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Frau Prof. Dr. Emilie Jäger gewidmet

Distribution of exchangeable cations in Mesozoic and Permo-Carboniferous sediments in the subsurface of Northern Switzerland

by *Tj. Peters*¹

Abstract

The exchangeable cations Na⁺, Ca²⁺, Mg²⁺ and K⁺ were determined in 91 samples of sedimentary rocks from deep drillholes in the Tabular Jura mountains in Northern Switzerland.

The total exchange capacities vary between 10–30 meq / 100 g and no obvious correlations with clay mineral variations were detected, which is attributed to the absence of smectites as major components. A general decrease in CEC with maximum burial depth was observed.

The cation occupancy in formations with deep groundwaters are quite constant and indicate water – rock interaction by cation-exchange. The original site occupancy must have taken place during early diagenesis.

Keywords: Cation exchange, exchange capacity, groundwater, diagenesis, Jura mountains, Switzerland.

Introduction

Cation exchange capacities of clay minerals have, for a long time, been mainly of interest in soil science. Lately, in the course of the search for suitable sites for waste disposal it has been realised, that the CEC of such minerals is an extremely important parameter for the retardation capacity of host rocks. Hydrochemists to some extent explain changes in chemistry of groundwaters by ion exchange with clay minerals in the aquifers (THORSTENSON et al., 1979; STUMM and MORGAN, 1981).

During the deep drilling programme of NAGRA (National Cooperative for the Storage of Radioactive Waste) in Northern Switzerland (Fig. 1) the cation exchange capacity of 91 samples of clay-bearing sediments was determined. This gave the opportunity to verify if the exchange capacity and the distribution of cations were related to formation, depth, mineralogy, groundwater or rock chemistry.

Methodology

Pieces of 2 to 3 kgs of air dried drillcore material were crushed to 5 mm size and split, whereof 500 g were ground to less than 100 micron. The samples were treated with 0.2 N BaCl₂ solution, the pH buffered at 8.7 with Triethanolamine. In the solution, the exchanged amounts of Na, K, Ca and Mg were determined by AAS. Results from samples containing evaporite minerals such as halite, gypsum or anhydrite had to be eliminated due to the water solubility of these minerals. Cations in trapped pore fluids, however, are included in the given amounts. In the extreme cases like the "Opalinus-Ton", where Na contents of the pore fluid can attain 6000 mg/l, with a porosity of 10 vol.%, this corresponds to about 1 meq / 100 g. Compared to the measured values of 5 to 6 meq / 100 g, this makes up to 20% of the "exchangeable" sodium. With the high pH of the buffer solution neither calcite nor dolomite seem to go into solution. Neglecting other absorbed ions like Sr and Ba

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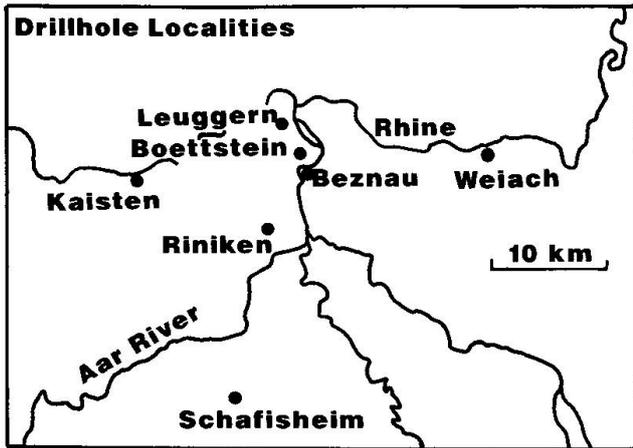


Fig. 1 Investigated drillhole localities.

the total CEC capacity is taken as the sum of exchangeable Na, K, Ca and Mg.

Results

The analytical results of the cation exchange, together with detailed sample description, mineralogy, clay mineralogy and geochemistry of the Beznau, Böttstein, Weiach, Riniken, Kaisten, Schafisheim and Leuggern drillholes are published in NAGRA (1984), PETERS et al. (1987a), MATTER et al. (1987a), MATTER et al. (1987b), PETERS et al. (1987b), MATTER et al. (1988) and Peters et al. (1987c) and summarised in tables 1, 2. From these the relative percentages of K^+ , Na^+ , Mg^{2+} and Ca^{2+} were calculated. The total exchange capacity and the occupancy are represented against depth in Figs 2 a, b, c.

TOTAL EXCHANGE CAPACITY

The total exchange capacity varies between 10 and 30 meq / 100 g with total clay mineral contents between 40 and 80 weight%. Within certain formations like the Rotliegendes, which mainly consists of detrital minerals (quartz, feldspars and clay minerals), the exchange capacity is positively correlated with the total clay mineral content. In carbonate rich samples such a relationship is much less evident.

Comparing the total exchange capacities of the different formations (see Fig. 2) it becomes evident that the values for the samples from the Carboniferous do not exceed 15 meq / 100 g and those from the Rotliegendes are not above 20 meq / 100 g, even with clay mineral contents up to 90 weight%. In the Mesozoic, however, even with clay mineral contents as low as 40 weight%, exchange capacities of 30 meq / 100 g are obtained.

No obvious relationship between cation exchange capacity and variations in clay mineral distribution can be detected. This is mainly due to the fact that clay minerals with extremely high exchange capacities like smectites do not occur in many samples. Among the clay minerals illite is predominating, kaolinite contents are strongly variable, chlorite and illite/smectite mixed layers are generally present in minor amounts (5–15%).

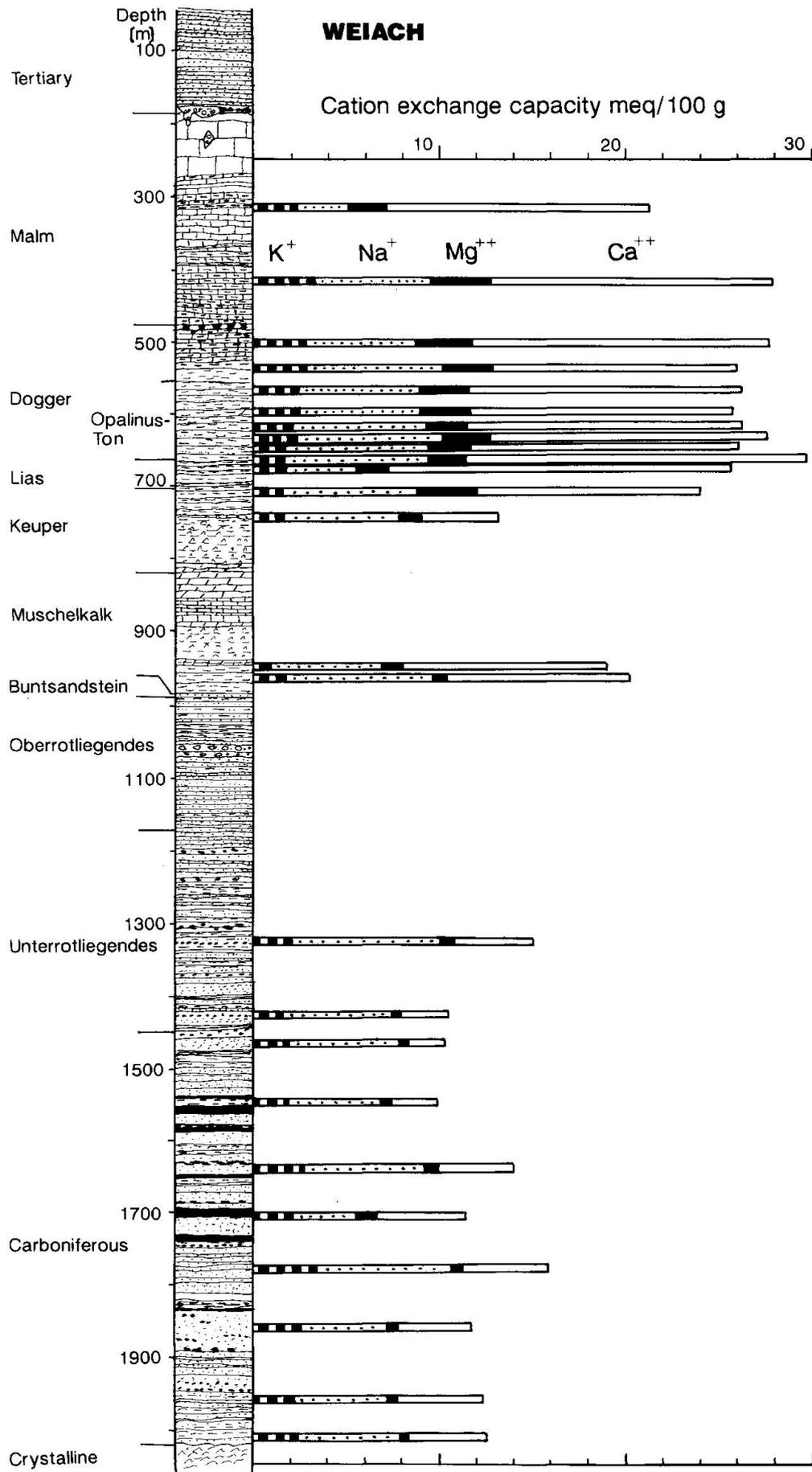
Ratios of exchangeable K^+ , Na^+ , Ca^{2+} and Mg^{2+}

Ca^{2+} and Na^+ make up the main part of the exchangeable cations. The amounts of Mg^{2+} generally vary between 5 and 10% and only in evaporitic series like the "Gipskeuper" and the "Sulfatschichten" may they reach 30% of the exchangeable cations. The proportion of K^+ is usually 10 to 20% with a few exceptions of up to 35% of the exchangeable cations in some Rotliegendes sandy siltstones. On the whole, in the deeper boreholes Weiach and Riniken (Fig. 2), the amount of Na^{2+} is higher, whereas Ca^{2+} is lower in the samples at great depths. On a much smaller scale, and probably for a different reason, the Na^+/Ca^+ ratio in the Beznau drillhole increases downwards in the uppermost 50 metres of the "Opalinus-Ton".

In order to find out if the nature of the exchangeable cations is a function of the sample mineralogy, their percentages were compared with the contents of the major minerals calcite, dolomite, illite, albite and potassium feldspar. Only in the Rotliegendes formation a weak positive correlation between K^+ and potassium feldspar and between Ca^{2+} and calcite was detected. In the 22 "Opalinus-Ton" samples, no significant correlation could be observed.

Discussion

The measured total exchange capacities of 10–30 meq / 100 g are common for illite dominated clays (GRIMM, 1953) and define a range that can be used in modelling retardation capacities of most clay rich formations in the subsurface of Northern Switzerland. The relatively lower CEC capacities in the Permo-Carboniferous formations in comparison with the Mesozoic cannot be attributed to lower total clay mineral contents nor to a different clay mineral distribution. However, the values for the specific surface as well as for the external surface (BET measured) are much lower in the Permo-Carboniferous than in the Mesozoic samples. This difference is not a function of today's depth as seen in the profiles of Fig. 2, but is more probably relat-



Figs 2 a, b, c Lithologic profile of sedimentary part of the drillholes with cation exchange capacity and site occupancy.

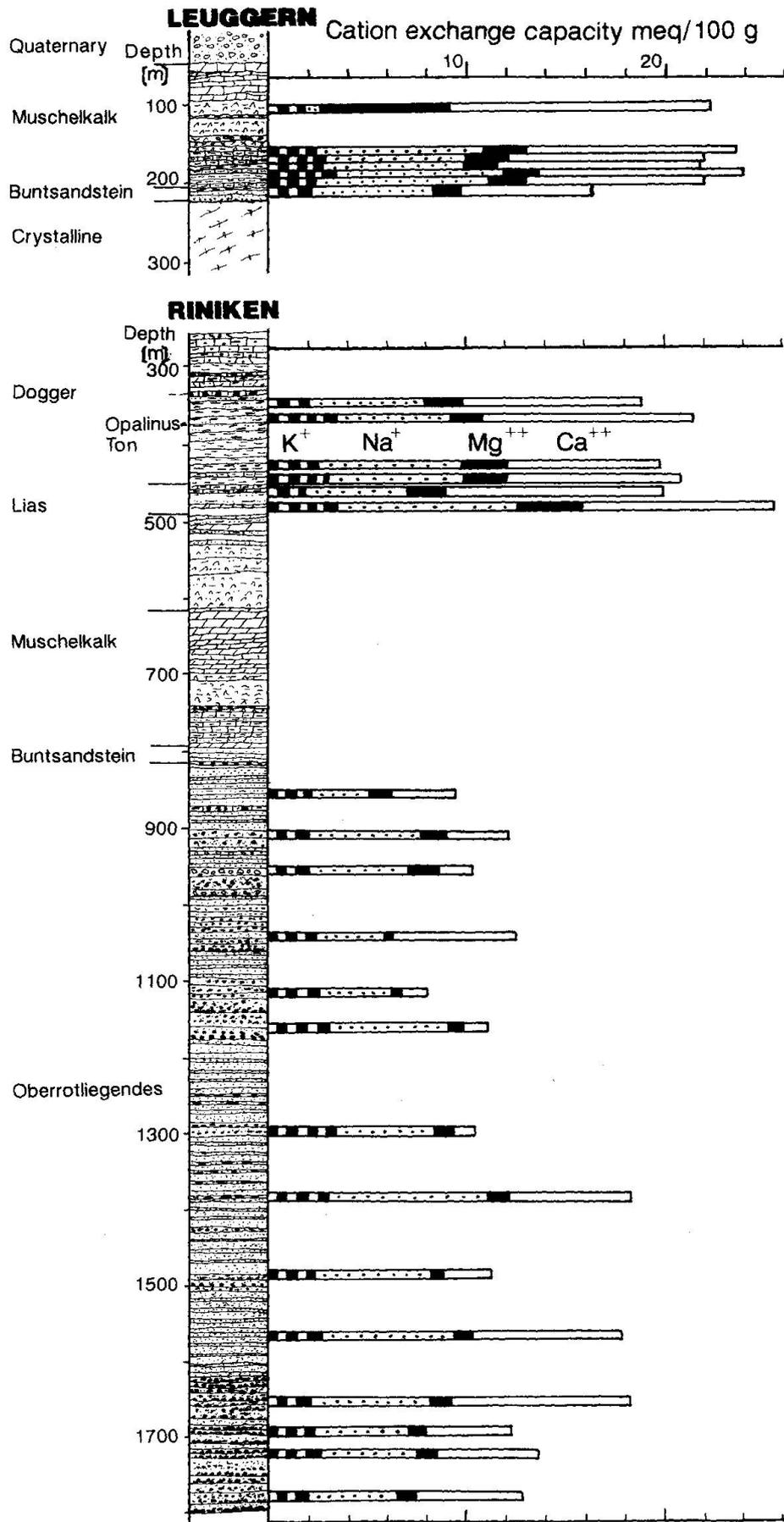


Fig. 2b

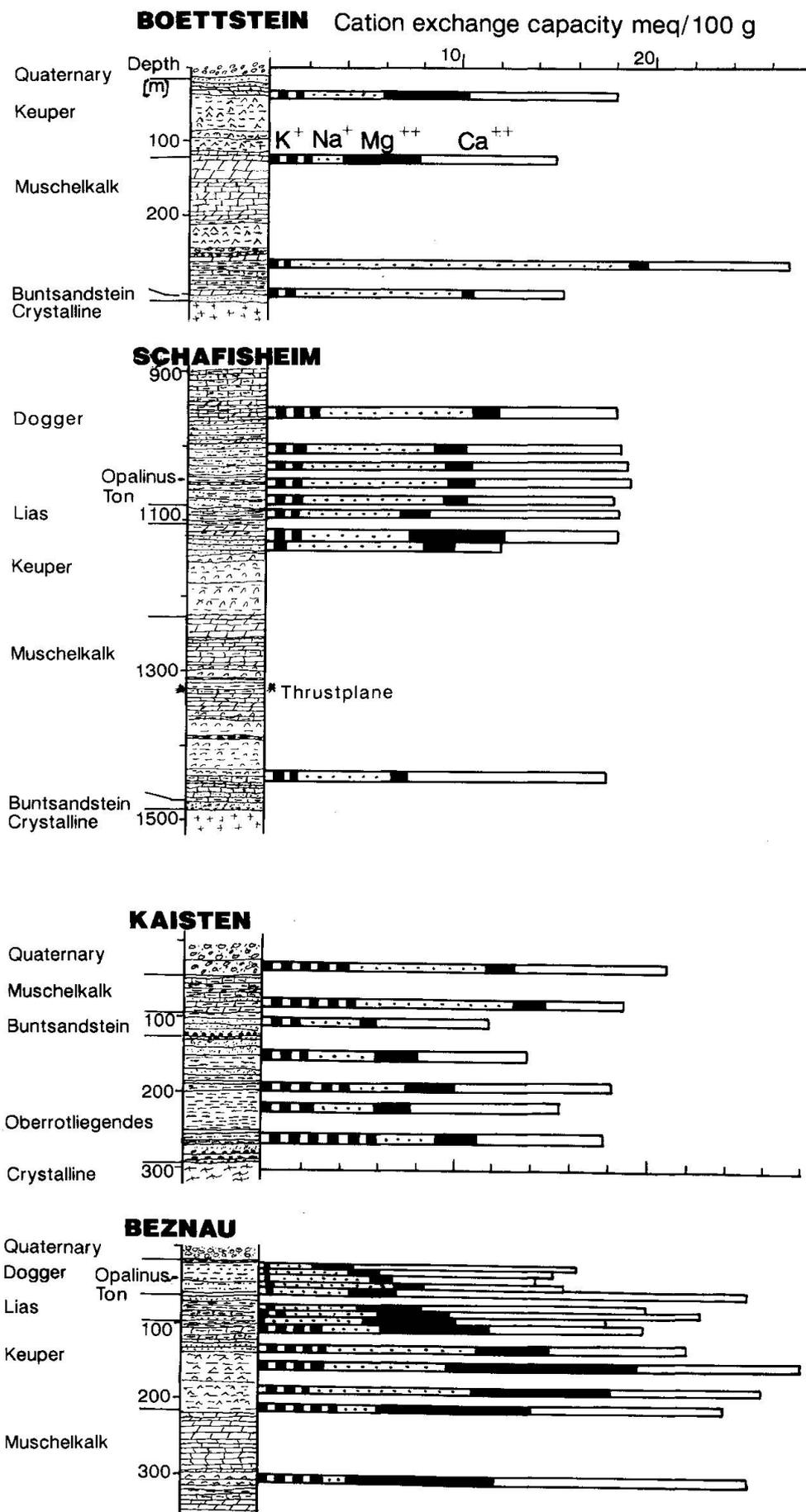


Fig. 2c

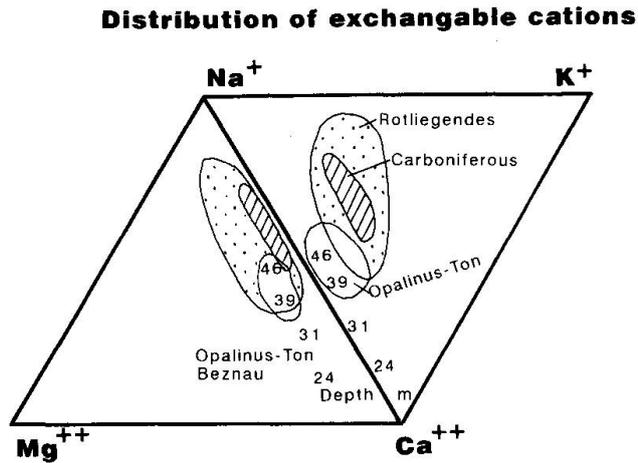
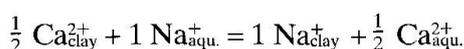


Fig. 3 Distribution of exchangeable cations in sediments from the Carboniferous, Rotliegendes and Opalinus-Ton. Numbers indicate depth of Opalinus-Ton samples from the Beznau drillsite.

ed to stronger diagenesis during the maximum depth of burial in Mesozoic times (MATTER et al., 1988). Most probably, there is also an overprint of the Permo-Carboniferous hydrothermal alteration which mainly took place in the crystalline (PETERS et al., 1987c) but might have also affected the Permo-Carboniferous sediments in the troughs.

The change in ratio of exchangeable Na^+ and Ca^{2+} in the uppermost samples from the "Opalinus-Ton" in the Beznau drillhole can be explained as being the result of either surface weathering before deposition of the Riss tills, or of a subsurface alteration by groundwaters of the overlying tills. As the carbonate content of the uppermost layers has not decreased, which is a typical sign of weathering of the "Opalinus-Ton", the second explanation is favoured. The change of Na^+ - Ca^{2+} occupancy would then be the result of an exchange with the Ca dominated overlying groundwaters in the gravels. With a depth of the affected zone of 20 m and an estimated age of 120,000 y of the tills, a mean velocity of the "exchange front" of 0.2 mm/year can be calculated. This "exchange front" does not indicate downward moving groundwaters.

In order to see whether the cation occupancy observed in the deeper strata could be the result of exchange reactions with deep groundwaters, selectivity coefficients (BOLT, 1982 a, b) were calculated. 8 CEC analysed samples are in the range where deep groundwaters were sampled and analysed (PEARSON et al., 1989). Assuming exchange reactions between the waters and the clay rich samples where, as an example Ca^{2+} on the exchange site of the clay is substituted by the Na^+ in the solution:



BOLT (1982 a, b) defined a selectivity coefficient as

$$K_c = \frac{\left(\frac{\text{Na}_{\text{clay}}^+}{\text{Na}_{\text{clay}}^+ + \text{Ca}_{\text{clay}}^{2+}} \right)^1}{\left(\frac{\text{Ca}_{\text{clay}}^{2+}}{\text{Na}_{\text{clay}}^+ + \text{Ca}_{\text{clay}}^{2+}} \right)^{\frac{1}{2}}} \times \frac{(\text{aCa}_{\text{aqu}}^{2+})^{\frac{1}{2}}}{(\text{aNa}_{\text{aqu}}^+)^1}$$

where

$$\frac{\text{Ca}_{\text{clay}}^{2+}}{\text{Na}_{\text{clay}}^+ + \text{Ca}_{\text{clay}}^{2+}}$$

is the molar ratio of exchangeable Na^+ over the sum of exchangeable Na^+ and Ca^{2+} ; $\text{aCa}_{\text{aqu}}^{2+}$ and $\text{aNa}_{\text{aqu}}^+$ are the activities of Ca^{2+} and Na^+ in solution. In table 2, the K_c -values for different ion pairs are listed.

The considerable variations in partitioning coefficients (CEC/conc. in aqueous phase) for instance Na/Ca varies from 0,05 in WEI 1418 to 2.4 ml/g in LEU 209 are mainly due to big variations in salinity of the groundwaters. On the other hand the selectivity coefficients (K_c) are less affected by total concentrations and show minor variations, well within the range given by BRUGGENWERT and KAMPHORST (1982) for the common clay minerals and soils. Particularly the selectivity coefficients for the pair Na-Ca and K-Na are very constant, despite the wide range in total ionic concentrations of the groundwaters. This behaviour confirms the assumption of hydrogeologists that water-rock interaction by ion exchange could be an important process in deep groundwater regimes for major cations.

Another problem is the original occupancy of the exchangeable sites. In the highly impermeable marine clays and marls like the Opalinus-Ton the occupancies of Ca, Na, K and Mg do not vary much and should mirror the situation during sedimentation and/or diagenesis. The selectivity coefficients of the "Opalinus-Ton" calculated with seawater or extracted pore fluid respectively (GAUTSCHI et al., 1991), are, compared to the values obtained from the aquifers in contact with marine formations, similar with respect to Na/Ca and K/Na (see Tab. 1). The K_c K/Ca of "Opalinus-Ton" with seawater is extremely low, which is probably an indication that the K/Na ratio of the pore waters is buffered by clay minerals. This process would decrease the K-content of trapped seawater. Compared to the other formations, the K_c Mg/Ca values for "Opalinus-Ton" are rather low. This is mainly due to the relatively high Mg-content of seawater and the extracted pore water as compared to the other groundwaters.

In the non-marine strata the exchangeable sites must have been filled during diagenesis when pore

Tab. 1 Selectivity coefficients of samples that might be in exchange with deep groundwaters.

	Leuggern Buntsand- stein	Kaisten Buntsand- stein	Kaisten Rotlie- liegendes	Riniken Rotlie- liegendes	Riniken Rotlie- liegendes	Weiach Rotlie- liegendes	Beznau Muschel- kalk	Beznau Gansinger Dolomit Ob.bunt. Mergel	Beznau Gansinger Dolomit Unt.bunt. Mergel	Opalinus/ Ton Mean Squeezed Water	Opalinus/ Ton Mean Seawater
Groundwater Sample	208-227	97-129.9	276-292	958-972	977-1010	1401-1415	220-304	104-109	104-109	Mt. Terry	Seawater
Na mol/l	0.0265	0.0777	0.0197	0.262	0.262	1.179	0.049	0.178	0.178	0.217	0.456
Ca mol/l	0.000434	0.0111	0.000809	0.0104	0.0117	0.228	0.0159	0.0127	0.0127	0.0197	0.01
K mol/l	0.000233	0.000965	0.000323	0.00364	0.00397	0.02	0.0022	0.00123	0.00123	0.0024	0.0097
Mg mol/l	0.000153	0.00232	0.000101	0.00332	0.00334	0.0425	0.00844	0.0088	0.0088	0.030	0.0562
Rock sample	209.13	101.32	262	948	1036.40	1418	303.1	105	110.4	Mean Op.	Mean Op.
Na meq/100g	6.31	3.21	3.18	5.10	3.50	5.85	1.22	4.8	10.11	32%	32%
Ca meq/100g	6.36	5.96	6.59	1.85	6.40	2.60	13.16	6.3	4.62	50%	50%
K meq/100g	2.21	1.83	5.98	2.00	2.25	1.55	3.36	0.5	3.39	10%	10%
Mg meq/100g	1.48	0.74	2.16	1.30	0.40	0.45	7.66	4.2	2.21	8%	8%
Kc Na/Ca	0.55	0.59	0.52	0.56	0.18	0.51	0.42	0.36	0.76	0.32	0.11
Kc K/Ca	34	46	52	21	8.6	17	13	6.6	50	10.6	1.87
Kc Mg/Ca	38	16.3	2.3	12	2.0	0.87	1.1	0.92	0.69	0.32	0.17
Kc K/Na	38	45	108	33	67	26	42	14	48	28	15

solutions were more concentrated than surface waters. In these cases the ions of the pore solutions are supplied by dissolution of rockforming minerals, i.e. sodium and calcium from plagioclase, potassium from K-feldspar, Mg from biotite and, more seldom, from lacustrine evaporite minerals. This would also explain the tendency for the higher Ca²⁺ occupancies in the Mesozoic strata compared to the Palaeozoic where the Na⁺ and K⁺ occupancies are, in general, more elevated.

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EXCHANGEABLE CATIONS IN MESOZOIC AND PERMO-CARBONIFEROUS SEDIMENTS

Tab. 2 Analytical data of samples from NAGRA drillholer.

DEPTH (m)	Geologic formation	Lithology	Exchangeable cations meq/100g					Surface area		Calcite	Dolomite/Asterite/Siderite	Quartz	K-feldspar	Albite	Illite	Kaolinite	Smectite	Chlorite	Illite/Smectite
			K+	Na+	Mg++	Ca++	Total	external m ² /g	internal m ² /g										
RINIKEN																			
348.57	Opalinus-Ton	claystone	2.00	5.85	1.90	9.15	18.90	27	184	8	-	23	1	1	32	15	-	12	8
360.39	Opalinus-Ton	sandy claystone	2.70	6.10	2.05	10.65	21.50	30	316	7	2	19	2	2	29	19	-	12	8
427.38	Opalinus-Ton	claystone	2.55	7.35	2.25	7.50	19.65	36	148	5	5	17	1	1	34	17	-	10	10
448.58	Opalinus-Ton	claystone	2.75	7.05	2.05	8.35	20.20	34	218	11	2	16	1	1	33	15	-	12	9
448.35	Posidonienschiefer	marl	1.75	5.50	1.75	10.80	19.80	25	50	23	2	9	1	2	39	8	-	11	4
484.24	Insektenmergel	claystone, black	3.40	9.10	3.35	9.85	25.70	47	126	7	1	9	-	-	48	7	-	14	14
848.43A	Rotliegendes	siltstone	2.10	2.85	1.20	3.35	9.50	22	0	-	1	19	5	3	54	3	-	3	12
905.65	Rotliegendes	siltstone	2.00	5.75	1.20	3.25	12.20	18	18	-	5	30	27	10	23	-	-	2	3
947.96	Rotliegendes	siltstone	2.00	5.10	1.30	1.85	10.25	17	22	-	9	19	17	7	37	-	-	3	8
1036.40	Rotliegendes	silty sandstone	2.25	3.50	0.40	6.40	12.55	8	46	6	-	38	25	5	23	-	-	-	3
1112.19	Rotliegendes	sandstone	2.45	3.70	0.50	1.45	18.00	9	28	1	-	39	25	7	24	-	-	-	4
1162.65	Rotliegendes	siltstone, red	3.60	6.10	0.70	1.25	11.05	17	12	1	-	24	8	4	53	-	-	-	10
1296.46	Rotliegendes	siltstone	3.35	5.15	0.80	1.25	10.55	13	36	-	-	-	-	-	-	-	-	-	8
1376.70	Rotliegendes	siltstone with nodules	3.10	8.15	1.00	6.00	18.25	20	48	6	-	18	2	2	52	-	-	-	12
1486.88	Rotliegendes	siltstone w. coarse sand	2.25	6.00	0.65	2.45	11.35	11	38	-	-	20	11	13	38	-	-	-	8
1558.83	Rotliegendes	siltstone with caliche	2.70	6.80	0.90	7.45	17.85	15	38	13	1	17	1	7	40	-	-	-	13
1638.75	Rotliegendes	caliche	2.05	6.10	1.05	9.15	18.35	18	84	7	-	15	1	7	47	-	-	-	16
1686.11	Rotliegendes	siltstone	2.20	4.90	0.80	4.45	12.35	13	72	-	-	21	6	10	38	-	-	-	11
1716.59	Rotliegendes	siltstone	2.60	5.95	1.00	4.10	13.65	15	66	1	-	18	3	11	50	-	-	-	10
1769.77	Rotliegendes	siltstone with caliche	1.90	4.55	0.90	5.45	12.80	13	78	2	-	21	6	14	36	-	-	-	13
KAISTEN																			
58.36	Wellenmergel	marl	4.46	7.23	1.44	7.85	20.98	25	46	16	3	18	1	3	43	-	-	-	9
84.00	Wellendolomit	claystone	4.74	8.35	1.69	4.04	18.82	37	44	-	8	15	1	6	55	-	-	-	7
101.32	Buntsandstein	claystone	1.83	3.24	0.74	5.96	11.74	33	50	2	2	32	4	-	47	2	-	-	10
152.31	Oberrotliegendes	siltstone	2.28	3.63	2.22	5.75	13.88	40	22	1	7	14	2	1	65	2	-	-	6
194.08	Oberrotliegendes	clay/siltstone	4.51	3.00	2.63	8.12	18.26	38	52	5	-	20	7	3	56	2	-	-	5
222.26	Oberrotliegendes	siltstone	2.59	3.21	1.93	7.88	15.61	41	48	2	-	17	2	1	64	-	-	-	10
262.44	Oberrotliegendes	siltstone	5.98	3.18	2.16	6.59	17.91	41	76	-	-	18	2	-	69	1	-	-	7
BOETTSTEIN																			
35.00	Schilfsandstein	claystone	1.7	4.3	4.4	7.8	18.2	90	0	-	-	2	-	-	90	-	-	-	-
120.65	"Lettenkohle"	sandy claystone	2.2	1.7	4.0	7.2	15.1	24	124	1.5	1.5	10	19	-	56	8	-	-	-
264.35	Orbicularis-Mergel	marl	1.1	17.8	0.9	5.7	27.3	15	184	28	5	10	-	3	44	-	-	-	5
297.90	Wellendolomit	marl	1.4	8.8	0.5	4.7	15.4	33	168	7	-	13	3	3	64	-	-	-	9
LEUGGERN																			
100.60	Oberer Sulfatschichten	dolomitic marl	1.81	0.87	6.52	13.10	22.30	22	348	20	22	10	4	4	11	-	26	3	-
162.59	Orbicularis-Mergel	marl black	2.54	8.51	2.20	10.48	23.73	21	226	23	5	12	-	3	41	-	-	-	11
169.44	Wellenmergel	marl	2.88	7.07	2.18	9.85	21.99	31	204	13	2	15	3	5	31	-	-	-	5
174.11	Wellenmergel	marl	2.76	7.18	1.56	10.35	21.85	28	130	23	-	13	2	4	42	-	-	-	6
187.82	Wellenmergel	marl	3.43	8.37	1.77	10.35	23.92	33	204	10	-	14	-	6	51	-	-	-	5
198.62	Wellendolomit	marl	2.36	8.86	1.79	8.98	21.99	34	68	10	2	12	4	15	50	-	-	-	8
209.13	Buntsandstein	medium fine-gr. sandstone	2.21	6.31	1.48	6.36	16.36	36	28	2	3	31	10	2	43	-	-	-	6
WEIACH																			
305.30	Schwarzach-Schichten	marl	2.30	2.80	2.15	14.10	21.35	21	108	68	3	4	-	-	16	2	-	-	5
410.50	Effinger-Schichten	marl	3.35	6.20	3.15	15.20	27.90	31	148	43	3	9	-	-	29	5	-	-	9
500.50	Württembergica-Schichten	sandy marl	2.85	5.85	2.95	16.10	27.75	30	204	25	1	21	4	2	23	10	-	-	8
535.50	Parkinsons-Schichten	sandy marl	2.80	7.25	2.60	13.35	26.00	37	200	10	-	22	1	-	34	13	-	-	10
565.30	Opalinus-Ton	sandy claystone	2.45	6.50	2.50	14.75	26.20	30	194	13	-	22	2	1	25	12	-	-	5
595.10	Opalinus-Ton	sandy claystone	2.50	6.40	2.60	14.30	25.80	36	150	9	6	16	3	1	27	21	-	-	6
610.50	Opalinus-Ton	sandy claystone	2.10	7.20	2.15	14.85	26.30	32	156	7	5	14	-	-	29	26	-	-	7
630.25	Opalinus-Ton	sandy claystone	2.30	7.90	2.50	14.85	27.55	34	208	5	5	14	-	-	30	26	-	-	11
645.40	Opalinus-Ton	sandy claystone	1.70	7.75	2.20	14.45	26.10	32	220	6	7	11	-	-	32	23	-	-	8
660.65	Opalinus-Ton	sandy claystone	1.75	7.70	2.00	18.25	29.70	33	200	7	3	13	-	1	32	23	-	-	9
676.91	Posidonen-Schiefer	marl, laminated	1.70	3.80	1.70	18.55	25.75	15	190	37	1	5	-	-	28	8	-	-	8
705.25	Knollenmergel	calclitic, dolomitic marl	1.55	7.20	3.05	12.30	24.10	40	104	24	28	4	-	-	22	-	-	-	9
735.50	Schilfsandstein	sandy claystone	1.60	6.15	1.25	4.20	13.20	29	22	-	-	19	10	7	49	-	-	-	15
945.50	Orbicularis Mergel	marl	1.00	6.00	1.00	11.00	19.00	13	134	59	11	3	-	-	20	-	-	-	6
959.95	Wellenmergel	sandy marl	1.75	8.00	0.65	9.85	20.25	22	84	27	2	9	2	3	47	-	-	-	10
1323.35	Rotliegendes	claystone	2.00	8.00	0.70	4.40	15.10	25	62	-	-	8	1	-	29	29	-	-	16
1418.50	Rotliegendes	clay-siltstone, bitum.	1.55	5.85	0.45	2.60	10.45	11	38	1	12	6	5	26	26	-	-	-	9
1466.90	Karbon	siltstone	1.90	5.85	0.45	2.05	10.25	13	22	3	-	8	2	-	25	50	-	-	6
1540.80	Karbon	siltstone, carbonif.	1.85	5.10	0.50	2.45	9.90	8	58	-	3	7	-	-	24	52	-	-	3
1630.75	Karbon	claystone	2.70	6.30	0.70	4.25	13.95	13	48	-	6	5	2	-	29	38	-	-	7
1688.45	Karbon	siltstone	2.10	3.35	1.05	4.80	11.30	4	22	-	43	8	4	-	21	14	-	-	11
1765.70	Karbon	sandy claystone	3.30	7.20	0.50	4.80	15.80	13	30	-	1	15	5	1	46	8	-	-	10
1840.55	Karbon	sandy claystone	2.60	4.50	0.55	4.05	11.70	7	10	-	1	42	5	1	31	7	-	-	4
1940.60	Karbon	fine grained sandstone	2.25	5.00	0.45	4.55	12.25	7	20	-	-	37	10	1	32	6	-	-	10
1992.40	Karbon	siltstone	2.30	5.50	0.40	4.30	12.50	9	40	-	-	22	1	1	47	11	-	-	9
BEZNAU																			
24.00-31.04	Opalinus-Ton	silty claystone	0.50	2.30	2.10	11.80	16.70	24.8	137	12	1	28	2	3	20	23	-	-	7
31.04-38.61	Opalinus-Ton	silty claystone	0.50	4.20	1.70	11.00	17.40	25.7	125	10	1	24	1	2	25	23	-	-	4
38.61-46.15	Opalinus-Ton	silty claystone	0.50	5.40	1.20	7.40	11.50	31.4	135	10	1	19	-	1	26	22	-	-	10
46.15-53.65	Opalinus-Ton	silty claystone	0.50	5.70	1.10	5.70	13.00	34.9	121	9	-	16	-	1	32	24	-	-	9
53.65-60.																			