given. Saturation with respect to $\text{CaCO}_3(\text{s})$ is assumed.

The system is characterized by eqn (9-12) and the mass balances

$$\begin{aligned} [\text{H}_2\text{CO}_3] + [\text{HCO}_3^-] + [\text{CO}_3^{2-}] &= \text{const. (analysis)} \\ [\text{H}^+] &= 10^{-\text{pH}} \quad (\text{analysis}) \end{aligned}$$

This would be the most common case encountered in natural systems.

The system is illustrated in Figure 1.

2.4 Dissolution of $\text{CaCO}_3(\text{s})$ in a closed water system with equal total carbonate and calcium concentrations

This case essentially illustrates the solubility of $\text{CaCO}_3(\text{s})$ in a closed system of various pH.

The system is characterized by eqn (9-12) and the mass balances

$$\begin{aligned} [\text{Ca}^{2+}] &= [\text{CO}_3^{2-}] + [\text{HCO}_3^-] + [\text{H}_2\text{CO}_3] \\ [\text{H}^+] &= 10^{-\text{pH}} \quad (\text{analysis}) \end{aligned}$$

and is illustrated in Figure 2.

2.5. Dissolution of $\text{CaCO}_3(\text{s})$ in a closed system with a CO_2 reservoir

This case illustrates the dissolution of $\text{CaCO}_3(\text{s})$ either in a closed system with a reservoir of constant $P(\text{CO}_2)$ or a system that becomes separated from such a reservoir, Figure 3.

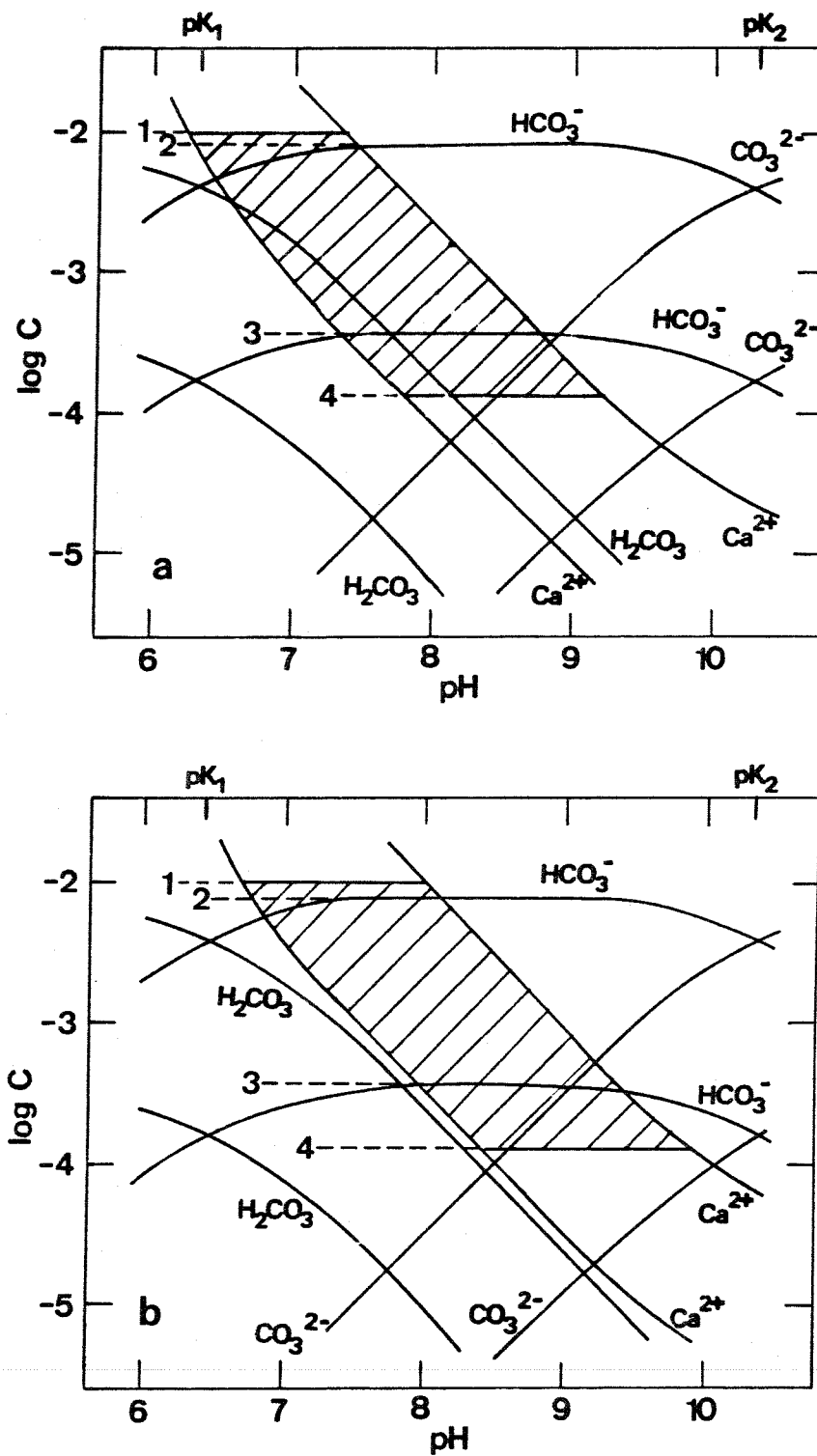


Figure 1 Dissolution of $\text{CaCO}_3(\text{s})$ in a closed water system with a fixed total carbonate concentration. (a) $I=0$, 25°C , (b) $I=4 \times 10^{-3}$, 10°C .

1: $(C_M)_{\text{max}}$, 2: $(C_T)_{\text{max}}$, 3: $(C_T)_{\text{min}}$, 4: $(C_M)_{\text{min}}$

Shaded areas indicate possible pH-ranges at equilibrium.

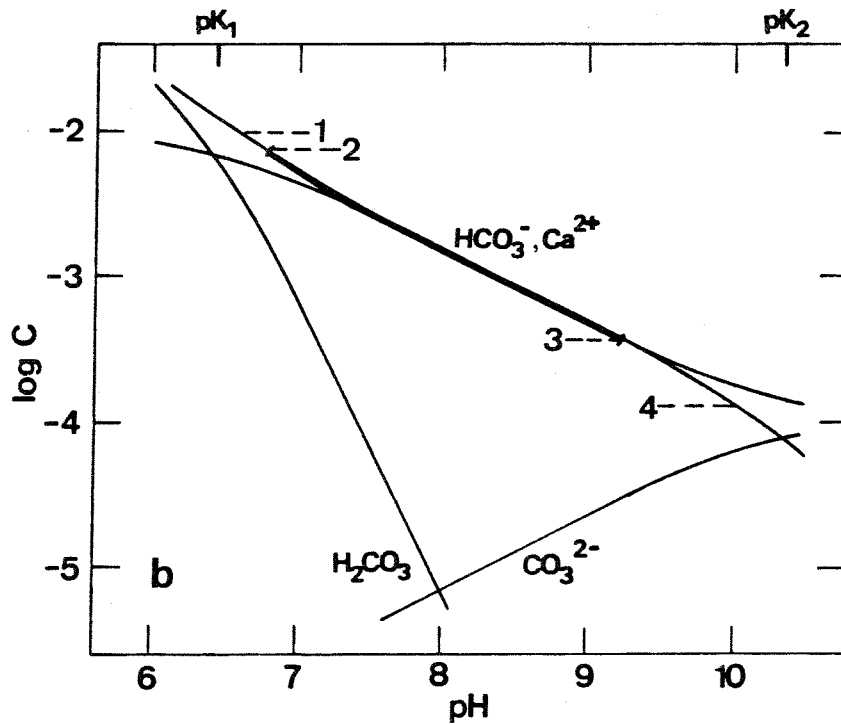
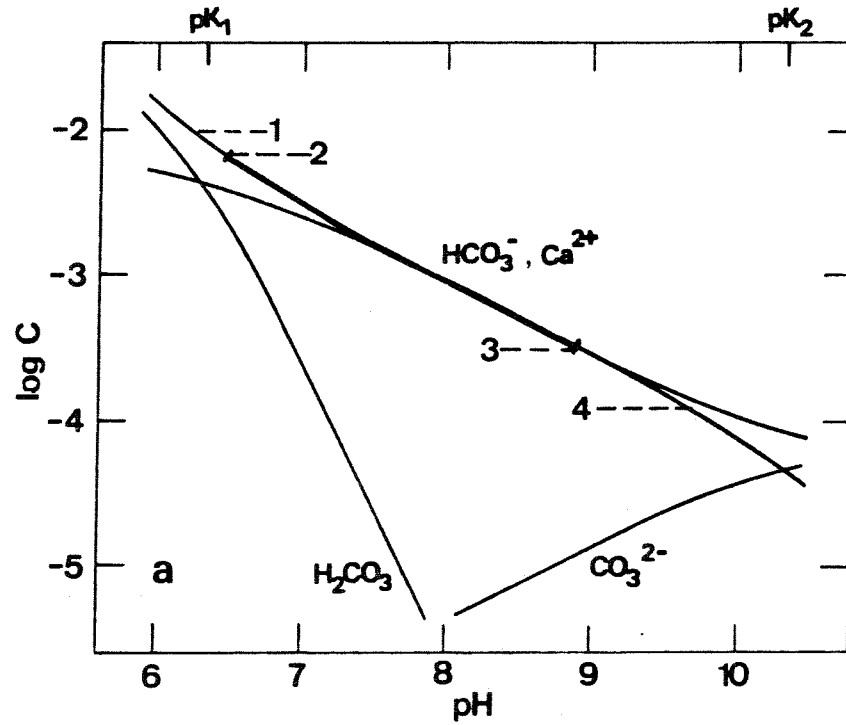


Figure 2 Dissolution of $\text{CaCO}_3(\text{s})$ in a closed water system with equal total carbonate and calcium concentrations. (a) $I=0$, 25°C , (b) $I=4 \times 10^{-3}$, 10°C .

1: $(C_M)_{\text{max}}$, 2: $(C_T)_{\text{max}}$, 3: $(C_T)_{\text{min}}$, 4: $(C_M)_{\text{min}}$.

Thick lines indicate possible pH-ranges at equilibrium.

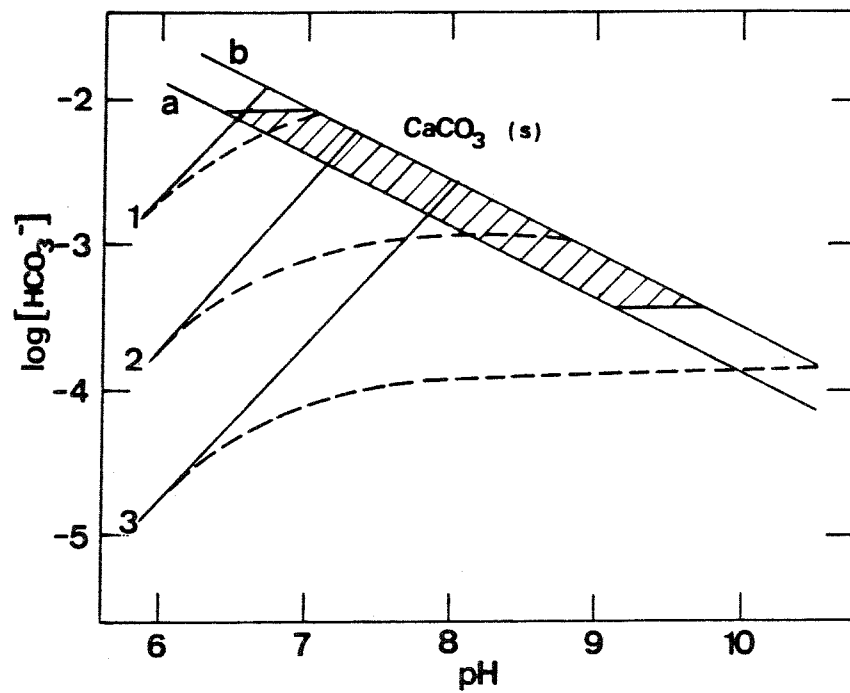


Figure 3 Dissolution of $\text{CaCO}_3(\text{s})$ in systems with a reservoir of CO_2 .

Solid lines: Constant $P(\text{CO}_2)$.

Dashed lines: System with enclosed CO_2 .

(a) $I=0$, 25°C , (b) $I=4 \times 10^{-3}$, 10°C .

1: $P(\text{CO}_2)=10^{-1}$, 2: $P(\text{CO}_2)=10^{-2}$, 3: $P(\text{CO}_2)=10^{-3}$ (atm)

Shaded area indicates possible pH-ranges at equilibrium.

The $\text{CaCO}_3(\text{s})$ equilibrium line in Figure 3 corresponds to

$$\log [\text{HCO}_3^-] = 0.5(\log K_s - \log K_2 - 2\text{pH}) \quad (13)$$

generated from eqn (9-12).

3. OBSERVED pH- AND CONCENTRATION RANGES

The calculated pH-ranges in CaCO_3 -saturated waters according to 2.1-2.5 above are summarized in Table 2, assuming $[\text{Ca}^{2+}]$ and total $[\text{CO}_3^{2-}]$ -ranges as given in 1.2.

Table 2 Calculated pH-values in $\text{CaCO}_3(\text{s})$ -saturated water systems

System	pH
(Pure water	9.9)
Open system, $P(\text{CO}_2)=10^{-3.5}$ atm	8.4
Closed system, constant C_T	6.3 - 9.9
Closed system, $C_T = C_M$	6.4 - 9.3
Closed system with CO_2 reservoir	6.4 - 9.7

In Figure 4 measured pH and HCO_3^- -concentration data are given for granitic groundwaters (from Finnsjön, Kråkemåla, Sternö, Svartboberget, Gideå, Fjällveden) (2-5) (C.f. Figure 1 and 3).

4. DISCUSSION AND CONCLUSIONS

From the observed field data and the calculated data for $\text{CaCO}_3(\text{s})$ -saturated systems the following observations can be made:

- o Most of the groundwaters are near saturation with respect to $\text{CaCO}_3(\text{s})$; many systems appear to be over-saturated.
- o The $\text{H}_2\text{O}-\text{CO}_2-\text{CaCO}_3$ -system buffers pH to the range 6.5-10, where a high pH would correspond to a low total carbonate concentration and often also a low calcium concentration; a low pH would correspond to high carbonate and calcium concentrations.

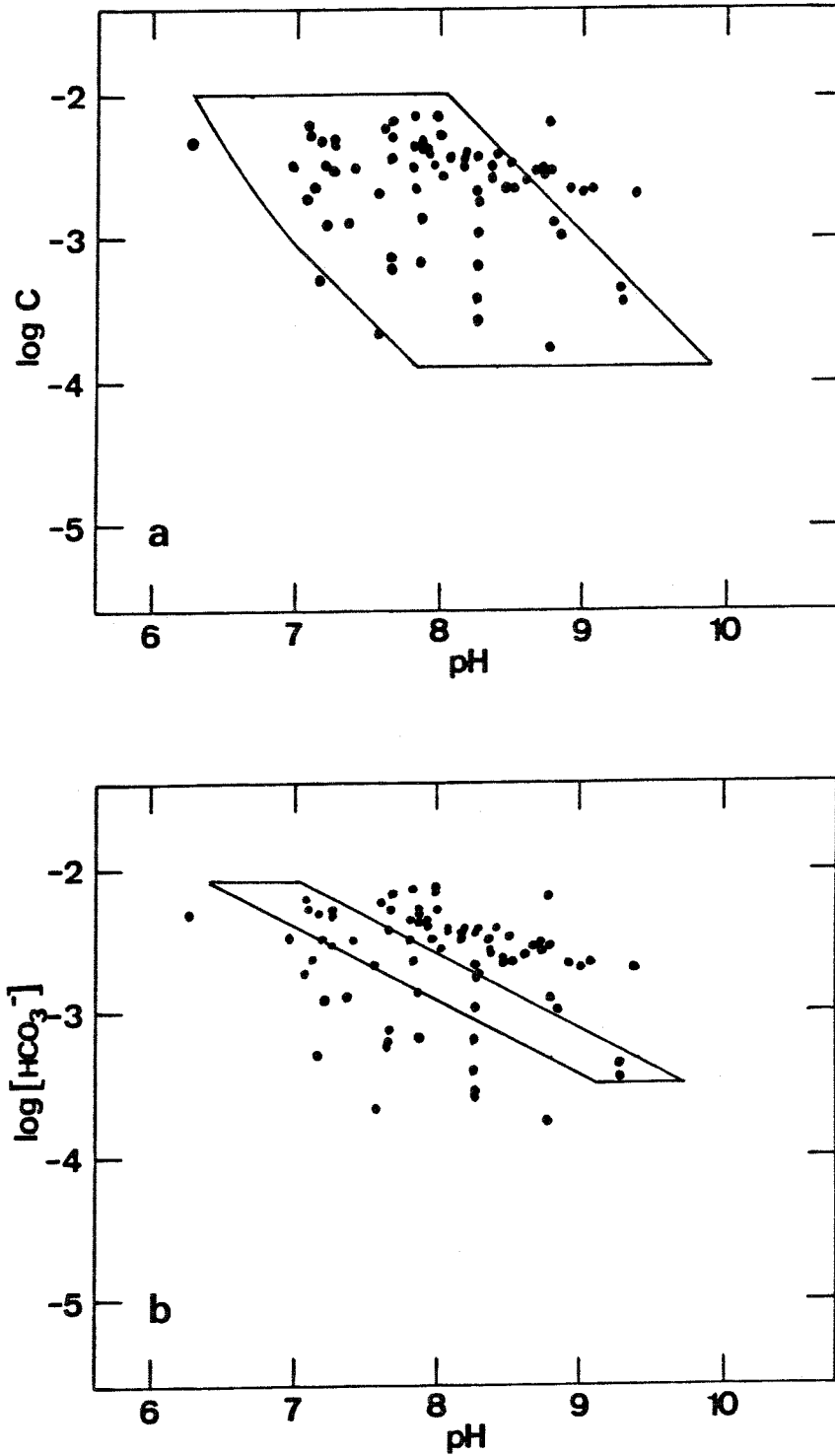


Figure 4 Measured pH and HCO_3^- -concentrations.

(a) Closed system with fixed total carbonate concentration (=Figure 1), (b) System with a CO_2 reservoir (=Figure 3).

The enclosed areas are the same as in Figure 1 and 3 (representing saturated systems)

- o The observed field data on pH and carbonate concentrations correspond reasonably well with the "closed system - fixed total carbonate concentration"-case. However, no effort is made to fully consider the variation of the stability constants with the ionic strength in the construction of Fig. 1 - 4.
- o For most of the deep groundwaters studied the observed pH is in the range 7-9 and the observed total carbonate concentration in the range 30-400 mg/l, 0.5-6.5 mM; (more often in the range 90-275 mg/l, 1.5-4.5 mM).

Calculated CO_3^{2-} -concentrations are given in Fig. 5. For waters with high total carbonate concentrations (more than 90 mg/l), the CO_3^{2-} -concentration can be estimated from

$$\log [\text{CO}_3^{2-}] = 0.76\text{pH} - 10.83 \pm 0.08$$

A lower limit is set by

$$\log [\text{CO}_3^{2-}] = \text{pH} - 14.$$

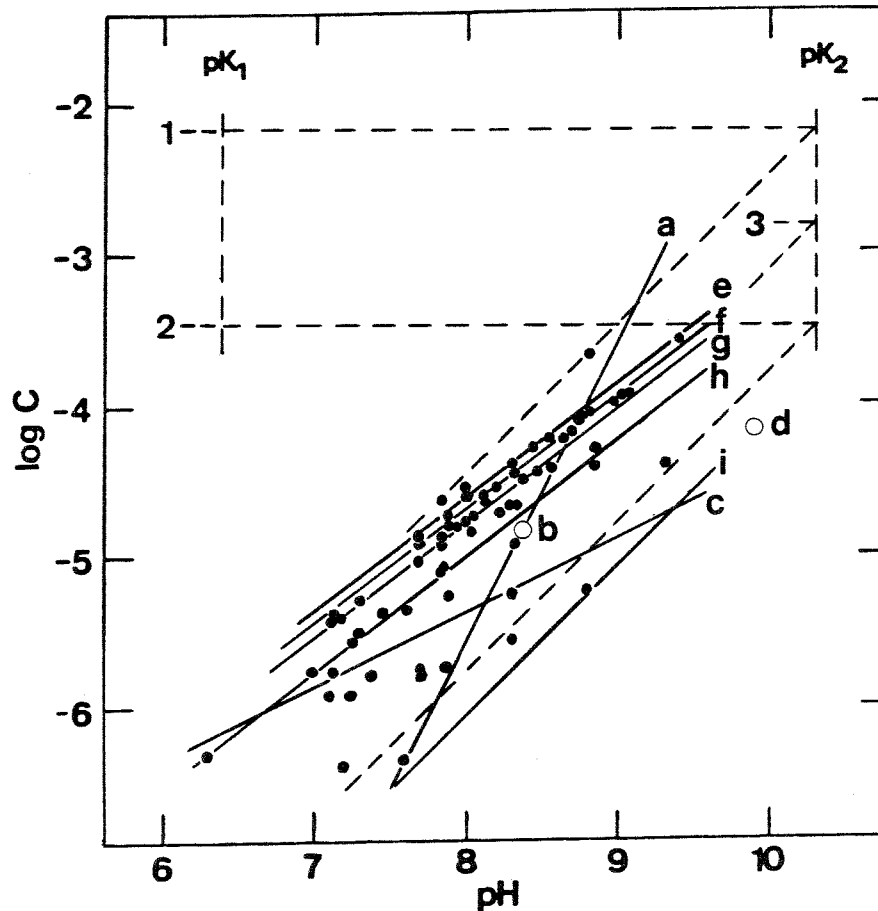


Figure 5 Calculated CO_3^{2-} -concentrations.

1. 400 mg/l HCO_3^- ; 2. 20 mg/l HCO_3^- ; 3. 90 mg/l HCO_3^- .
- (a) $\log [\text{CO}_3^{2-}] = 2\text{pH} - 18.1 + \log P(\text{CO}_2)$, $\log P(\text{CO}_2) = -3.5$
Open system without $\text{CaCO}_3(\text{s})$.
- (b) $\log [\text{CO}_3^{2-}] = -4.9$, $\text{pH} = 8.4$
Dissolution of $\text{CaCO}_3(\text{s})$ in an open system.
- (c) $\log [\text{CO}_3^{2-}] = 0.5\text{pH} - 9.35$
Dissolution of $\text{CaCO}_3(\text{s})$ in a closed system with equal total carbonate and calcium concentration.
- (d) $\log [\text{CO}_3^{2-}] = -4.4$, $\text{pH} = 9.9$
Dissolution of $\text{CaCO}_3(\text{s})$ in water.
- (e) $\log [\text{CO}_3^{2-}] = 0.76\text{pH} - 10.76$
Equal to or higher than 95% of measured data.
- (f) $\log [\text{CO}_3^{2-}] = 0.76\text{pH} - 10.83$
Equal to or higher than 75% of measured data.
- (g) $\log [\text{CO}_3^{2-}] = 0.76\text{pH} - 10.91$
Equal to or higher than 50% of measured data.
- (h) $\log [\text{CO}_3^{2-}] = 0.76\text{pH} - 11.16$
Equal to or higher than 25% of measured data.
- (i) $\log [\text{CO}_3^{2-}] = \text{pH} - 14$
Approximate minimum level.

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OBSERVED pH, Ca^{2+} - AND HCO_3^- -CONCENTRATIONS

Observed pH, $[\text{Ca}^{2+}]$ and $[\text{HCO}_3^-]$ in natural granitic groundwaters are compiled below (3-5).

FJ	= Fjällveden
SV	= Svartboberget
GI	= Gideå
KR, KS	= Kråkemåla
ST, SS	= Sternö
FI, FS	= Finnsjön
FO	= Forsmark
G	= Gåvastebo

a, b and c indicate the variation in composition observed after long pumping times.

Location	Hole no.	Depth	pH	$\log[\text{HCO}_3]$	$\log[\text{Ca}^{2+}]$
FJ	2	106	8.05	-2.58	-3.29
FJ	2	293	7.14	-2.64	-3.33
FJ	2	409	7.45	-2.55	-3.28
FJ	2	506	8.85	-2.85	-3.53
FJ	4	131a	7.85	-2.49	-3.18
FJ	4	131b	8.2	-2.46	-3.44
FJ	4	272	8.0	-2.50	-3.17
FJ	4	349a	8.2	-2.49	-3.57
FJ	4	349b	8.4	-2.49	-3.38
FJ	4	420a	7.1	-2.74	-3.28
FJ	4	420b	8.85	-2.48	-3.48
FJ	8	402	8.29	-2.68	-3.21
FJ	8	562	8.85	-2.67	-3.18
SV	4	82	8.46	-2.64	-3.21
SV	4	394	9.4	-2.68	-3.53
SV	4	373	9.08	-2.67	-3.36
SV	4	551	9.04	-2.68	-3.39
GI	2	157	8.75	-2.58	-3.61
GI	2	288	8.80	-2.57	-3.61
GI	2	353	8.63	-2.58	-3.63
GI	2	478	8.77	-2.58	-3.63
GI	2	528	8.70	-2.58	-3.62
GI	4	91	7.84	-2.65	-3.10
GI	4	212	8.96	-2.66	-3.62
GI	4	385	9.33	-3.45	-3.30
GI	4	498	8.31	-2.71	-3.14
GI	4	596a	7.25	-2.90	-3.10
GI	4	596b	8.32	-2.99	-2.87
GI	4	596c	8.85	-2.97	-2.80
SS	-	0	7.7	-3.21	-3.23
ST	3	232	6.3	-2.36	-2.51
ST	4	226	7.3	-2.32	-2.74
ST	4	312	7.2	-2.32	-2.68
ST	4	397	7.3	-2.32	-2.71
G	1	60	7.0	-2.49	-2.43
G	2	66	8.0	-2.28	-2.62

Location	Hole no.	Depth	pH	$\log[\text{HCO}_3^-]$	$\log[\text{Ca}^{2+}]$
FS	-	0	-	-3.21	-3.41
FI	1	206	7.1	-2.28	-2.83
FI	1	293	7.1	-2.24	-2.91
FI	2	385	7.65	-2.28	-3.14
FI	4	152	8.8	-2.20	-3.25
FI	4	247	8.0	-2.19	-3.23
FI	4	368	7.7	-2.19	-3.27
FI	4	534	7.7	-2.19	-3.27
FI	5	141	8.4	-2.58	-1.81
FI	5	205	7.9	-2.87	-1.66
FI	5	297	7.9	-3.19	-1.44
FI	5	384	7.7	-3.17	-1.36
FI	6	184	7.6	-2.69	-1.85
FI	6	250	7.4	-2.91	-1.54
FI	6	398	8.3	-3.58	-1.33
FI	6	688	8.3	-3.21	-1.33
FI	7	123	-	-2.26	-3.11
FI	7	301	8.3	-2.42	-2.56
FI	7	322	-	-2.32	-2.58
FI	7	511	7.9	-2.34	-2.46
FI	8	103	7.9	-2.38	-3.07
FI	8	196	7.2	-3.31	-1.42
FI	8	283	7.6	-3.67	-1.39
FI	8	395	8.8	-3.74	-1.40
KS	1	0	-	-2.48	-2.80
KR	1	103	7.7	-2.44	-3.01
KR	1	264	8.1	-2.43	-3.17
KR	1	406a	8.3	-2.45	-3.15
KR	1	406b	8.45	-2.46	-3.23
KR	1	491a	7.95	-2.46	-3.33
KR	1	491b	8.1	-2.44	-3.29
KR	1	491c	7.85	-2.44	-3.27
KR	2	291	7.3	-2.53	-2.99
KR	2	510	7.25	-2.53	-2.97
FO	1	458	7.85	-2.20	-3.06

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