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Sorption of actinides in granitic rock

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SORPTION OF ACTINIDES IN GRANITIC ROCK

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SUMMARY

The sorption of americium(III), neptunium(V) and plutonium on geologic media under oxic conditions has been measured by a batch technique. The aqueous phase was a synthetic groundwater or 4M NaCl solution. The solid phase was a pure mineral, representative of igneous rocks, or granite. Altogether 40 different minerals and rocks were used. The effects of pH and the ionic strength of the aqueous phase as well as of the cation exchange capacity and the surface/mass ratio of the solid sorbent are discussed. Empirical equations giving the distribution coefficient as a function of pH in the environmental pH-range 7-9 are suggested. Some observations and conclusions concerning sorption mechanisms are given.

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1. INTRODUCTION

The actinides with their daughter products (Th, Pa, U, Np, Pu, Am, Cm) will largely dominate the biological hazards from spent nuclear fuel already some hundred years after the discharge from the reactor and onwards. Thus, it is of prime importance that the chemical behaviour of the actinides in environmental systems is well understood. This would be the necessary basis for predictions of actinide reactions and mobilities in nature, e.g. in connection with accidental releases from the nuclear fuel cycle or from an underground repository for spent fuel or high-level waste.

In the present investigation the sorption of Am, Np and Pu on geologic material has been studied. This report is a compilation of data from three different groups and time periods.

1. Dep. of Nuclear Chemistry, Chalmers University of Technology, Sweden; 1977-78.
   (B. Allard, H. Kipatsi, J. Rydberg, B. Torstenfelt).
   Most results from this period have previously been reported (1-4).
2. Transuranium Research Lab., Oak Ridge Nat. Lab., USA; 1978-79.
   (B. Allard, G.W. Beall, T. Krajewski).
   All data from this period has previously been presented (5-21).
3. Dep. of Nuclear Chemistry, Chalmers University of Technology, Sweden; 1980-82.
   (B. Allard, K. Andersson, U. Olofsson, B. Torstenfelt).
   Some data from this period has previously been reported (22-29) although most results will be presented elsewhere (30).

Some qualitative and semi-quantitative observations on actinide behaviour in geochemical systems are briefly summarized in this report. Detailed discussions of sorption mechanisms etc., as well as comparisons with literature data, will appear elsewhere (30).

2. THE GROUNDWATER - ROCK SYSTEM

Spent nuclear fuel or high-level reprocessing waste will be stored in granitic bedrock at great depth in the present Swedish concept for radioactive waste deposition. The studies of actinide behaviour under the conditions expected in a granitic repository have been concentrated on the following approach:

- Description of the geochemistry of a deep repository in granite (mineralogy of the bedrock and fracture filling materials, groundwater chemistry).
- Description of actinide chemistry in groundwater systems.
- Description of actinide sorption behaviour in geologic systems.

2.1. Major minerals of igneous rocks

Igneous rocks like granite are composed of a small number of major constituents like quartz, feldspars (orthoclase, plagioclase), micas (biotite), amphiboles, pyroxenes and olivine, as illustrated in Fig. 1. Moreover, there are some accessory minerals, e.g. fluorite, calcite, apatite, magnetite, etc. that would have some influence on the properties of water exposed surfaces and on the composition of the groundwater (see below).

2.2. Composition of granitic groundwaters

The complex relations between solid geologic media (major and accessory components, weathering products), species in solution and the biosphere and atmosphere are schematically illustrated in Fig. 2.

The most important chemical system in the aqueous phase appears to be the H_2O-CO_2-system, which largely determines pH (through the protolysis of H_2CO_3). In most groundwaters, the total carbonate concentration would determine the maximum calcium concentration (saturated $CaCO_3$(s)-solution). Some interrelations exist between the calcium, sodium and magnesium concentrations through ion

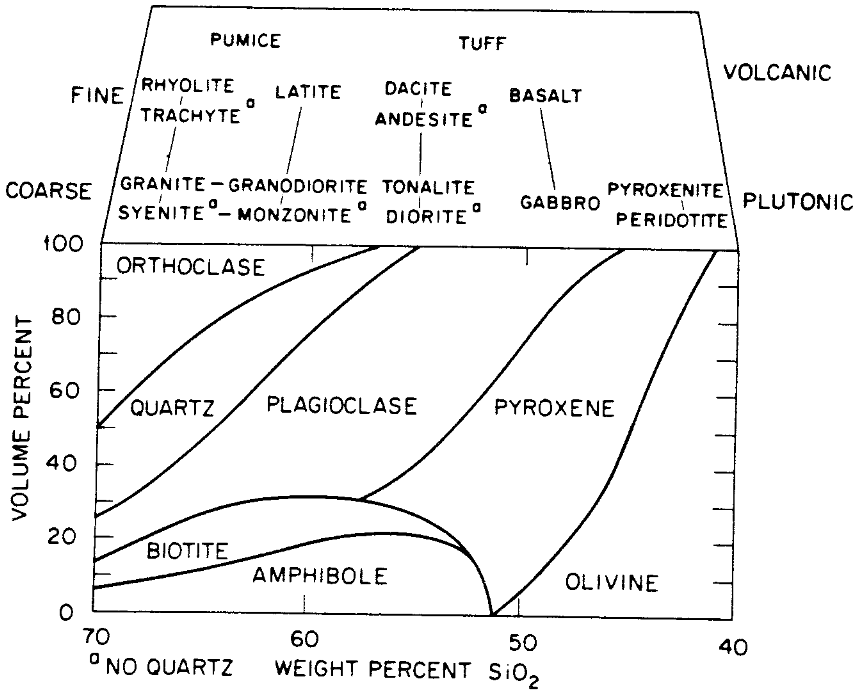


Figure 1 Major components of igneous rocks (7)

exchange reactions with weathering products (mostly clay minerals). Moreover, the calcium concentration would limit the total fluoride and phosphate concentrations due to the low solubility of fluorite and apatite. Thus, the concentrations of $HCO_3^- - Ca^{2+} - Na^+ - Mg^{2+} - F^- - HPO_4^{2-}$ as well as pH would to some extent be interdependent and related to the presence of weathering products and some accessory minerals.

Another interdependent chemical system would be $Fe^{2+} - O_2$ with Eh and pH, which would be related to the presence of iron containing minerals. (Further aspects on groundwater equilibria are given e.g. in ref. 31).

Observed concentration ranges of major species in deep granitic groundwaters are given in Table 1. (32, 33) Moreover, saline waters with total salt concentrations ($Na^+ - Ca^{2+} - Cl^-$) of up to 10 g/l have been encountered.

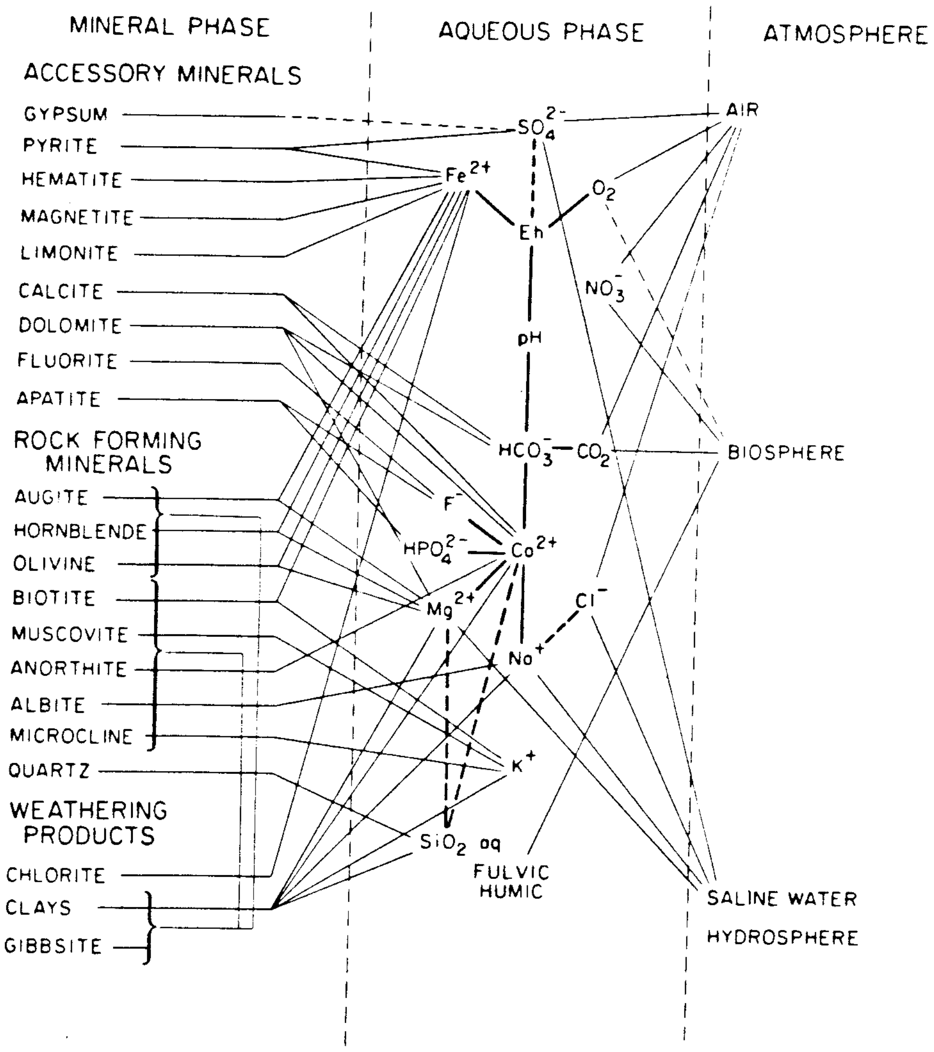


Figure 2 The groundwater - rock system (6)

2.3. Fracture filling minerals

The mineralogical composition of fracture filling products would be of importance for the retention of radionuclides in the groundwater, since the fracture minerals and not the bulk rock itself would primarily be exposed to the groundwater. Three categories of fracture minerals will be expected (34).

- o Weathering and alteration products of e.g. micas, feldspars, amphiboles, etc. (clay minerals).
- o Precipitates and crystallization products from aqueous solutions, not necessarily formed under hydrothermal conditions (e.g. calcite).
- o Metamorphic products.

Table 1 Groundwater composition (12, 32)

Species	Concentration in nature [a] mg/l	Artificial groundwater mg/l
HCO_3^-	30-400	123
SO_4^{2-}	1-25	9.6
Cl^-	5-50	70
F^-	0.01-5	
HPO_4^{2-}	0.01-0.5	
SiO_2(tot)	5-30	12
Ca^{2+}	10-50	18
Mg^{2+}	2-20	4.3
Na^+	10-100	65
K^+	1-5	3.9
Fe^{2+}	0.5-20	
pH	7.2-8.5	8.2

[a] Deep undisturbed non-saline groundwater in granitic rock.

A list of common fracture minerals in granitic rock is given in Table 2 (34).

2.4. Selection of minerals and artificial groundwaters for sorption studies

In the selection of minerals for the experimental studies the general principle has been to include most of the common minerals in igneous rocks that for various reasons might be expected to have some significant influence of the actinide sorption behaviour. The following groups of minerals are represented:

- Major rock forming minerals
- Common accessory minerals
- Accessory minerals known to contain minor amounts of actinides or lanthanides
- Weathering products of the rock forming minerals

Table 2 Fracture minerals in granite (34)

Mineral	Occurrence [a]
Calcite	A
Dolomite	S, Me
Gypsum	P
Pyrite	A
Epidote	Me, H, S
Prehnite	Me, H
Chlorite	A
Kaolin	W, H
Montmorillonite	S, H
Illite	W, H
Quartz	A
Laumontite	S, H
Stilbite	S, H
Analcime	S, H

[a] A = in all geologic environments
S = in sedimentary rocks
Me = in metamorphic rocks
W = as weathering product
H = as hydrothermal product
P = precipitate

- o Common fracture minerals
- o Minerals containing anions that would form strong actinide complexes (F^-, PO_4^{3-}, CO_3^{2-})
- o Minerals representing all of the six major structure classes of the silicates

A list of the minerals is given in Table 3.

An artificial groundwater, Table 1, representative of deep non-saline granitic groundwater, was used as aqueous phase, as well as 4 M NaCl, representing very saline waters or brine.

Table 3 Minerals and rocks selected for sorption studies

Class	Mineral[a]	Class	Mineral[a]
Sulfides	Pyrite	-Cyclo silicates	Beryl
	Chalcopyrite		
	Galena	-Ino silicates	Augite
	Molybdenite		Hornblende
	Chalcosite		Attapulgite
Oxides, Hydroxides	Magnetite	-Phyllo silicates	Kaolin
	Hematite		Halloysite
	Limonite		Serpentine
	Corundum		Montmorillonite
	Gibbsite		Muscovite
			Biotite
Halides, Sulfates, Carbonates, Phosphates	Fluorite		Chlorite
	Anhydrite		
	Calcite	-Tecto silicates	Albite
	Dolomite		Microcline
	Apatite		Bytownite
	Monazite		Anorthite
			Quartz
Silicates	Olivine		
-Neso silicates	Almandine	Igneous rocks	Basalt (36)
	Zircon		Climax Stock granite (36)
	Sphene		Westerly granite (36)
-Soro silicates	Epidote		Finnsjön granite (34)

[a] See e.g. ref. 35.

3. EXPERIMENTAL

3.1. Preparation and characterization of the mineral samples

Pure, crystalline products from natural locations were selected. Mineral grains that appeared to be reasonably pure were selected from a coursely ground material and then further ground and sieved. Usually the size fraction 0.044-0.063 mm was used in the sorption studies. The purity was checked by X-ray diffractometry, and for some of the minerals also by chemical analysis. Normally, the impurities were low (within 5%). Mineralogic compositions of the rock samples are given in ref. 34 and 36. (Detailed data concerning mineral composition, sources, etc. are available, but not included or further discussed in this report).
For most of the minerals used the surface/mass ratio was determined by the ethylene glycol method (37) as well as the BET-method in a few cases. (C.f. ref. 43).

The anion and cation exchange capacities were determined as a function of pH by a batch technique (38). (C.f. ref. 43).

3.2. Sorption studies

3.2.1. Radionuclides

Radionuclides used in the distribution measurements are given in Table 4. The short-lived nuclides ^{235}Np and ^{237}Pu were obtained from Argonne Nat. Lab. and Harwell (in the experiments at Oak Ridge and Chalmers, respectively). The long-lived nuclides ^{237}Np and ^{239}Pu were used in some of the early measurements, primarily on montmorillonite and Finnsjön granite.

3.2.2. Batch-wise distribution measurements

The crushed and sieved solid material was prewashed with aqueous phase for five days. The solid phase was allowed to settle by gravity or by light centrifugation (for clay samples) and the washing solution was discharged. After a second identical wash a known amount of aqueous phase was added and the system was allowed to preequilibrate for another five days before an acidic active spike solution was added and pH adjusted (with 0.1 or 0.01 M NaOH or HCl) to a preselected value.

The activity remaining in solution (after phase separation by centrifugation) was measured, and the distribution coefficient Kd (kg/kg)[a], calculated according to

$$Kd = (c_0 - c)V\rho/cm$$

where c_0 = initial concentration (M)
c = measured concentration after a certain contact time (M)
V = volume of aqueous phase (m^3)
ρ = density of aqueous phase (kg/m^3)
m = mass of solid phase (kg)

The sorption on the walls of the vials was measured and found to be negligible in the presence of crushed minerals or rocks, due to the very large surface provided by the crushed material in comparison with the exposed surface of the vial.

The composition of the artificial groundwater was not significantly altered under the present experimental conditions except in a few systems (e.g. quartz, hornblende, anhydrite) (C.f. Fig. 3).

Experimental conditions etc. are given in Appendix. A detailed discussion of the proper choice of experimental parameters and conditions is given elsewhere (39).

Table 4 Radionuclides used in the sorption studies

Nuclide	Half-life
^{235}Np	396d
^{237}Np	2.14×10^6y
^{237}Pu	45.4d
^{239}Pu	2.41×10^4y
^{241}Am	433y

[a] The distribution coefficient is denoted by Kd (kg/kg) throughout this report. In most of the references the distribution coefficient K_d (m^3/kg) is used. Thus, Kd = 1000(K_d), assuming that the density of the aqueous phase is 1000 kg/m^3.

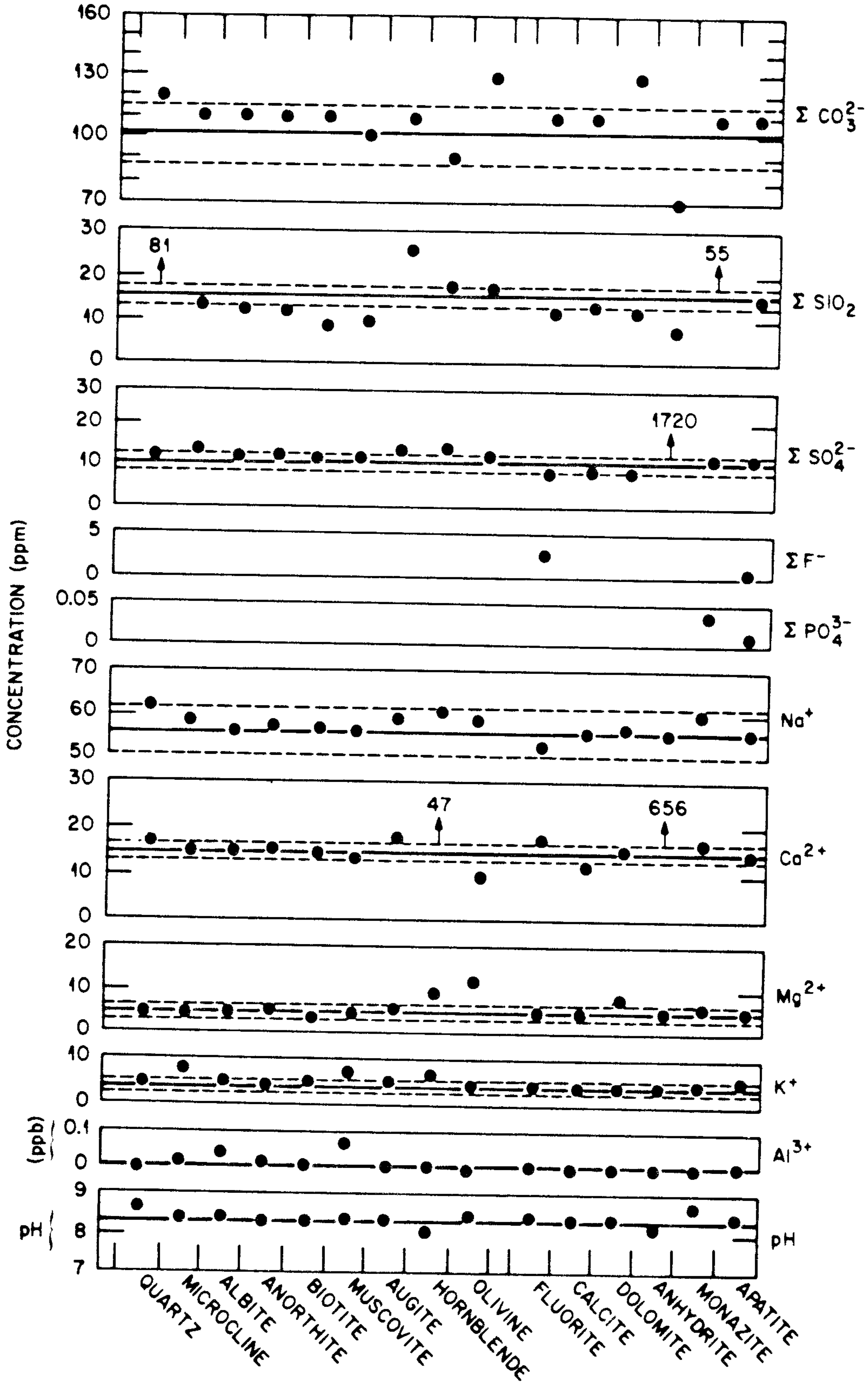


Figure 3 Composition of artificial groundwater after contact with crushed minerals for four weeks.

4. RESULTS AND DISCUSSION

Under the present conditions, americium and neptunium would be tri- and pentavalent, respectively, while plutonium would be predominantly tetravalent (24,27). The observed distribution coefficients (Kd) for the individual mineral systems (Appendix). are summarized in Fig. 4 - 14. The effects of pH, cation exchange capacity (CEC), surface area and the composition of the aqueous phase (groundwater, GW, or 4M NaCl) are briefly discussed separately below.

4.1. Influence of pH on the sorption

The most important single chemical parameter with influence on the sorption of actinides in well-defined oxidation states appears to be pH of the aqueous phase. Similar sorption isotherms were obtained for the individual actinides in most of the mineral systems with some exceptions (see 4.6.).

For americium (Fig. 4 - 8) Kd generally increases from low values at pH below 4 to maximum values at pH 6.5 to 8 and then tend to level out or slightly decrease at higher pH. This general behaviour, which seems to be relatively little affected by the nature of the various minerals, indicates a sorption mechanism that would be related to the solution chemistry (hydrōlysis) rather than to sorbent properties (see 4.5).

For neptunium (Fig. 9 - 13), similar sorption isotherms were also obtained for the various mineral systems, however significantly different from the americium systems. Very low Kd-values were obtained at pH below 7-7.5 but increasing with pH above this level. The number of minerals that deviate from this general behaviour is larger than for americium (see 4.6).

For plutonium (Fig. 14) very high Kd-values were obtained in the whole observed pH-range 4-9, however possibly with a slight decrease at pH above 7-8.

The effects of the solution chemistry, determined by pH, on the sorption behaviour is further discussed in 4.5.

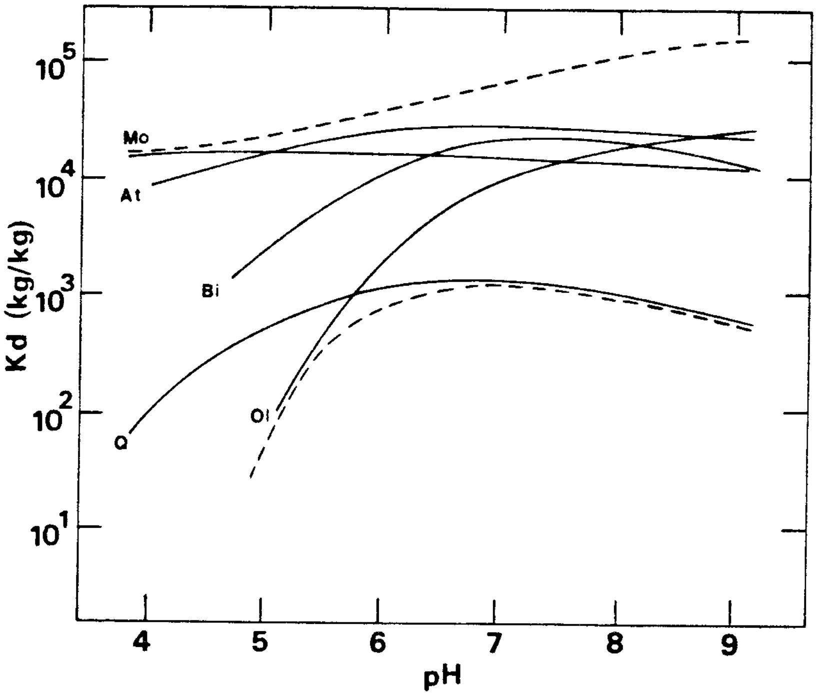


Figure 4 Kd for Am in groundwater - silicate systems. At=attapulgite, Bi=biotite, Mo=montmorillonite, Ol=olivine, Q= quartz.

Dotted lines indicate the ranges of observed values for totally 21 mineral systems

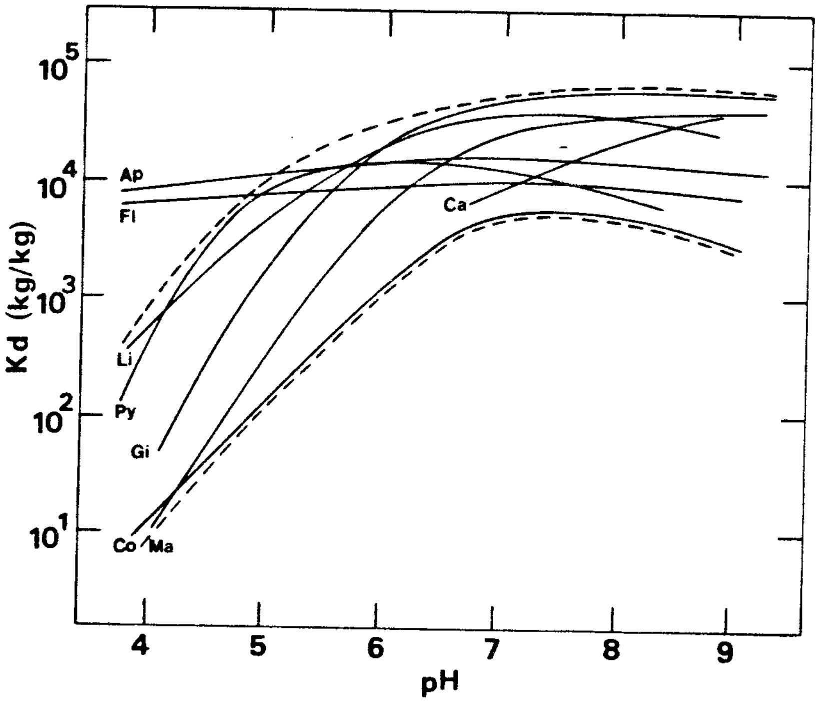


Figure 5 Kd for Am in groundwater - non-silicate systems. Ap=apatite, Ca=calcite, Co=curundum, Fl=fluorite, Gi=gibbsite, Li=limonite, Ma=magnetite, Py=pyrite.

Dotted lines indicate the ranges of observed values for totally 13 mineral systems

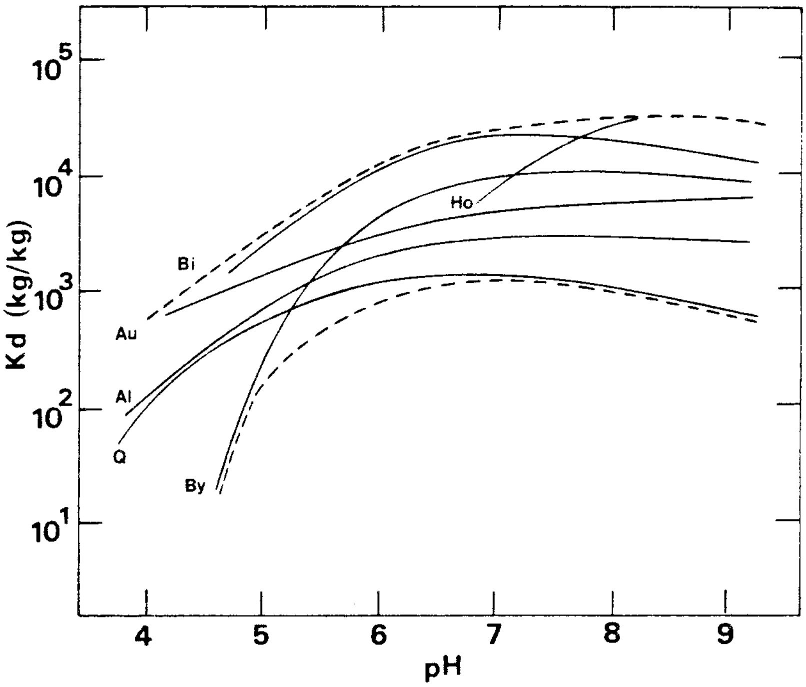


Figure 6 Kd for Am in groundwater - major granite mineral systems.
Al=albite, Au=augite, Bi=biotite, By=bytownite, Ho=hornblende, Q=quartz.
Dotted lines indicate the ranges of observed values for totally 10 mineral systems

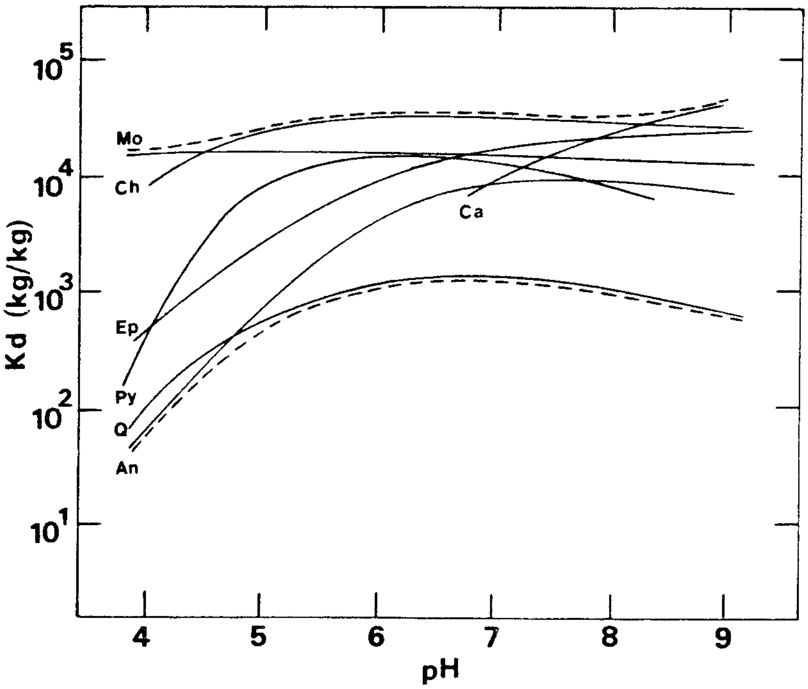


Figure 7 Kd for Am in groundwater - major fracture mineral systems.
An=anhydrite, Ca=calcite, Ch=chlorite, Ep=epidote, Mo=montmorillonite, Py=pyrite, Q=quartz.
Dotted lines indicate the ranges of observed values for totally 10 mineral systems

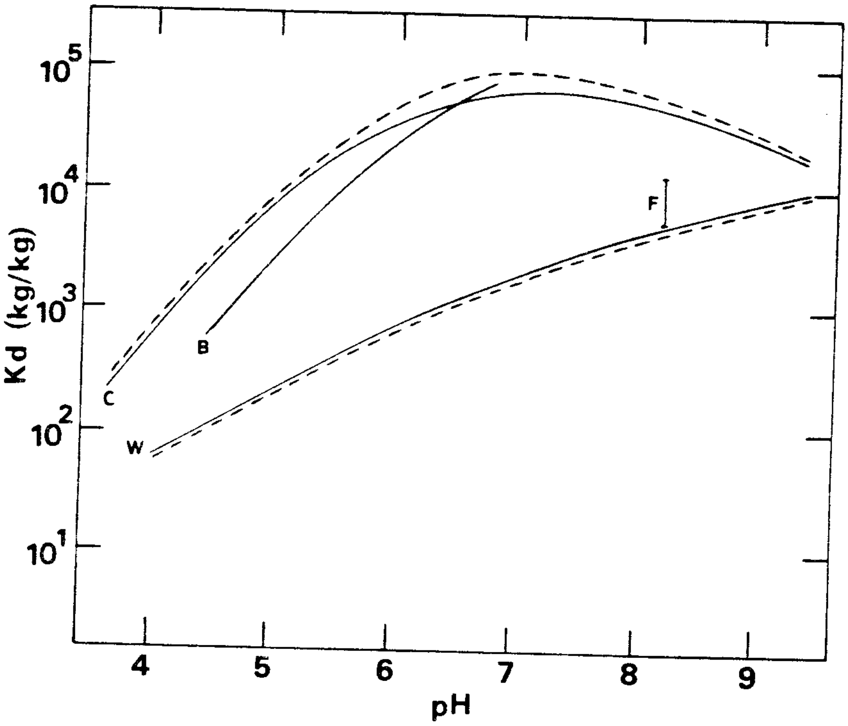


Figure 8 Kd for Am in groundwater - igneous rock systems.
B=basalt, C=granite (Climax Stock), F=granite (Finnsjön), W=granite (Westerly)

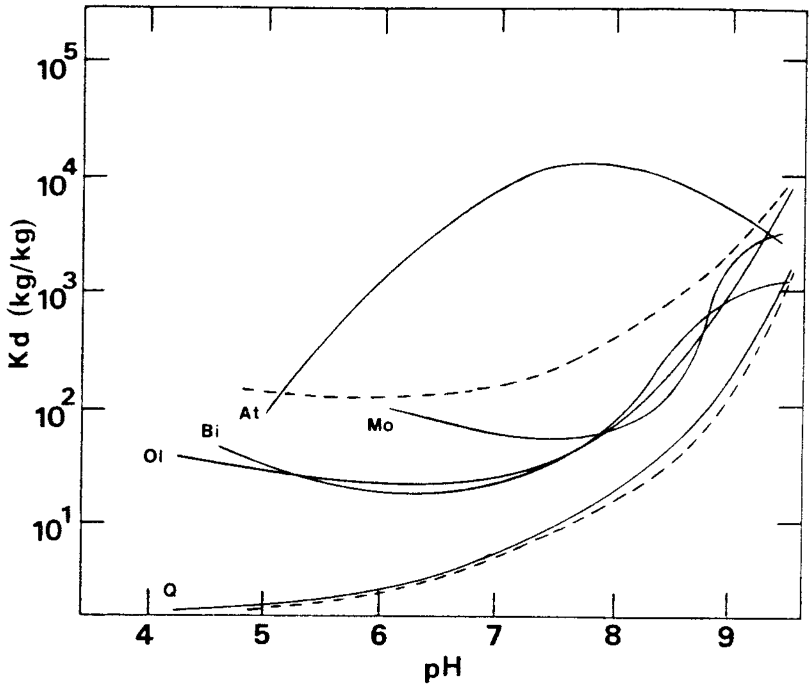


Figure 9 Kd for Np in groundwater - silicate systems. At=attapulgite, Bi=biotite, Mo=montmorillonite, Ol=olivine, Q= quartz.

Dotted lines indicate the ranges of observed values for totally 15 mineral systems

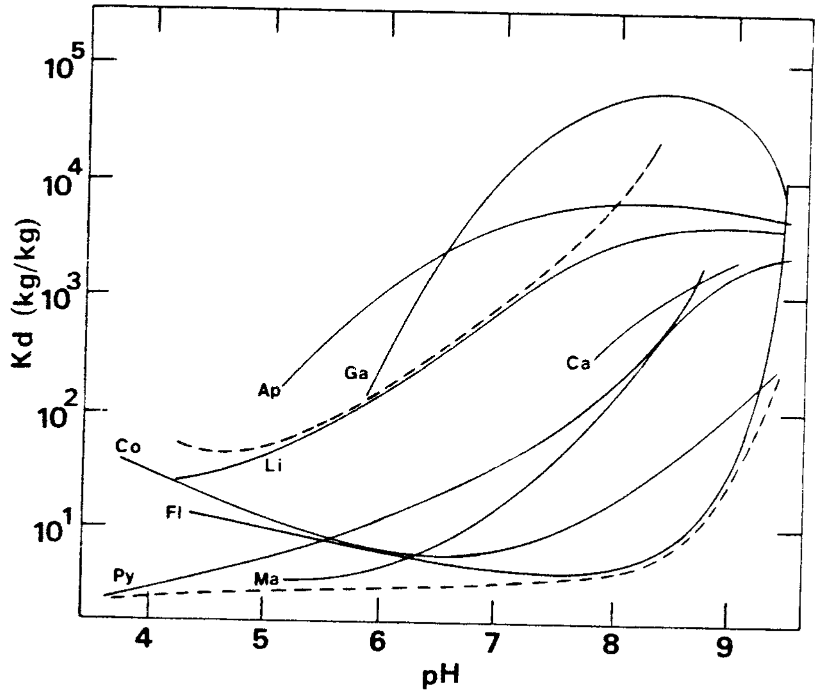


Figure 10 Kd for Np in groundwater - non-silicate systems. Ap=apatite, Ca=calcite, Co=corundum, Fl=fluorite, Ga=galena, Li=limonite, Ma=magnetite, Py=pyrite.

Dotted lines indicate ranges of observed values for totally 14 mineral systems

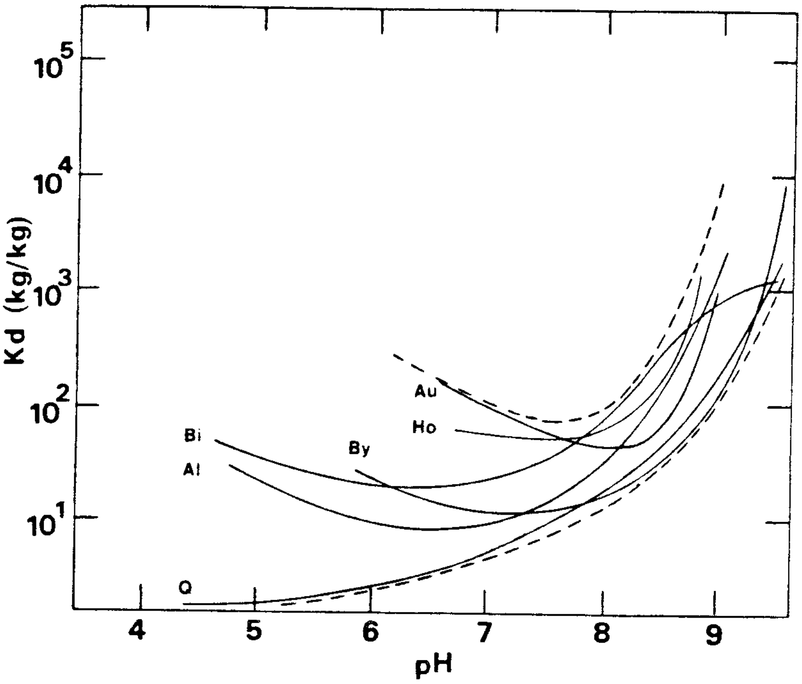


Figure 11 Kd for Np in groundwater - major granite mineral systems.
Al=albite, Au=augite, Bi=biotite, By=bytownite, Ho=hornblende, Q=quartz.
Dotted lines indicate ranges of observed values for totally 7 mineral systems

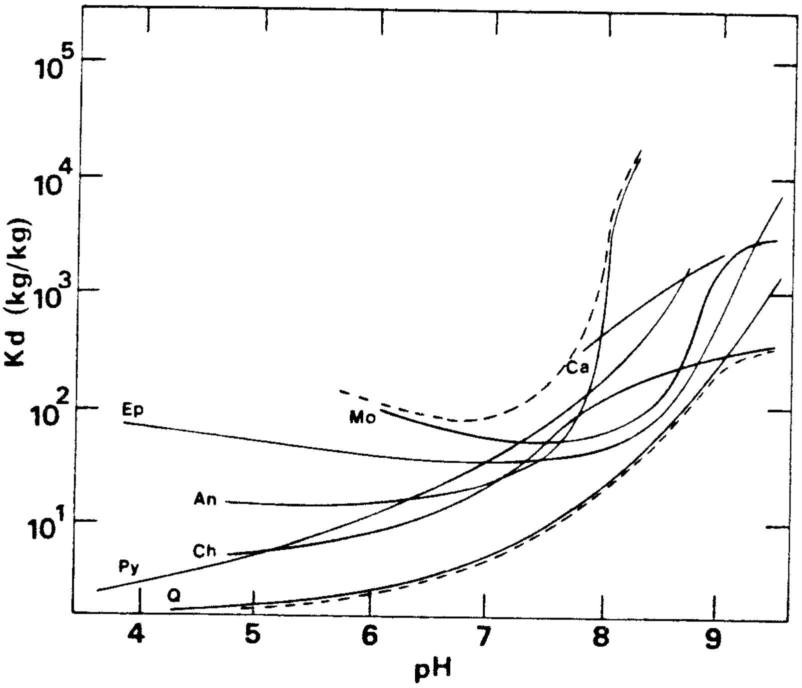


Figure 12 Kd for Np in groundwater - major fracture mineral systems.
An=anhydrite, Ca=calcite, Ch=chlorite, Ep=epidote, Mo=montmorillonite, Py=pyrite, Q=quartz.
Dotted lines indicate ranges of observed values for totally 10 mineral systems

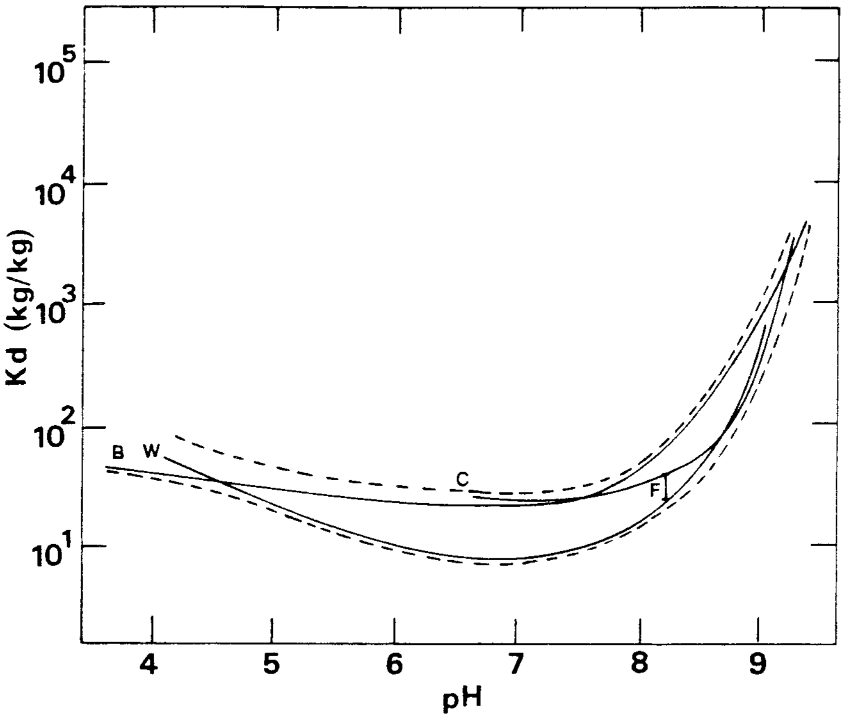


Figure 13 Kd for Np in groundwater - igneous rock systems. B=basalt, C=granite (Climax Stock), F=granite (Finnsjön), W=granite (Westerly)

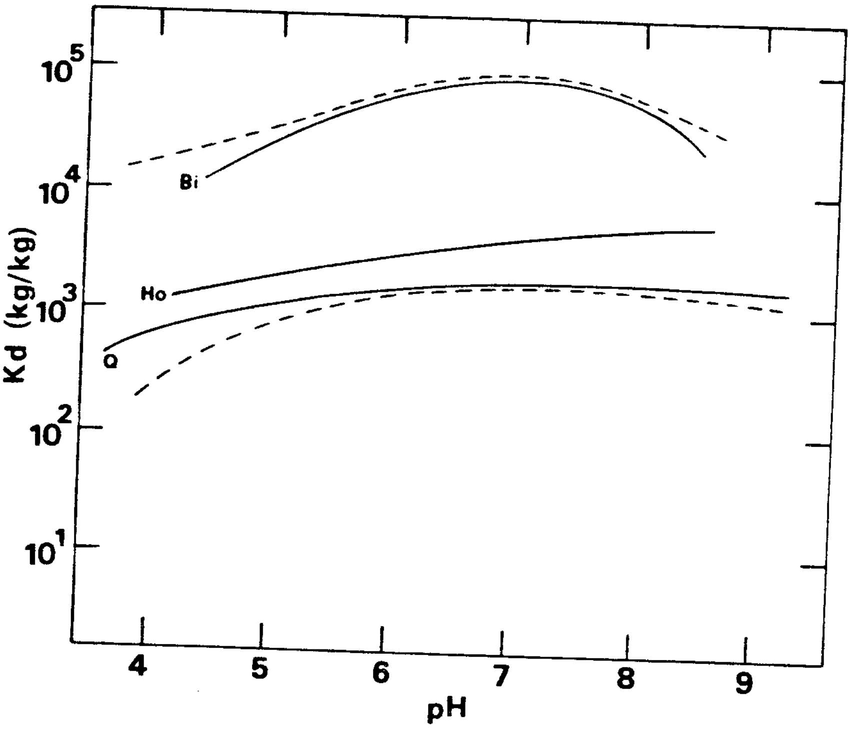


Figure 14 Kd for Pu in groundwater - major granite mineral systems.

Bi=biotite, Ho=hornblende, Q=quartz.

Dotted lines indicate ranges of observed values for totally 5 mineral systems

4.2. Influence of the cation exchange capacity on the sorption

The relation between Kd and CEC is illustrated in Fig. 15 and 16 for americium and neptunium, respectively (at pH 5 and 8 in GW).

Both for americium and neptunium Kd is generally increasing with CEC at pH 5, especially for the silicate systems. At pH 8 such a correlation is less obvious or possibly not significant.

4.3. Influence of the ionic strength on the sorption

The relation between Kd in GW and 4M NaCl is illustrated in Fig. 17 and 18 for americium and neptunium, respectively (at pH 5 and 8).

At pH 5 significantly higher Kd-values are obtained in GW than in 4M NaCl for most of the americium systems. This would be expected for a predominantly ion exchange process, as indicated in 4.2. However, the Kd-ratio between GW and 4M NaCl is generally below 10, which is less than expected since the ionic strength ratio is ca 500.

For neptunium the data are too scattered at pH 5 to allow any conclusions.

At pH 8 the differences in observed KD-values between GW and 4M NaCl have become much less than at pH 5 for the americium systems.

For neptunium significantly higher values were obtained in the 4M NaCl systems than in GW.

The difference between americium and neptunium in the dependence of the salt concentration is further illustrated in Fig. 19 and 20, showing Kd vs pH for two minerals with high CEC (biotite and montmorillonite). For americium the sorption exhibits a very pronounced ionic strength dependence at low pH. The difference decreases with increasing pH, and at pH above 7-8 there appears to be no significant difference between the sorption curves for the two aqueous phases.

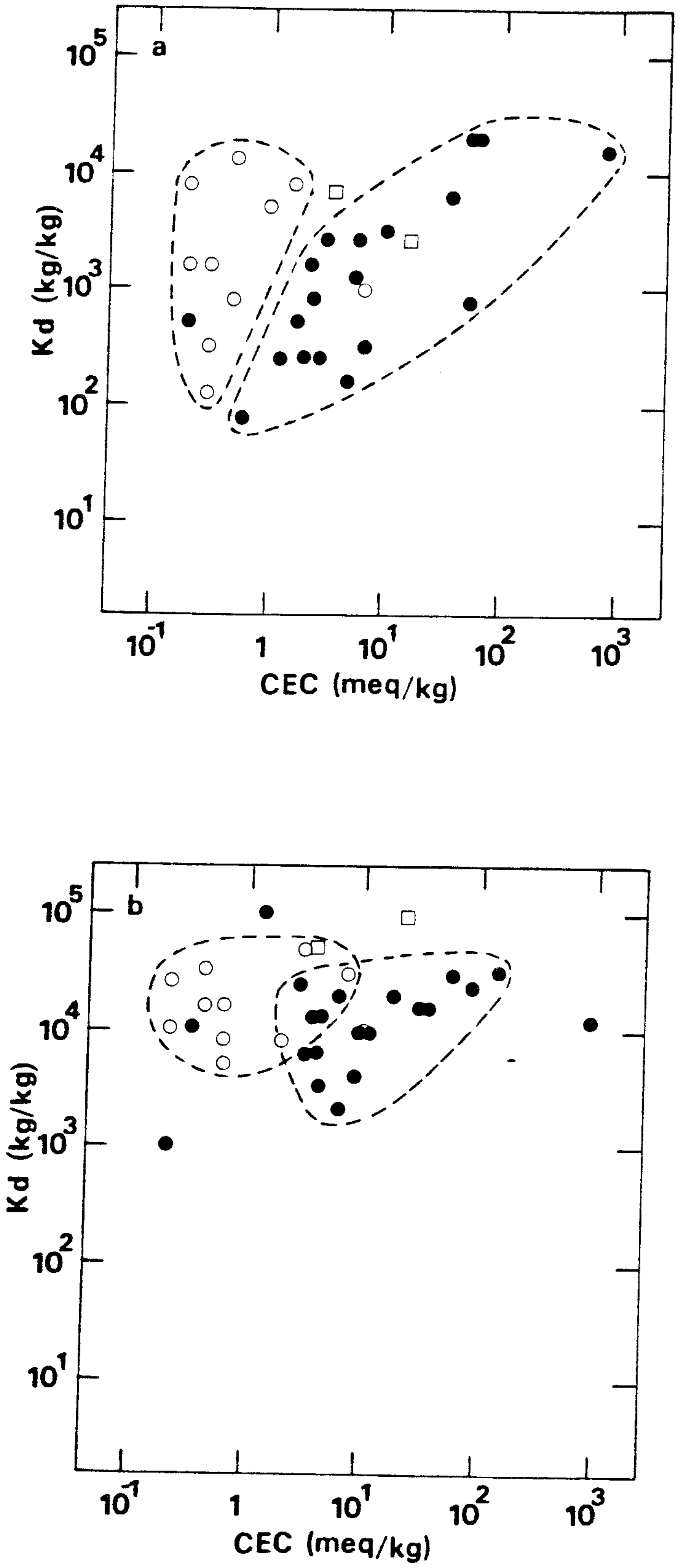
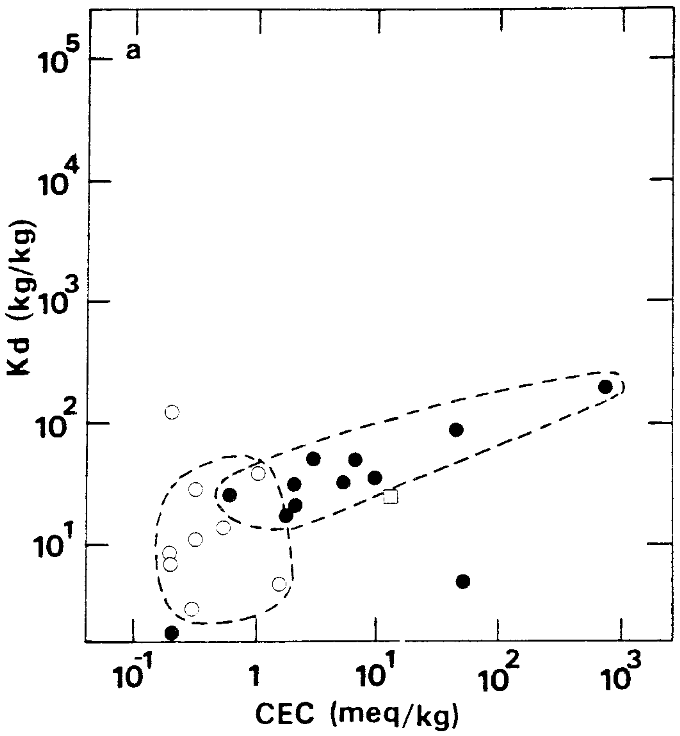


Figure 15 Kd vs CEC for Am at pH 5 (a) and pH 8 (b).
○ non-silicates, ● silicates, □ granites.



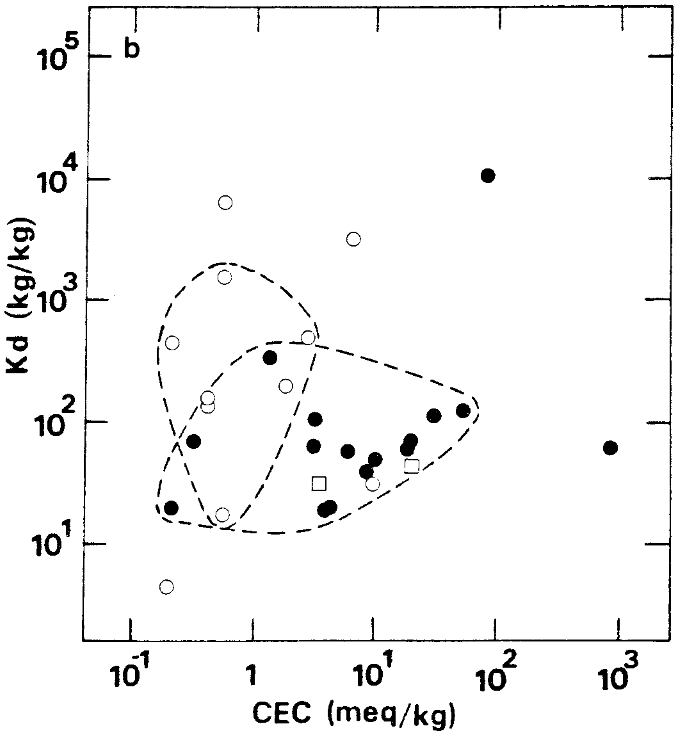
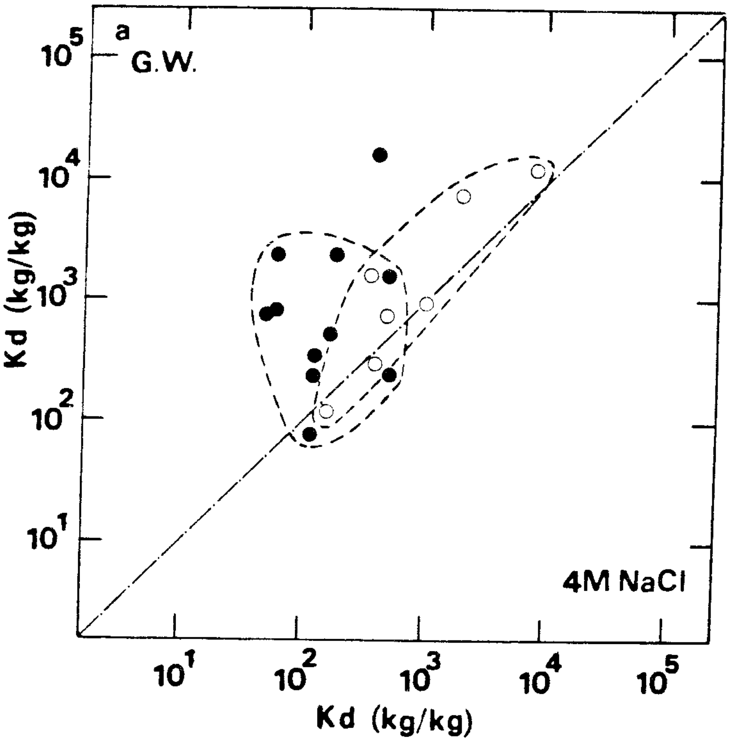


Figure 16 Kd vs CEC for Np at pH 5 (a) and pH 8 (b).
○ non-silicates, ● silicates, □ granites.



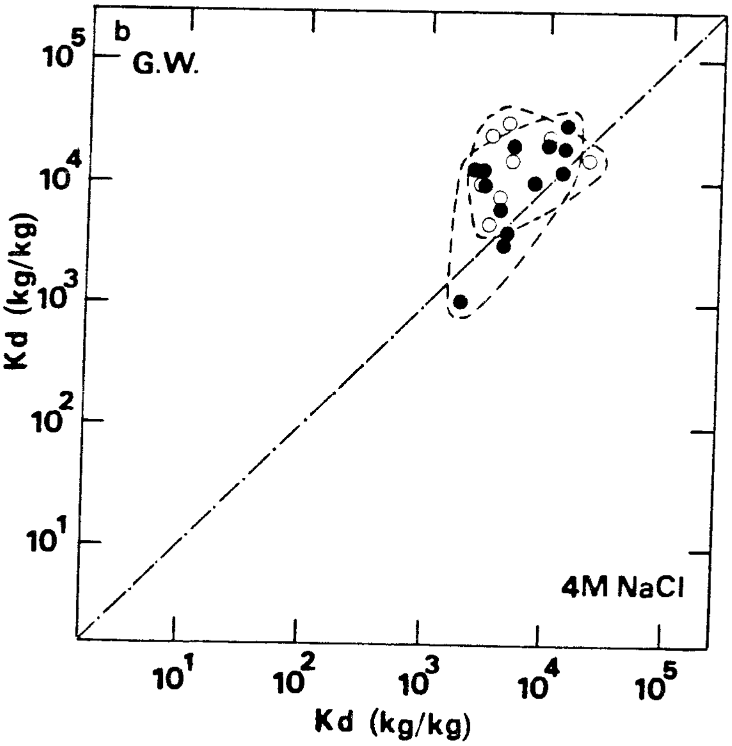
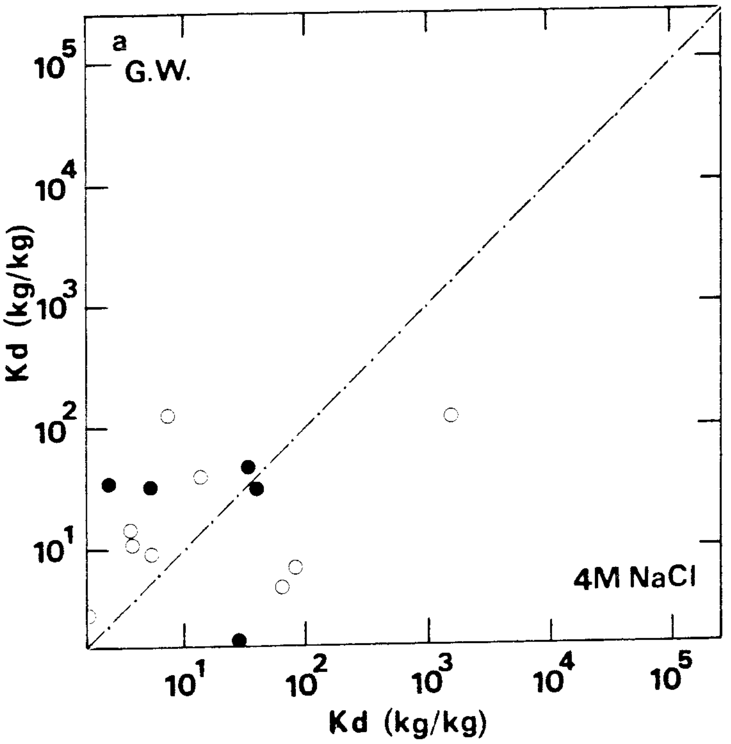


Figure 17 Kd for Am in groundwater and 4 M NaCl at pH 5 (a) and pH 8 (b).
○ non-silicates, ● silicates.



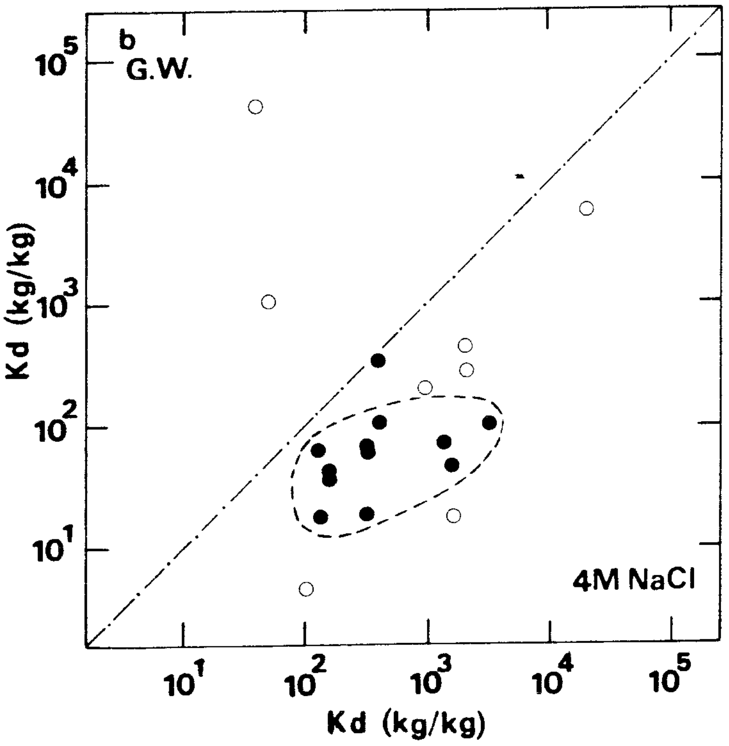


Figure 18 Kd for Np in groundwater and 4 M NaCl at pH 5 (a) and pH 8 (b).

○ non-silicates, ● silicates.

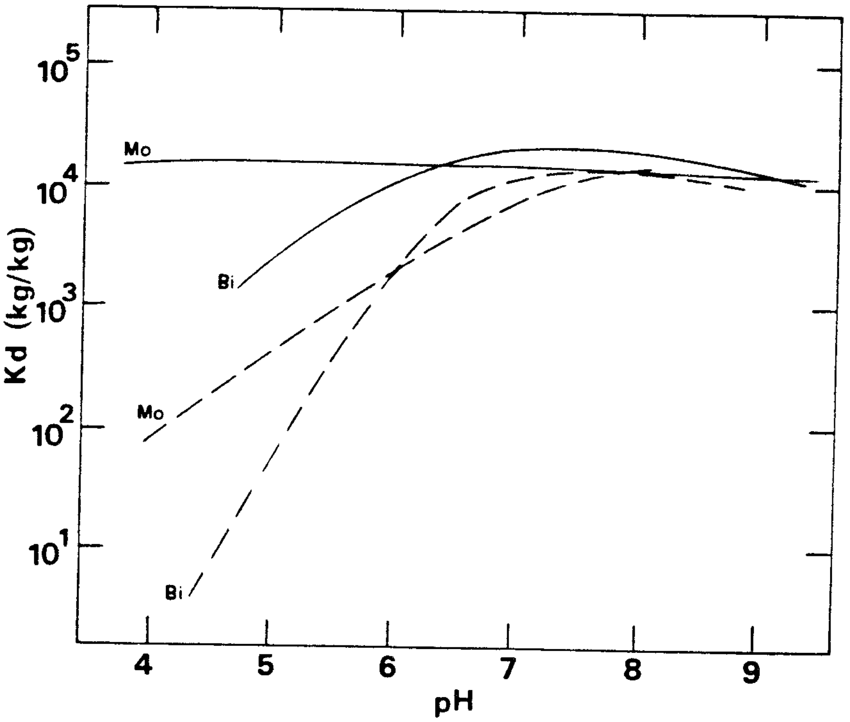


Figure 19 Kd vs pH for Am in groundwater (solid lines) and 4 M NaCl (dashed lines).
Bi=biotite, Mo=montmorillonite.

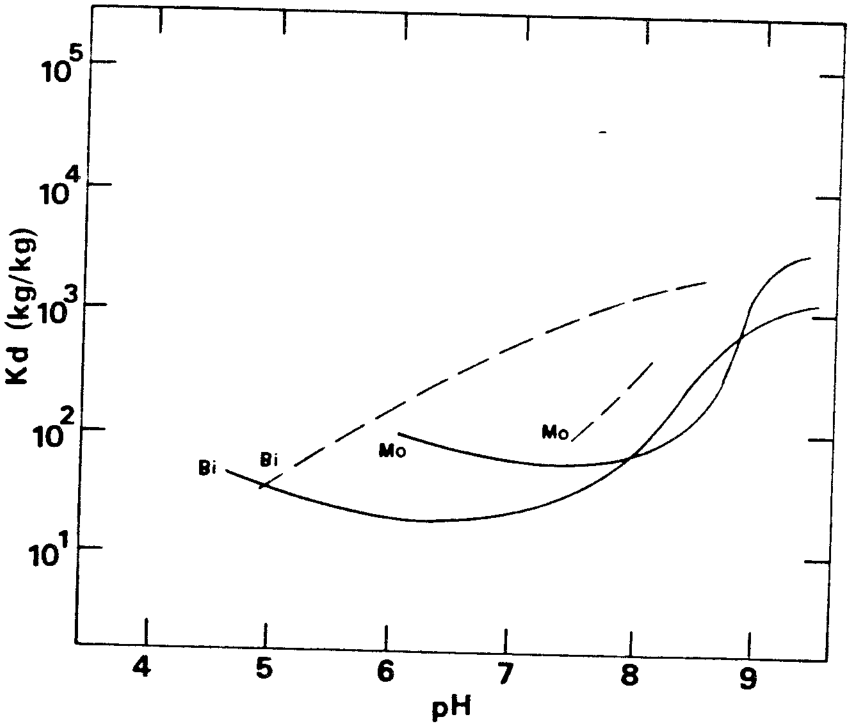


Figure 20 Kd vs pH for Np in groundwater (solid lines) and 4 M NaCl (dashed lines).
Bi=biotite, Mo=montmorillonite.

For neptunium the sorption curves in the 4M NaCl seem to be displaced 1-2 pH-units toward lower pH in comparison with the GW-systems (see further discussion in 4.5).

4.4. Influence of the surface area on the sorption

In Fig. 21 and 22 Kd vs the surface/mass ratio (A) is illustrated for americium and neptunium. Possibly a tendency of increasing Kd with increasing A can be noticed. However, also CEC is generally increasing with A (C.f. Fig. 2 - 9).

4.5. Influence of chemical speciation on the system

The pH-dependencies of the relative concentrations of americium and neptunium hydroxy and carbonate species (29) as well as of the observed distribution coefficients are illustrated in Fig. 23 and 24.

In both the americium and neptunium systems the sorption appears to increase when hydrolysis becomes significant (at pH above 5-6 for americium and at pH above 7-8 for neptunium). This observation is in qualitative agreement with sorption models based on hydrolysis and complex formation described in the literature (e.g. ref. 40). Also the maximum in sorption at pH when neutral hydrolysis products would dominate has previously been pointed out (e.g. in ref. 41).

The slightly decreasing Kd with increasing pH (above pH 8-9) for americium could reflect the formation of anionic species such as $Am(CO_3)_2^-$ and $Am(CO_3)_3^{3-}$. However, a similar decrease is not observed for neptunium in the pH-range where the predicted species $NpO_2(CO_3)_x^{1-2x}$ would dominate.

The displacement of the sorption curves towards lower pH observed for neptunium in the 4M NaCl system is not likely to reflect the chloride complexation. However, an increase of the hydrolysis constant, when going from groundwater (<0.01M) to a 4M salt solution, leading to a higher degree of hydrolysis at a certain pH, can not be excluded. Reliable hydrolysis constants determined at various ionic strengths are not available for neptunium(V).

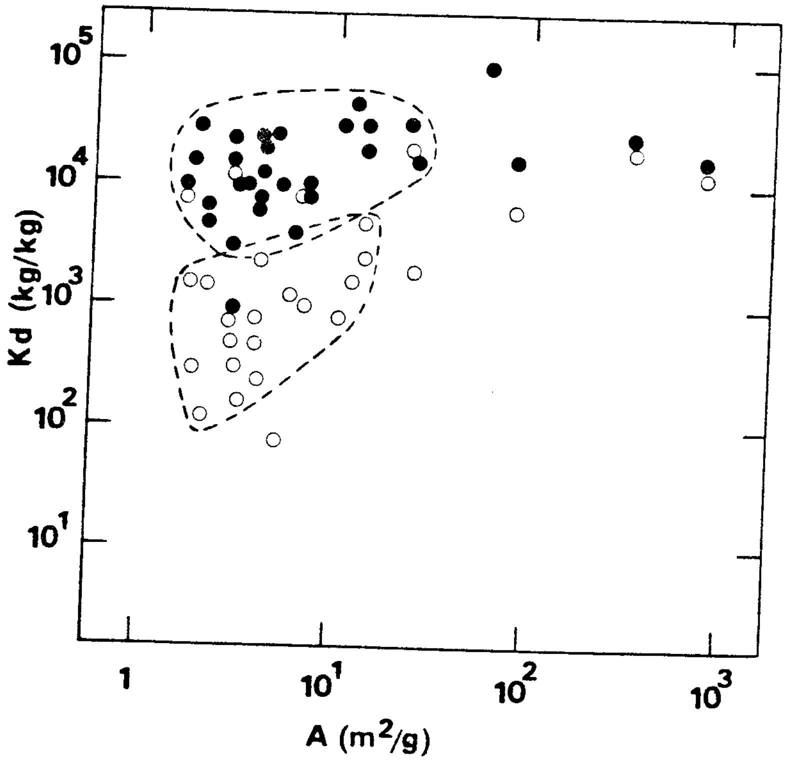


Figure 21 Kd vs A for Am at pH 8.
○ non-silicates, ● silicates.

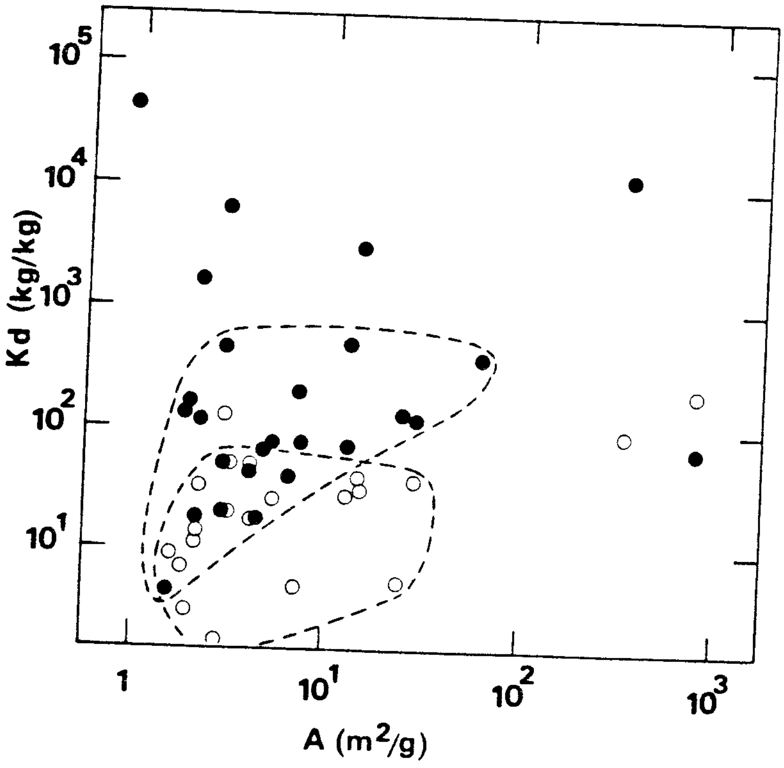


Figure 22 Kd vs A for Np at pH 8.
○ non-silicates, ● silicates.

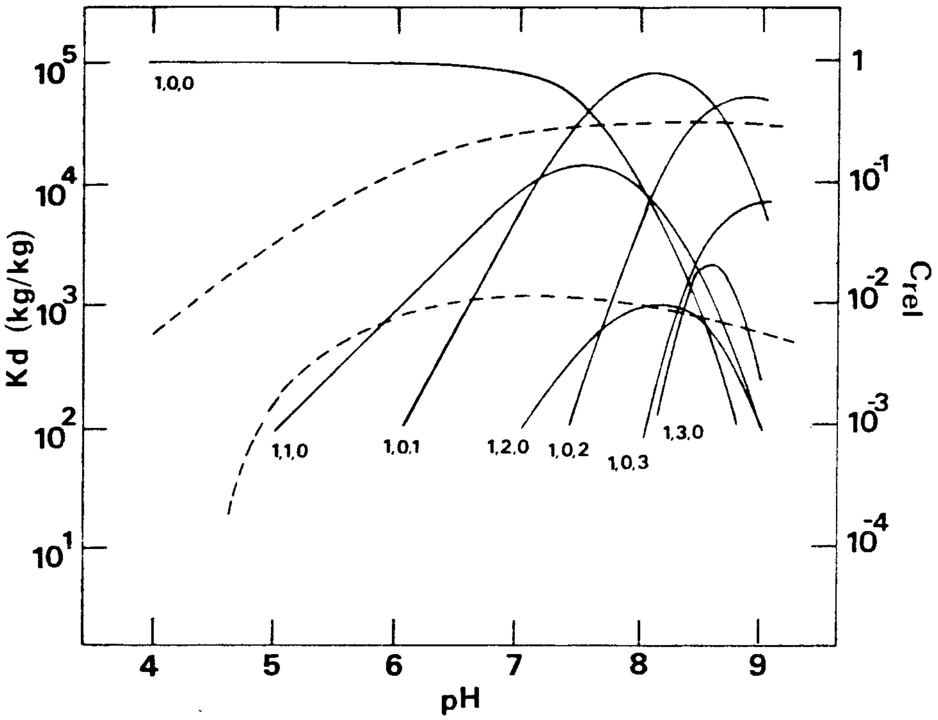


Figure 23 Kd vs speciation for Am.

Solid line = relative concentrations of $Am_x(OH)_y(CO_3)_z$.

Dashed line = Kd.

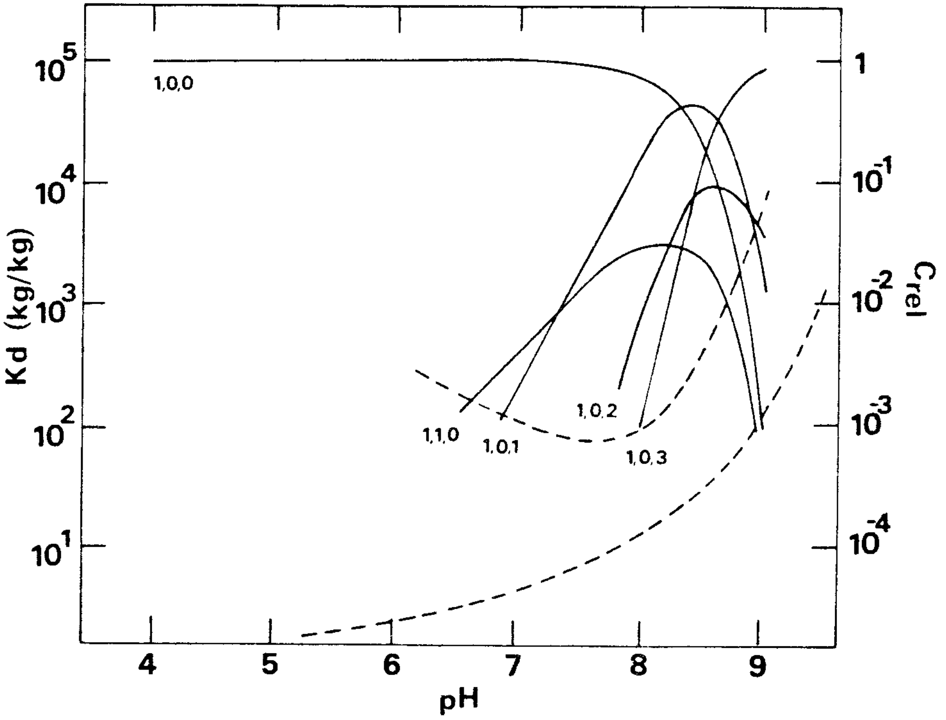


Figure 24 Kd vs speciation for Np.

Solid line = relative concentration of $(NpO_2)_x(OH)_y(CO_3)_z$.

Dashed line = Kd.

4.6. Other factors affecting the sorption

Deviations from the general pH-dependence of the sorption isotherms are obtained for a few mineral systems. These are typically high-capacity minerals giving high sorption at pH below the level of significant hydrolysis (e.g. americium - montmorillonite).

An enhanced sorption is also observed for minerals containing anions that would form sparingly soluble actinide compounds (e.g. F^- and PO_4^{3-}), probably due to chemisorption reactions.

Effects of nuclide concentrations, ionic strength, presence of complexing agents in the aqueous phase, temperature and time have previously been noticed (e.g 3,18,27,28). The influence of these factors on the sorption behaviour is discussed elsewhere and only briefly summarized below.

The variation of the surface charge of the minerals and its effect on the sorption has not been studied.

5. CONCLUSIONS

5.1. Sorption meachanisms

The experimental observations on the sorption behaviour of actinides on geologic material, both for Am, Np and Pu as presented in this report, as well as for Th, Pa and U presented elsewhere (42), can qualitatively be summarized as follows:

- For all systems the sorption is drastically increased when hydrolysis starts. Sorption maxima are generally obtained in the pH-range where neutral hydroxy complexes would dominate in solution.

- A decrease in sorption at high pH seems to coincide with the formation of anionic species (e.g. with OH^- or CO_3^{2-}).

- In the pH-range where hydrolyzed products dominate in solution the sorption is less affected by the cation exchange capacity of the solid sorbent than in non-hydrolyzed systems.

- The sorption is fairly independent of the ionic strength for actinides in their lower oxidation states in the pH-range where hydrolysis dominates. For unhydrolyzed systems the sorption is decreasing with increasing ionic strength.

- The sorption of hydrolyzed species is slowly increasing with time. The time to reach equilibrium is decreasing with increasing ionic strength and/or increasing temperature.

- The formation of colloidal fractions e.g. of clayish materials, that would lead to lower apparent distribution coefficients, is decreasing with increasing ionic strength.

- The sorption of actinides in the lower oxidation states (III, IV) is fairly independent of the nuclide concentration (for total concentrations well below the solubility product of any sparingly soluble compound). For the higher oxidation states, in cases when polynuclear anionic species can be formed (e.g. $An(VI)-CO_3^{3-}$), a drastic reduction of the sorption with increasing nuclide concentration (or concentration of e.g. CO_3^{2-}) can be obtained.

- The presence of organic complexes can lead to a reduced sorption.

Also should be pointed out the changes in sorption behaviour that would occur when the oxidation state is varied, e.g. from III or IV, which could be highly hydrolyzed in the environmental pH-range, to V, with a low degree of hydrolysis, or VI that would form strong anionic carbonate complexes.

In general, the sorption of actinides in the environmental pH-range can best be considered as a physical adsorption process, caused by non-specific forces of attraction between the sorbent and the solute. This sorption can result in the binding of the solute in several consecutive layers on the sorbent. This would be a reversible and fairly rapid process, highly pH-dependent but relatively independent of the nuclide concentration (in terms of distribution coefficients) at low total concentrations as well as of the ionic strength.

However, there are considerable contributions from electrostatic interactions (ion exchange) particularly in the low pH-range (cationic species, with a lower degree of hydrolysis) and in the high pH-range (anionic species). Such processes are dependent on both nuclide concentration and ionic strength, as well as pH.

For certain systems, specific actions of chemical forces between solute and solid would lead to chemisorption reactions which are selective, concentration dependent and possibly partly irreversible.

Sorption mechanisms will be further discussed in detail elsewhere (30).

5.2. Distribution functions for actinides in granite

The presence of "dark" minerals (biotite, amphiboles, pyroxenes) as well as weathering products (clay minerals) will largely determine the sorptive properties of granitic rocks. The variation in distribution coefficients for most of the individual major components of the rock are usually within 1-1.5 orders of magnitude, which has the consequence that the overall sorptive properties are fairly insensitive to minor variations in the mineralogic composition (c.f. Fig. 4 - 14).

The presence of fracture minerals in a weathered rock as well as of accessory non-silicate minerals would generally enhance the sorption. Usually higher distribution coefficients are obtained for a natural rock than what can be estimated from the arithmetic sum of the contributions from the individual mineral components, as calculated using data for pure minerals.

Based on the observed distribution coefficients (Appendix) and considering the composition of igneous rocks with respect to major rock forming minerals and pH in deep groundwaters the following empirical distribution functions have been formulated:

$$\log Kd = apH^2 + bpH + c \pm d \qquad (2a)$$

or

$$\log Kd = apH^2 + bpH + c + f \qquad (2b)$$

where $f = \log (mQ + nD + o) \pm 0.2$ (3)

Here a,b,c,d,m,n and o are constants

Q = fraction of quartz

D = fraction of "dark" minerals (biotite, hornblende, etc.).

From (2a) values for an "average" granite (c.f. Fig. 1) would be estimated, the constant d indicating the range of observed values.

In (2b) an effort is made to give a simple estimate of Kd for a granite with known composition with respect to the low-sorbing quartz and the high-sorbing "dark" minerals.

Calculated empirical values of the constants in eqn. (2) are given in Table 5, and estimated distribution coefficients in Table 6.

Table 5 Constants in eqn. (2)

Element	a	b	c	d	m	n	o
Th(IV)[a]	-0.0451	0.747	1.16				
Pa(V)[a]	-0.172	3.099	-9.33				
U(VI)[b])	-0.0349	0.483	1.31	0.35	-0.26	1.53	0.71
Np(V)	0.469	-6.759	25.47	0.35	-0.26	1.80	0.71
Pu[c]	-0.0699	1.049	-0.22	0.45	-0.35	5.60	0.71
Am(III)	-0.0562	1.207	-2.13	0.40	-0.62	1.80	0.71

[a] Data estimated from measurements on Al_2O_3 and SiO_2 only (27) $[Th] = 2.5 \times 10^{-9}M$, $[Pa] = 4 \times 10^{-12}M$

[b] From 18. $[U] = 9.5 \times 10^{-12}M$

[c] Predominantly Pu(IV), but probably with some contributions from Pu(V).

Table 6 Estimated distribution coefficients (Kd, kg/kg)[a]

Element	pH			
	7	8	8.2[b]	9
Th(IV)[c]	15000	18000	18000	17000
	11000	13000	13000	12000
Pa(V)[c]	8600	28000	23000	43000
	6100	20000	23000	30000
U(VI)[d]	960	870	840	680
	680	620	590	480
Np(V)	14	26	38	420
	10	19	28	310
Pu[e]	5000	5000	4800	3600
	4400	4400	4300	3200
Am(III)	3700	8500	9700	15000
	2300	5200	6000	9300

[a] Upper value obtained by eqn. 2a. This would correspond to an "average" granite. Lower value obtained by eqn. 2b, assuming a silica rich rock (30% quartz, 5% dark minerals). This could be considered as a conservative "reference" value representative of the lower range of expected values (but not minimum).

[b] pH in the artificial groundwater used as a reference system (Table 3).

[c] See foot-note a, Table 5.

[d] See comment in text on the concentration dependence.

[e] See foot-note c, Table 5.

Data for oxidizing and reducing conditions that could serve as reference values for granitic groundwater systems are given in Table 7.

Table 7 Reference distribution coefficients at pH 8.2 (upper and lower values as in Table 6)

Element	Kd(kg/kg) Oxic conditions	 Reducing conditions
Th	18000 13000	18000 13000
Pa	33000 23000	(18000)[a] (13000)
U	840[b] 590	(4800)[c] (4300)
Np	38 28	(18000)[a] (13000)
Pu	4800[d] 4300	(9700)[e] (6000)
Am	9700 6000	9700 6000

[a] Assuming tetravalent state and a sorption behaviour similar to Th. Probably higher values would be expected for Pa, considering the hydrolysis properties of this element.

[b] At low uranium concentrations and closed system (see comments in text).

[c] Predominantly U(IV), but possibly some contribution from higher oxidation states, similar to plutonium.

[d] See foot-note c, Table 5.

[e] Assuming trivalent state and a sorption behaviour similar to Am.

Eqn. (2) and the values in Table 5-7 would be valid under the following assumptions:

o The aqueous phase is a typical groundwater, not necessarily of low salinity, but with pH in the 7-9 range.

o The solid phase is granite.

o The total nuclide concentration is well below maximum solubility, as determined by any sparingly soluble compound.

Thus, effects of e.g. nuclide concentration, presence of strong complexing agents, temperature and time, etc. are neglected. Significant increases of the distribution coefficient with time (by up to one order of magnitude) have been observed, particularly for the actinides in their lower oxidation states and at pH where neutral species would dominate.

For rocks rich in "dark" minerals, weathering products or chemisorbing accessory minerals considerably higher distribution coefficients would be obtained than the "average" values in Table 6 (by up to 1.5 orders of magnitude), which is obvious when considering the sorption data for the individual minerals (C.f. Appendix).

It is not likely that rock compositions in igneous rocks would be encountered that would give distribution coefficients well below the "reference" values. Thus, the values in Table 6 and 7 would reasonably well define the lower range of expected actinide distribution coefficients in granite-groundwater systems.

An exception is the uranium system. At high nuclide concentrations and/or high carbonate concentrations under oxic conditions (e.g in systems open to air) much lower distribution coefficients (by up to 2 orders of magnitude) have been reported in the literature.

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In particular the cooperation with Dr G.W. Beall and Ms T. Krajewski at Oak Ridge as well as the support and interest from Dr R.L. Hahn and the staff members at the Transuranium Research Lab. should be mentioned, and of course also the cooperation within the group at Chalmers University that is still working on the project.

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DISTRIBUTION COEFFICIENTS

Distribution coefficients (Kd, kg/kg) for Am(III), Np(V) and Pu as a function of pH are given in Figure 1-1 to 1-9 (Am), 1-10 to 1-18 (Np) and 1-19 to 1-21 (Pu). Aqueous phase is groundwater (GW) or 4 M NaCl. Concentrations etc. are given in Table 1-1.

Table 1-1 Conditions for distribution measurements

Solid phase: Minerals according to Table 3.	
Particle size fraction (after sieving)	0.044-0.063 mm
Amount of solid in batch experiments	0.5-1 g [a]
Aqueous phase: Artificial groundwater (Table 2) or 4 M NaCl.	
Volume in batch experiments	40-50 ml [a]
Solid/liquid ratio	6-15 g/l [a]
Initial radionuclide concentrations	$(1.8\text{-}5.0)\times 10^{-9}$ M (Am) $(2.0\text{-}5.3)\times 10^{-11}$ M (Np)[a,b] $1{,}8\times 10^{-11}$ M (Pu)[a,b]
Other parameters	
Temperature	22±2°C
Contact time	5d
Phase separation	Centrifugation (1h, 4000g)
Oxic conditions	
Equipment	Polycarbonate, polypropylene

[a] Exact amounts of solid, aqueous phase as well as nuclide concentration for the ca 1100 measurements given in Fig. 1-1 to 1-21 are available, but not given in this report.

[b] Also some preliminary measurements with concentration around 10^{-8} M.

All curves in Figure 1-1 to 1-21 are arbitrarily drawn.

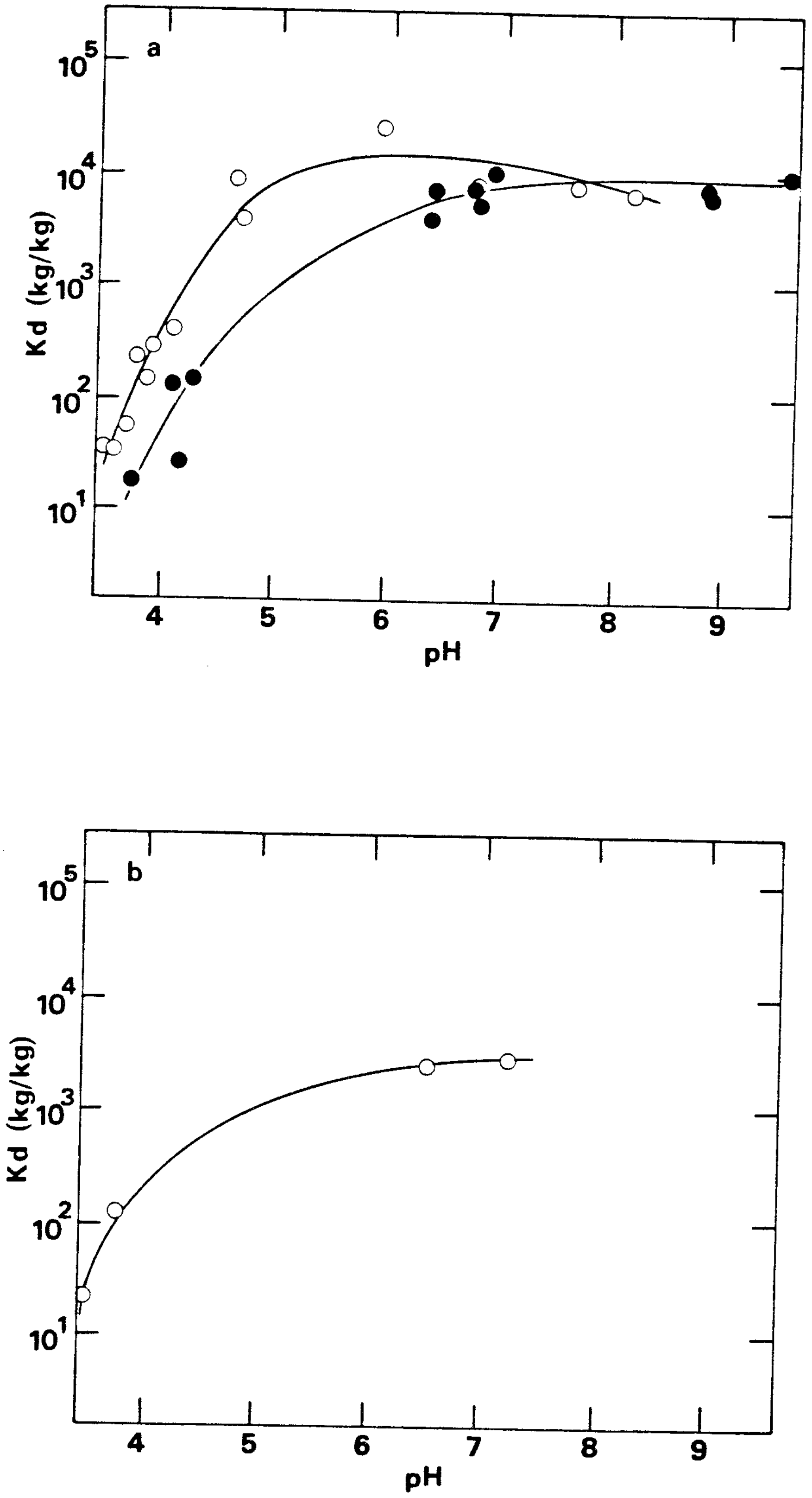


Figure 1-1 Kd for Am in GW (a), 4 M NaCl (b) - sulfides.
○ pyrite, ● chalcopyrite

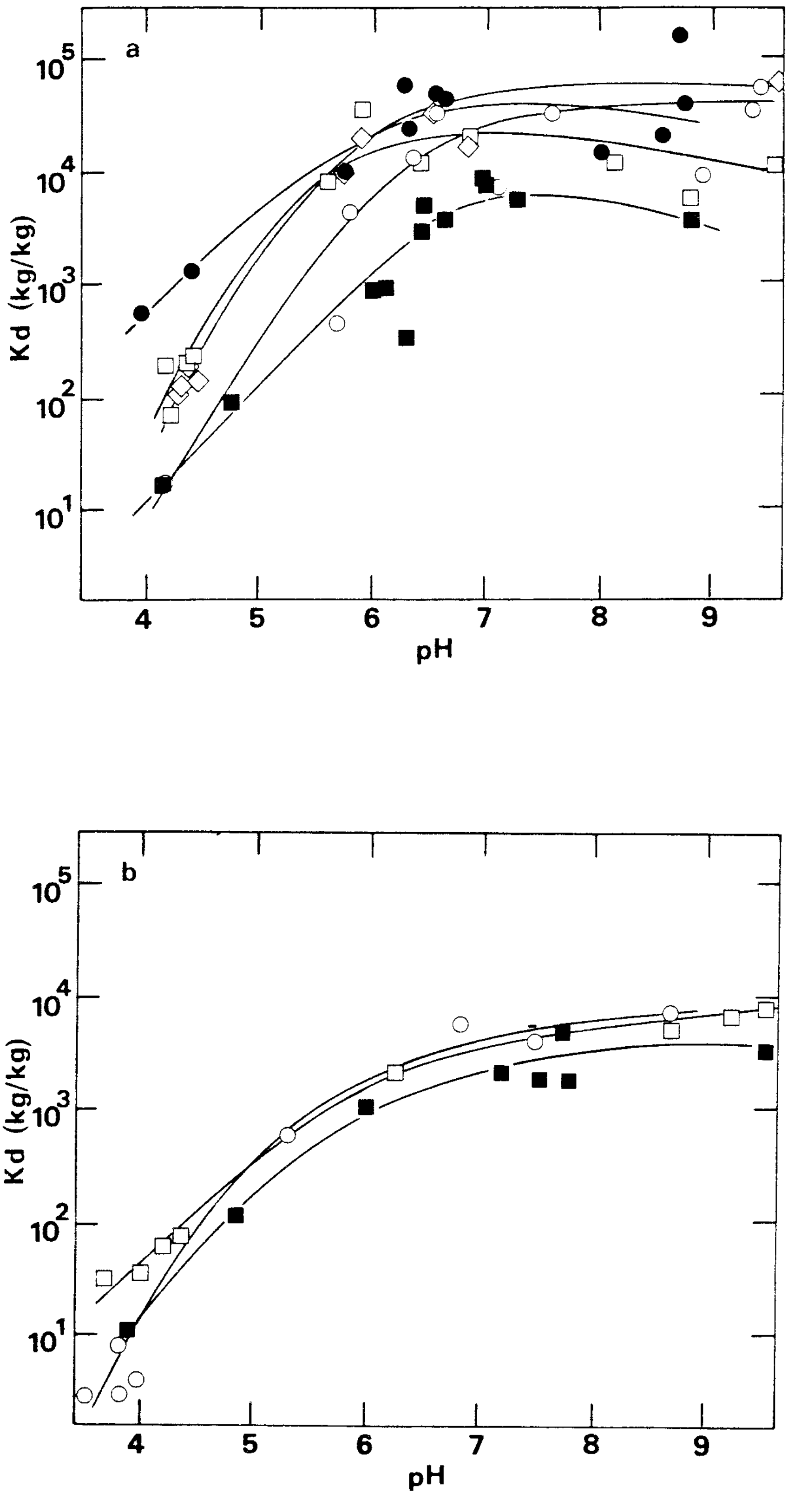


Figure 1-2 Kd for Am in GW (a), 4 M NaCl (b) - oxides.
○ magnetite, ● limonite, □ hematite, ■ corundum,
◇ gibbsite

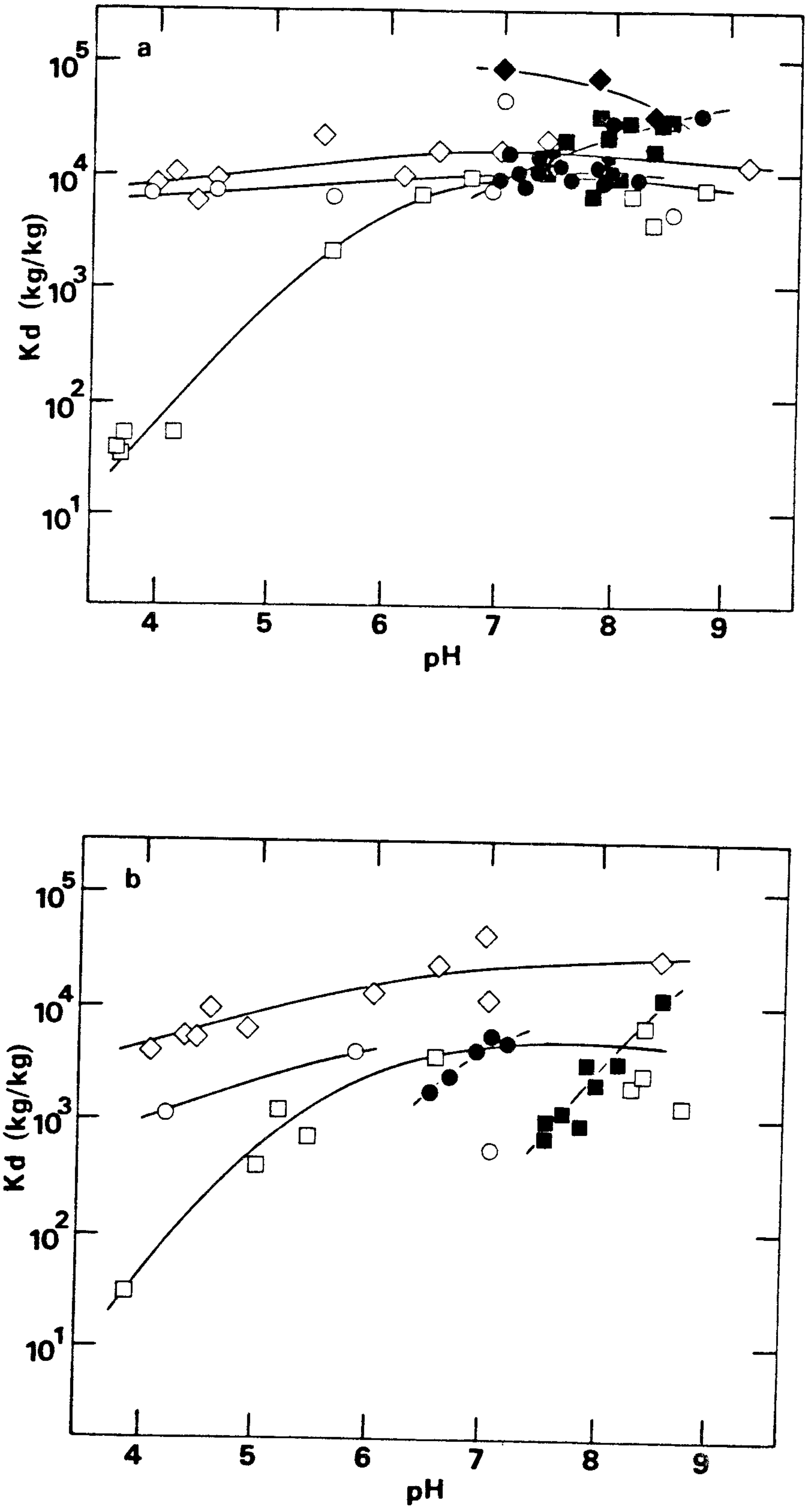


Figure 1-3 Kd for Am in GW (a), 4 M NaCl (b) - fluorides, sulfates, carbonates and phosphates.
○ fluorite, □ anhydrite, ● dolomite, ■ calcite,
◇ apatite, ◆ monazite

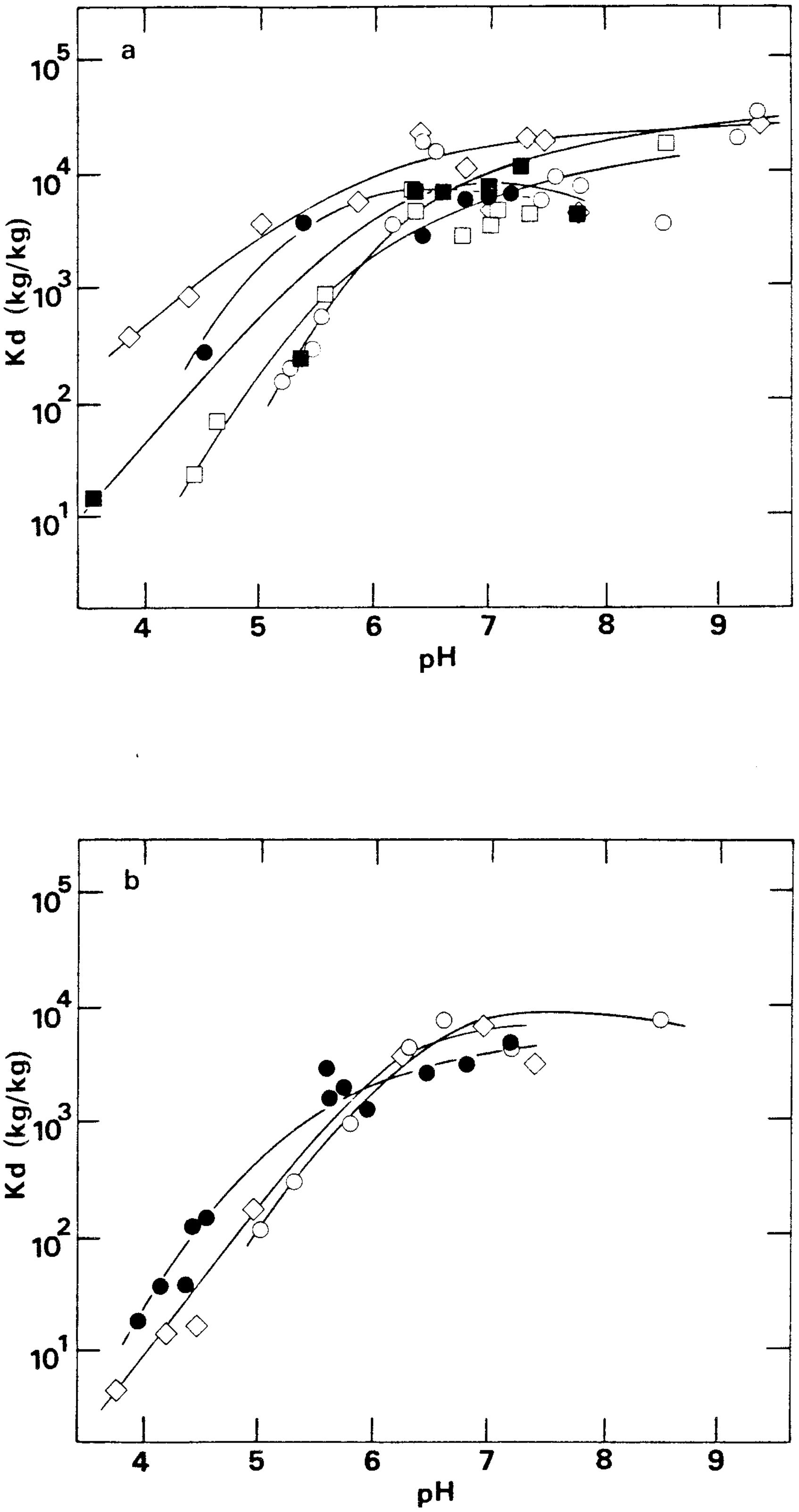


Figure 1-4 Kd for Am in GW (a), 4 M NaCl (b) - neso and soro silicates.
○ olivine, ● almandine, □ sphene, ■ zircon,
◇ epidote

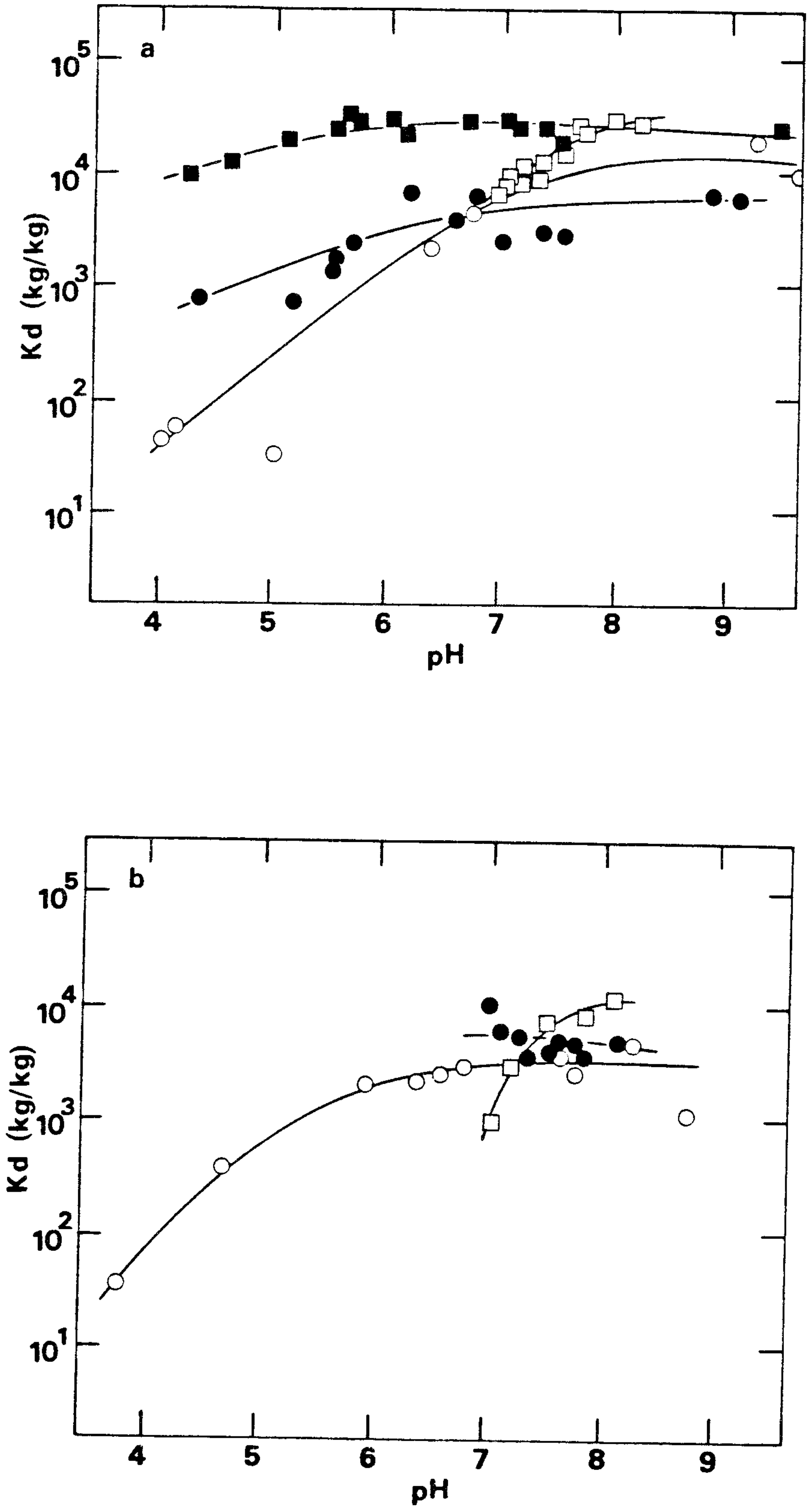


Figure 1-5 Kd for Am in GW (a), 4 M NaCl (b) - cyclo and ino silicates.

○ beryl, ● augite, □ hornblende, ■ attapulgite

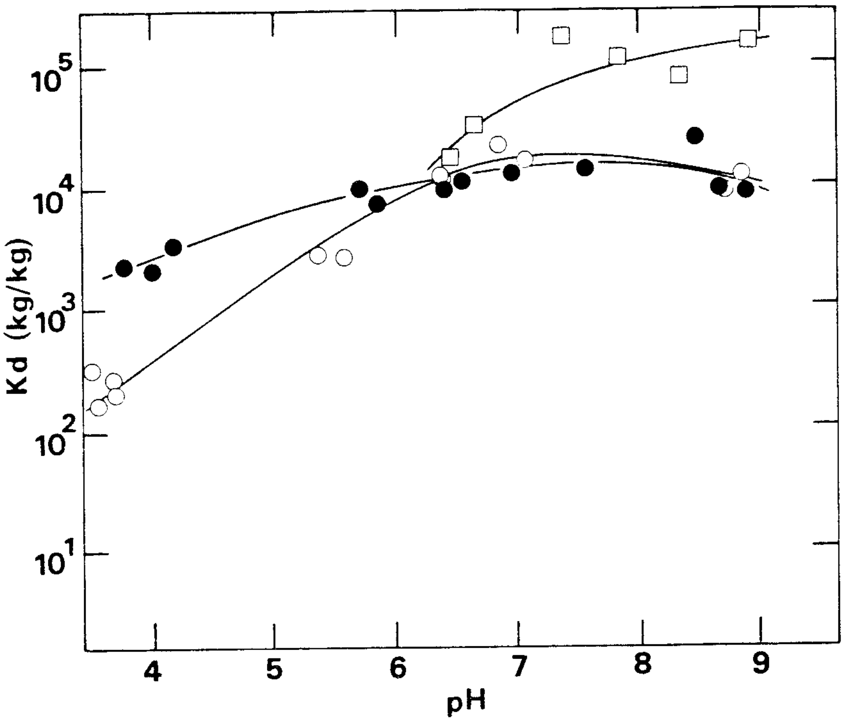


Figure 1-6 Kd for Am in GW - phyllo(2) silicates.
○ kaolin, ● halloysite, □ serpentine

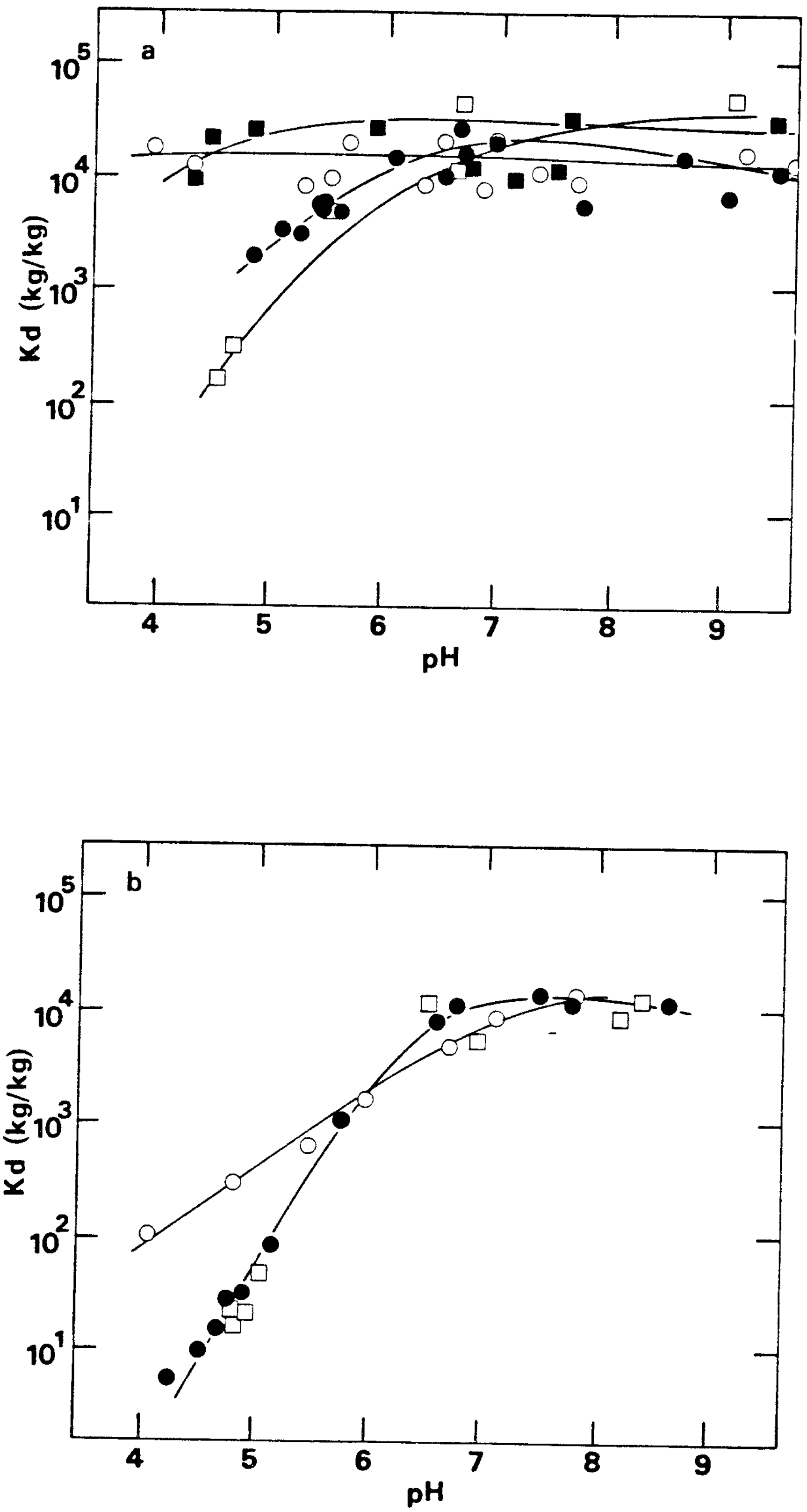


Figure 1-7 Kd for Am in GW (a), 4 M NaCl (b) - phyllo(3) silicates.
○ montmorillonite, ● biotite, □ muscovite,
■ chlorite

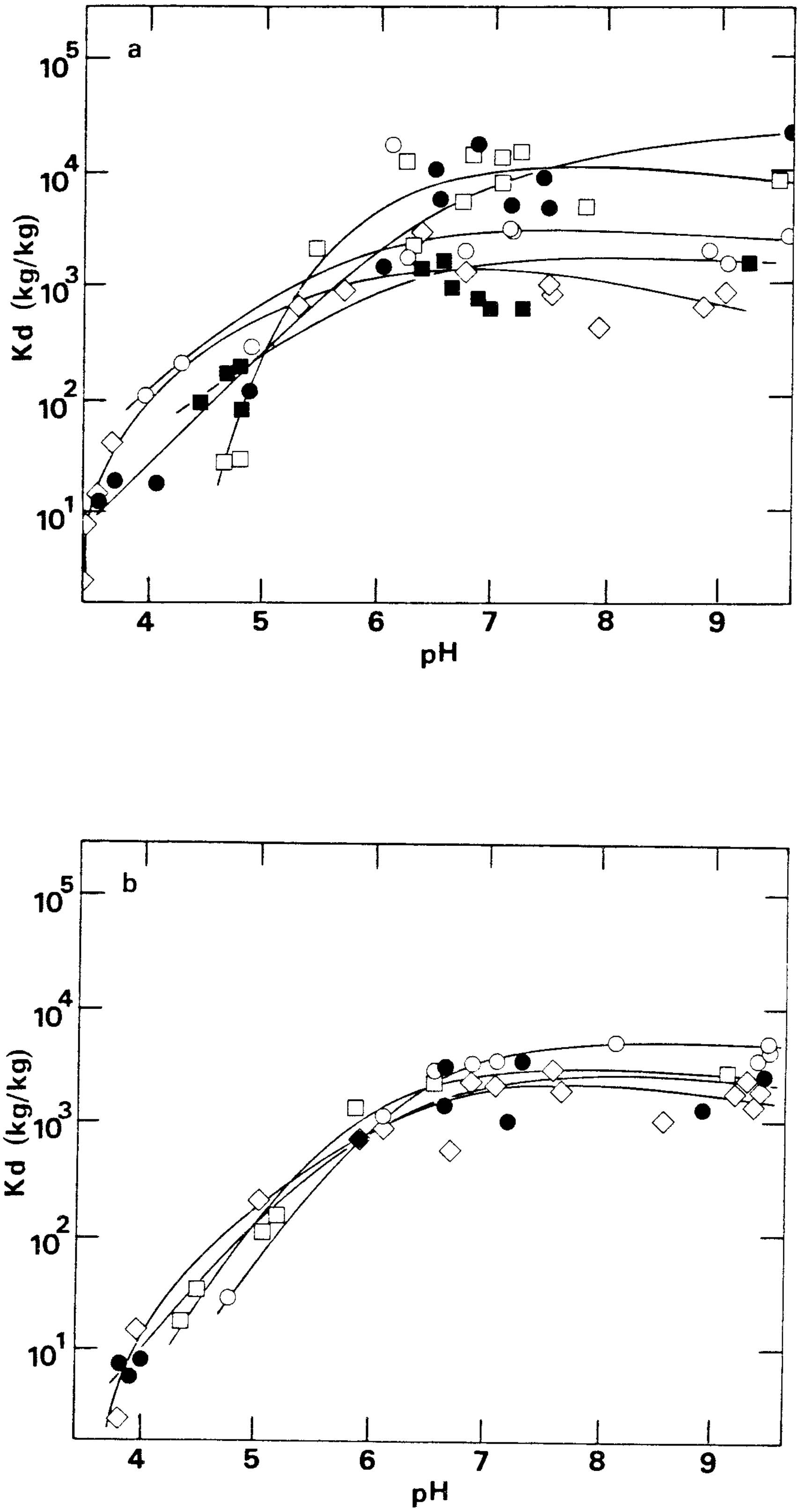


Figure 1-8 Kd for Am in GW (a), 4 M NaCl (b) - tecto silicates.
○ albite, ● microcline, □ bytownite, ■ anorthite,
◇ quartz

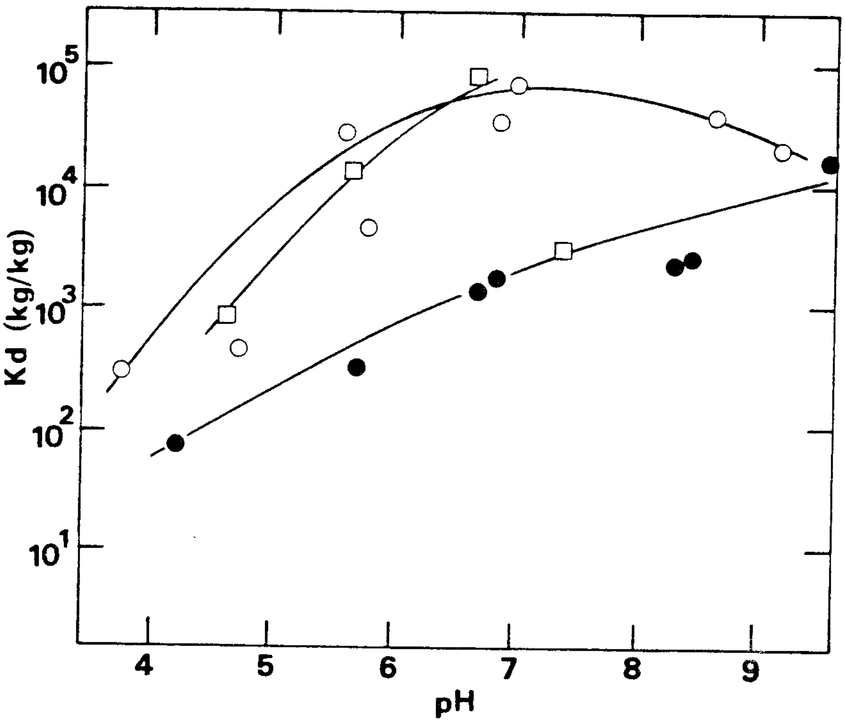


Figure 1-9 Kd for Am in GW - igneous rocks.
○ granite (Climax Stock), ● granite (Westerly),
□ basalt

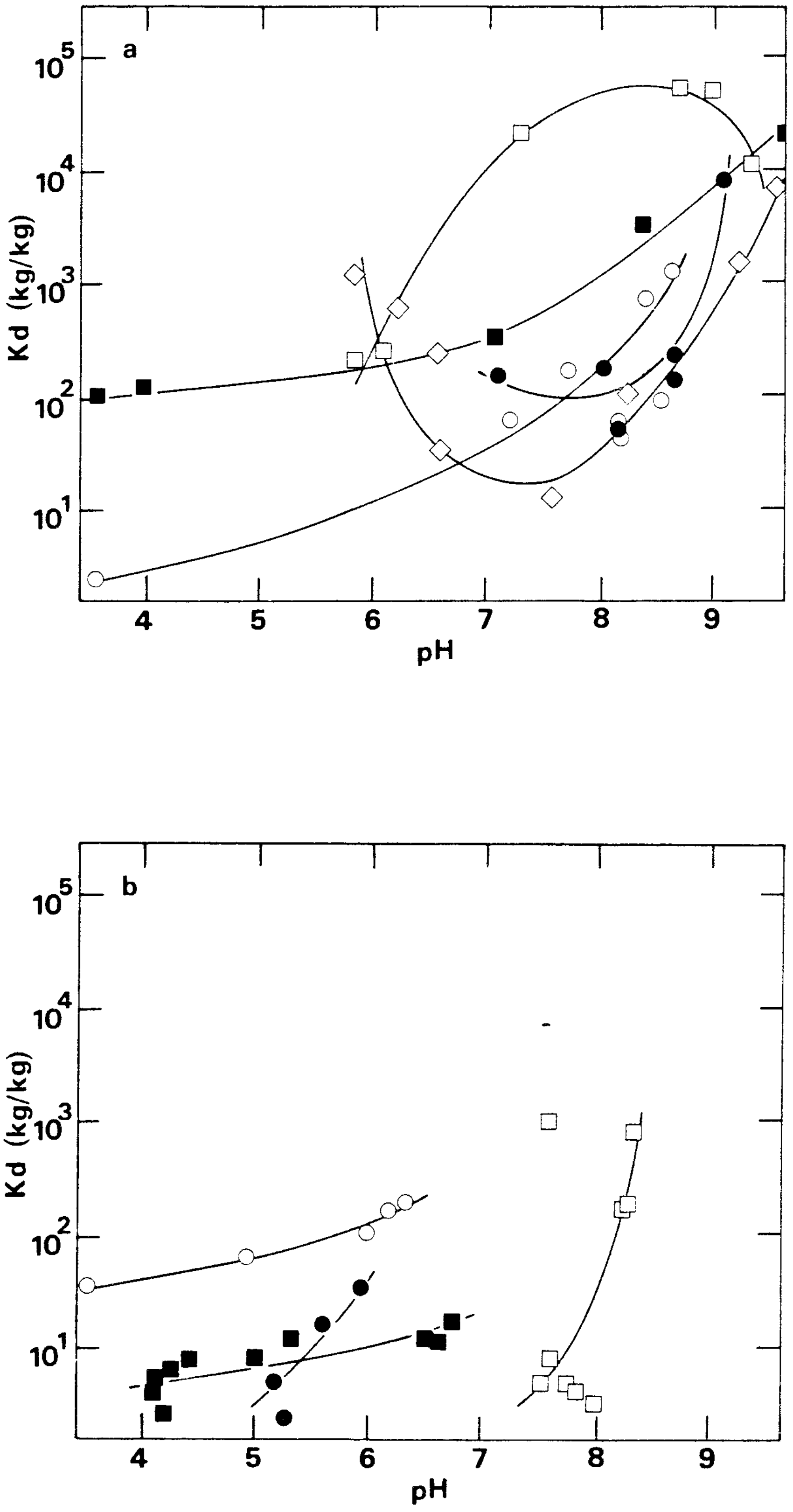


Figure 1-10 Kd for Np in GW (a), 4 M NaCl (b) - sulfides.
○ pyrite, ● chalcopyrite, □ galena, ■ molybdenite,
◇ chalcosite

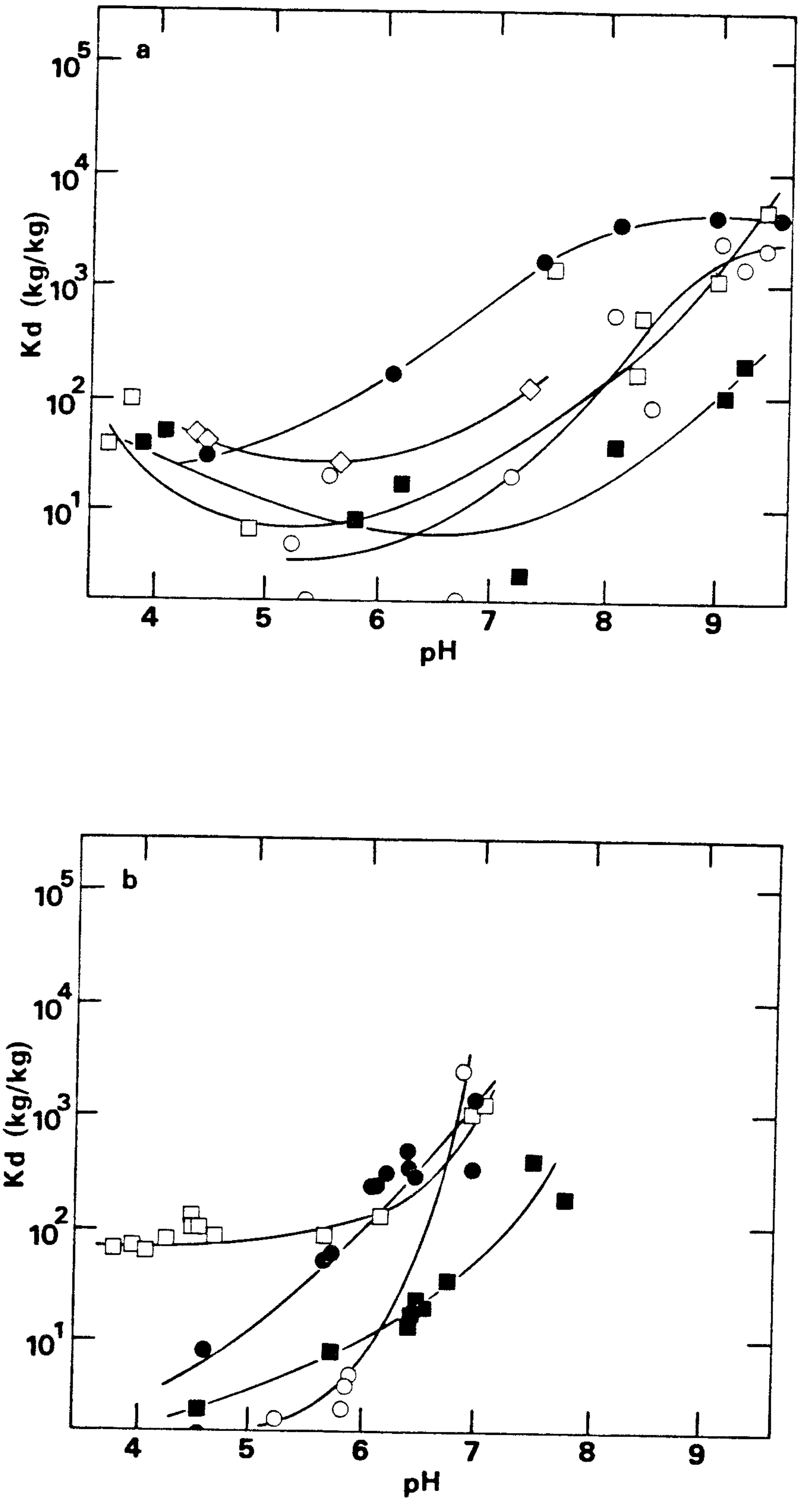


Figure 1-11 Kd for Np in GW (a), 4 M NaCl (b) - oxides.
○ magnetite, ● limonite, □ hematite, ■ corundum,
◇ gibbsite

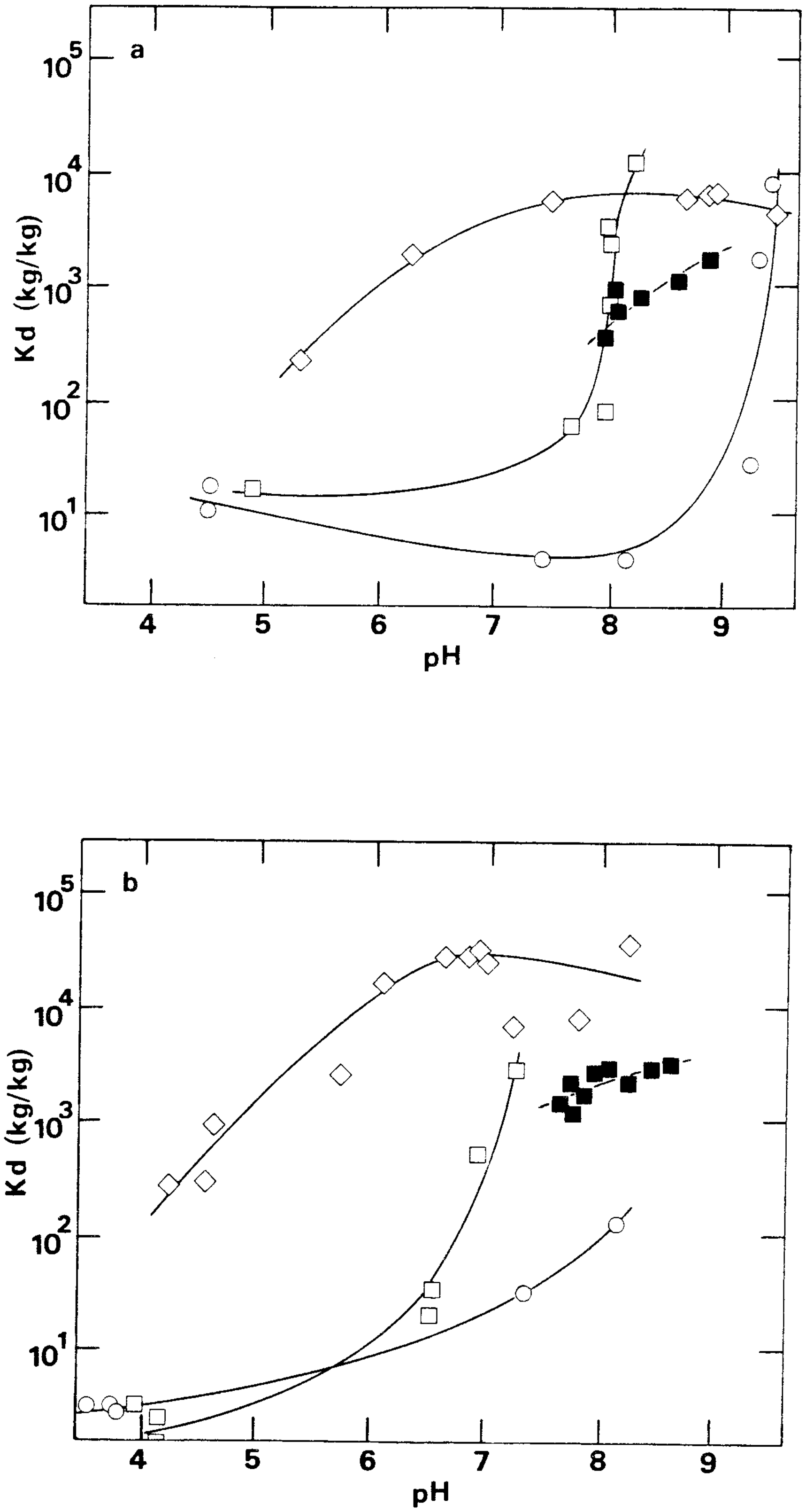


Figure 1-12 Kd for Np in GW (a), 4 M NaCl (b) - fluorides, sulfates, carbonates and phosphates.
○ fluorite, □ anhydrite, ■ calcite, ◇ apatite

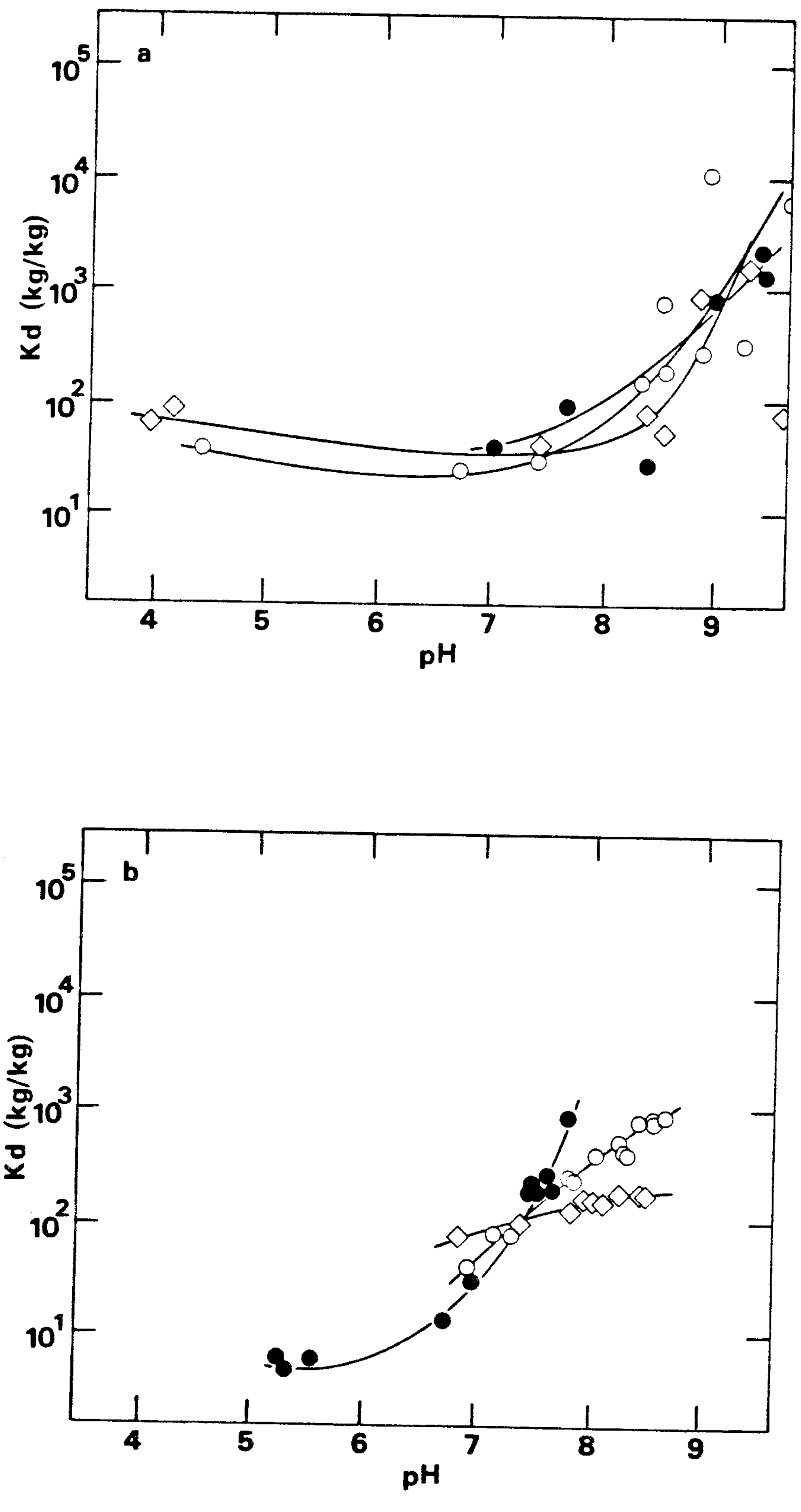


Figure 1-13 Kd for Np in GW (a), 4 M NaCl (b) - neso and soro silicates.
○ olivine, ● almandine, ◇ epidote

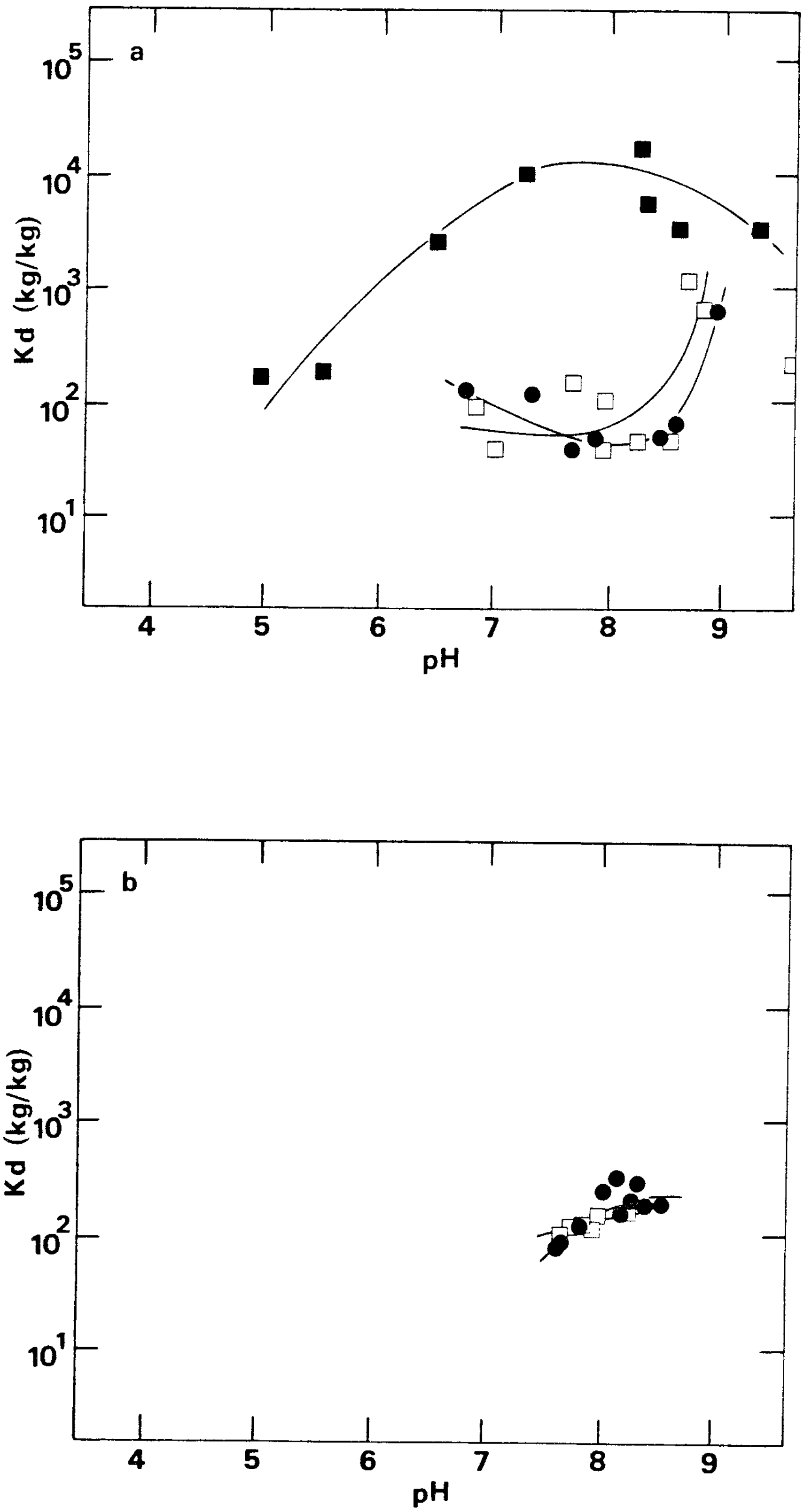


Figure 1-14 Kd for Np in GW (a), 4 M NaCl (b) - ino silicates.
● augite, □ hornblende, ■ attapulgite

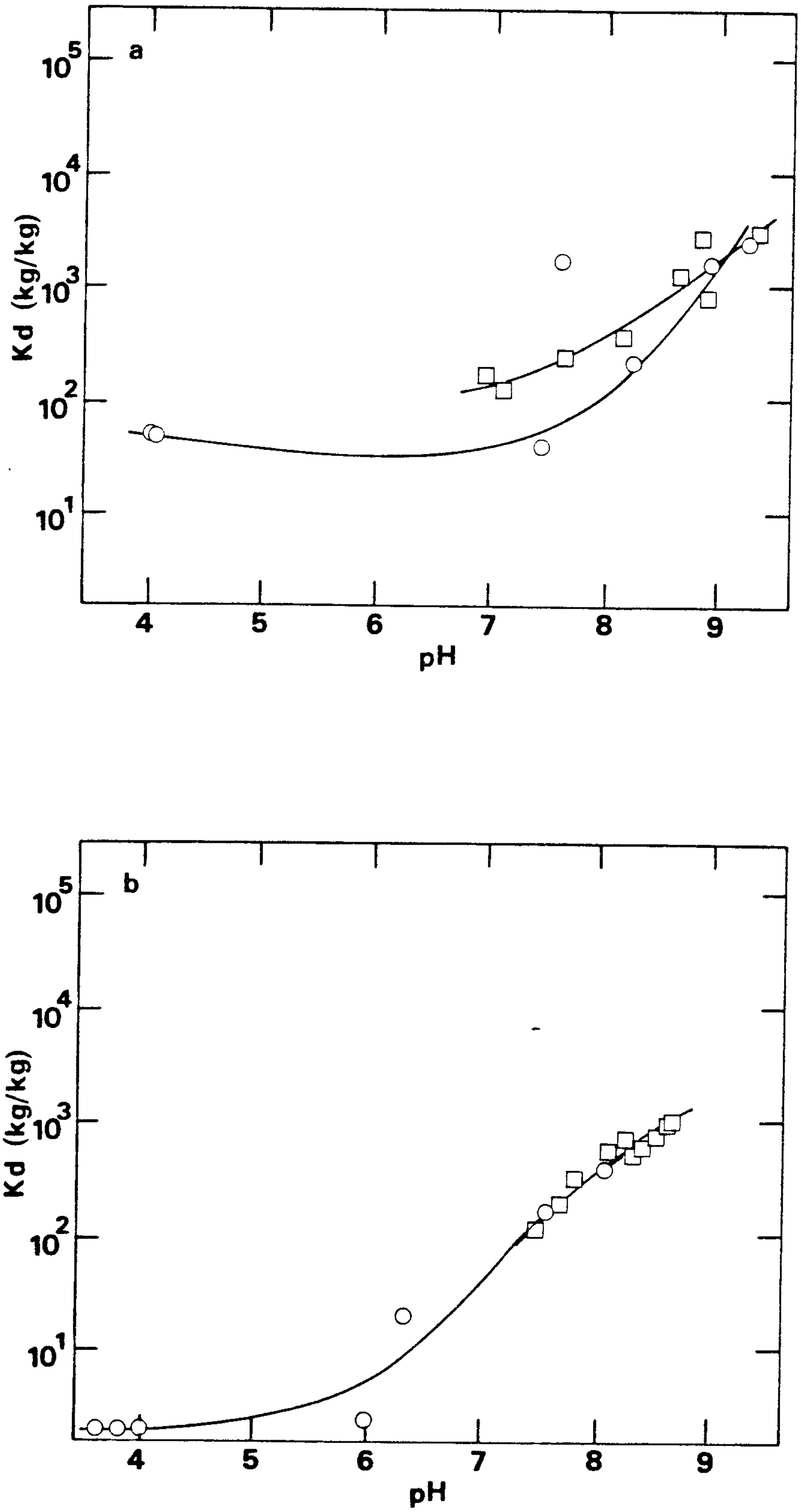
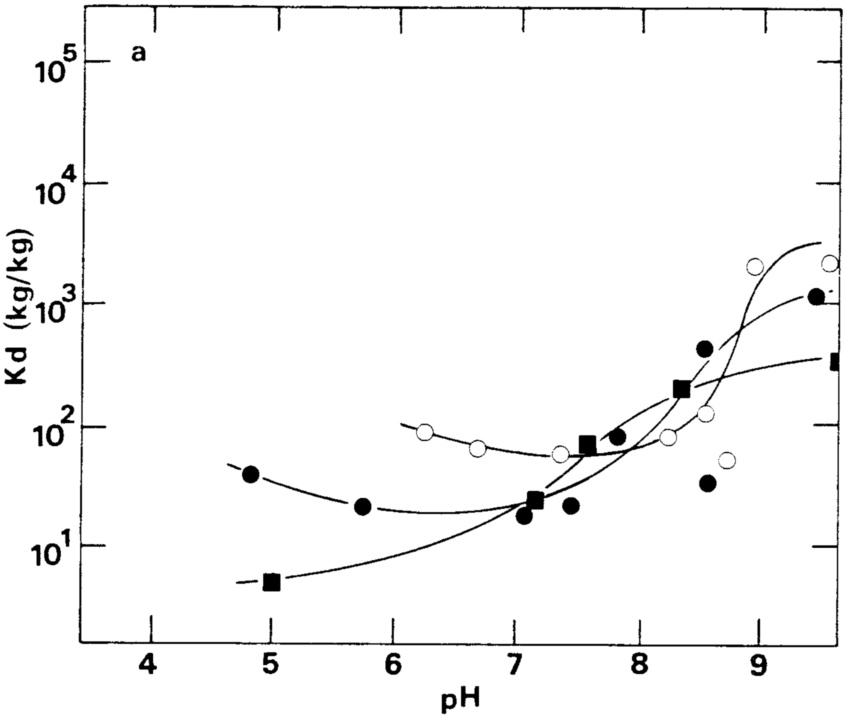


Figure 1-15 Kd for Np in GW (a), 4 M NaCl (b) - phyllo(2) silicates.
○ kaolin, □ serpentine



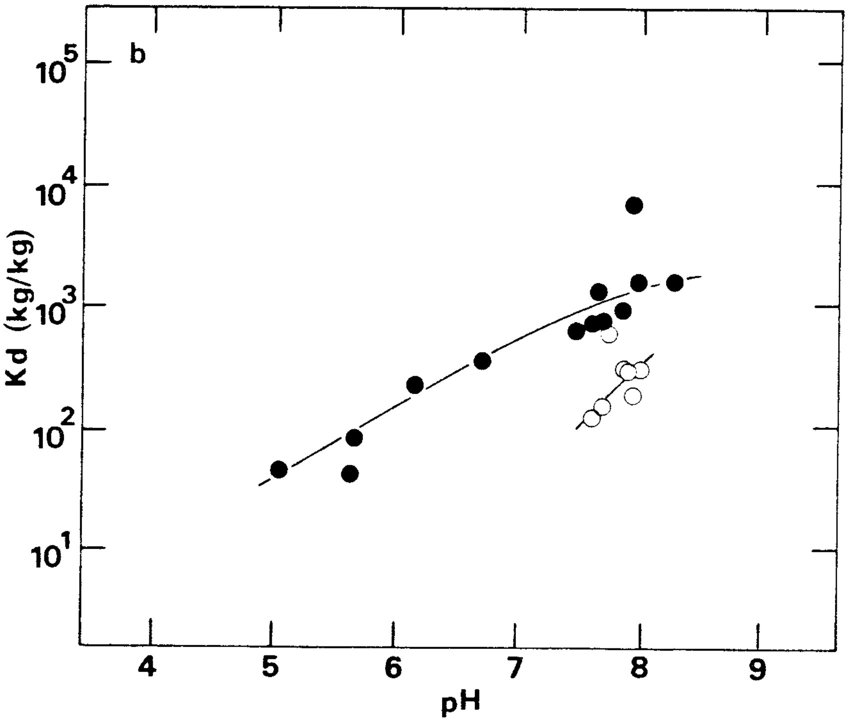


Figure 1-16 Kd for Np in GW (a), 4 M NaCl (b) - phyllo(3) silicates.

○ montmorillonite, ● biotite, ■ chlorite

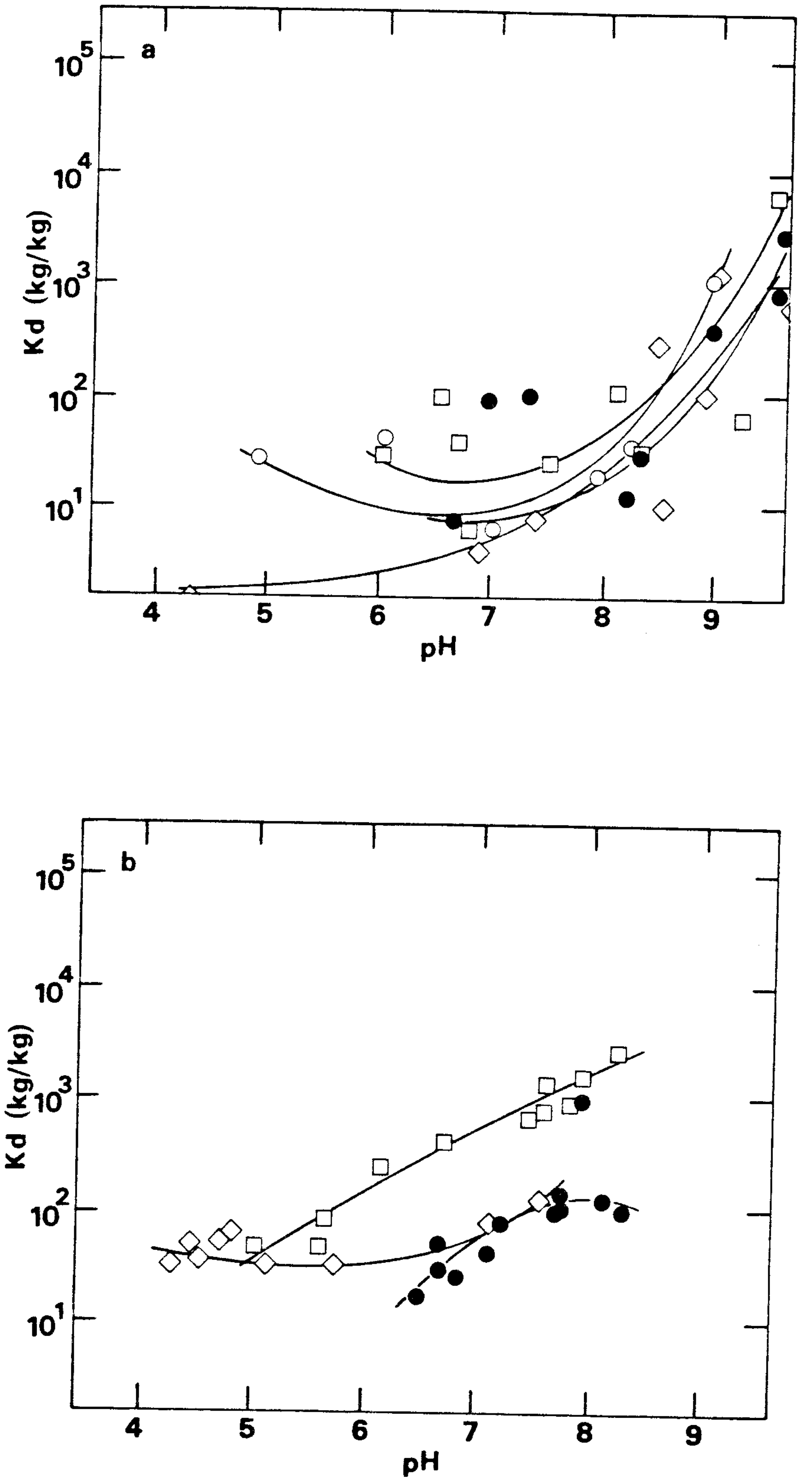


Figure 1-17 Kd for Np in GW (a), 4 M NaCl (b) - tecto silicates.
○ albite, ● microcline, □ bytownite, ◇ quartz

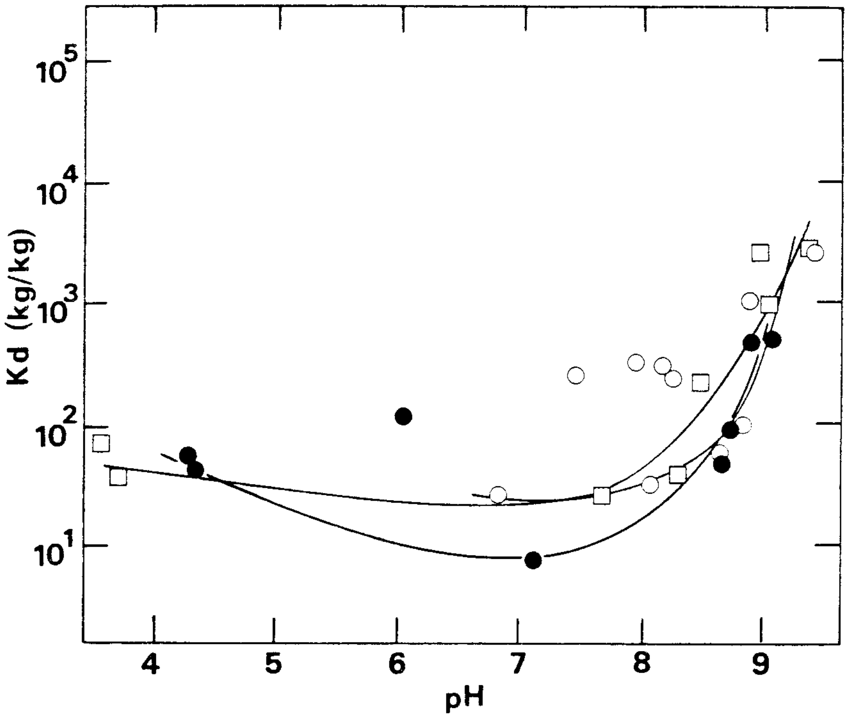


Figure 1-18 Kd for Np in GW - igneous rocks.
○ granite (Climax Stock), ● granite (Westerly),
□ basalt

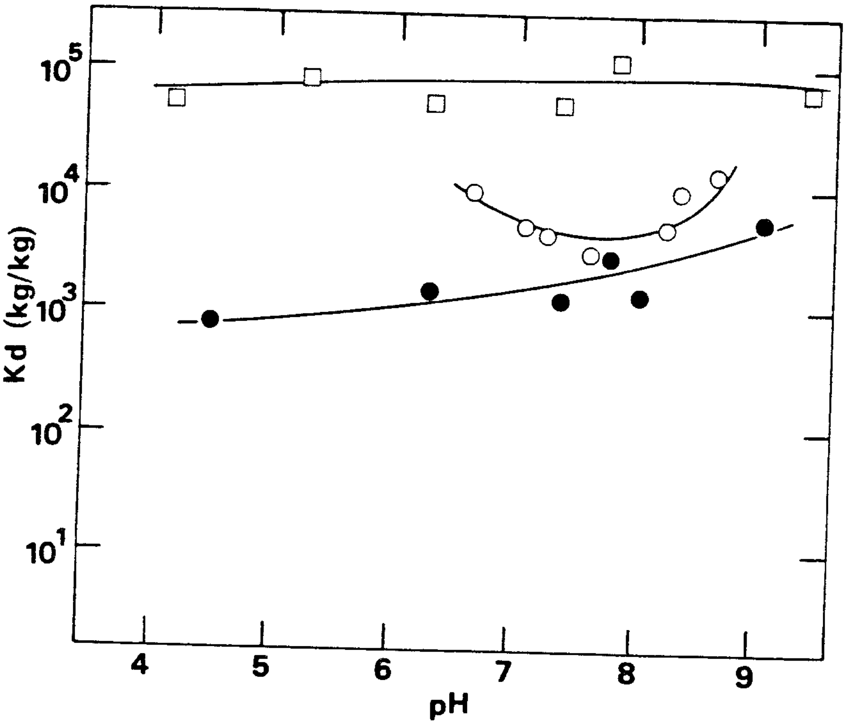


Figure 1-19 Kd for Pu in GW - non-silicates.
○ pyrite, ● magnetite, □ apatite

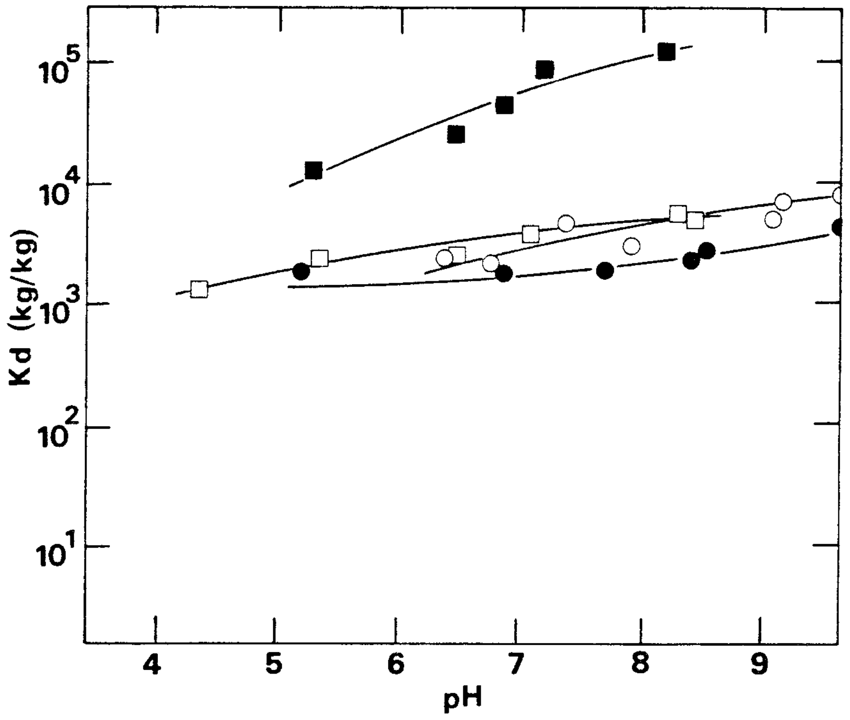


Figure 1-20 Kd for Pu in GW - neso and ino silicates.
○ olivine, ● almandine, □ hornblende, ■ attapulgite

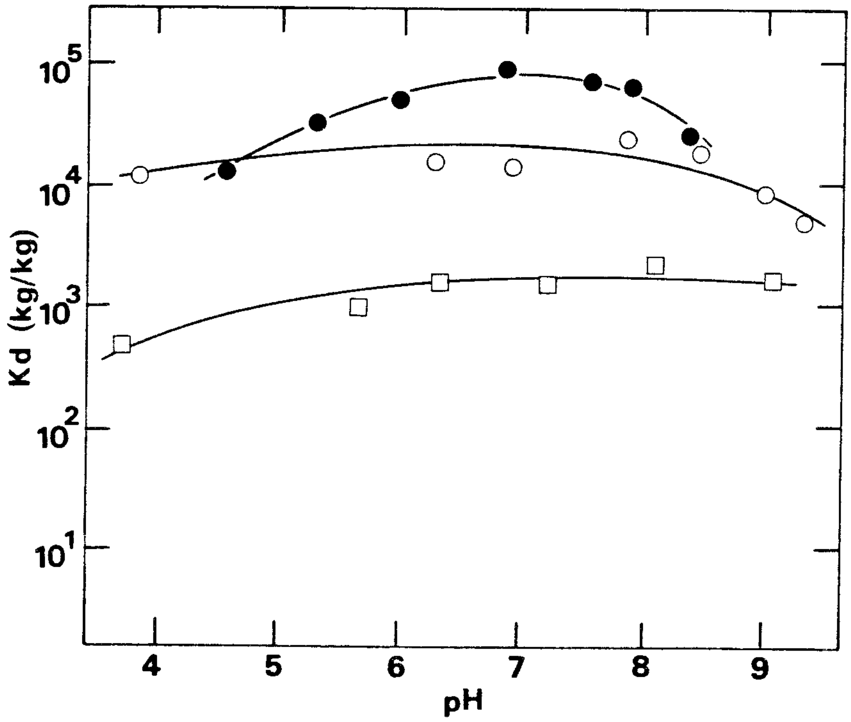


Figure 1-21 Kd for Pu in GW - phyllo and tecto silicates.
○ montmorillonite, ● biotite, □ quartz

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Ove Persson
Geological Survey of Sweden, Uppsala
Ove Landström
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A review of biological factors of relevance for assessment of absorbed dose at long term waste storage
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