ments k_d in the cluster according to $(k_d + m_d)!/k_d!m_d!$. If this is the case, the addition of an element to a group of elements implies self-structurizing properties if the following holds for the interaction between the fundamental elements:

$$\Pi \frac{(k_c + m_c)!}{k_c!m_c!} > \Pi \frac{(k_d + m_d)!}{k_d!m_d!}. \qquad (2)$$

In summary, elementary reductionism comprising dynamic interaction of different elements may cause self-organization or dissolution of existing organization, e.g. by environmental impact, in several real systems found in nature, and may also be relevant to phenomena in human society including economics and politics and to mental concept-forming processes including pattern recognition and the evolution of science. To explore the exact conditions for self-organization in these systems in terms of increased cognitive stability would be a challenging scientific task.

1 Acknowledgment. This work was supported by travel grants from the University of Uppsala and the Sweden-Japan Foundation for Research and Development 1982–1983.

2 Bertalanffy, L. v., General Systems Theory. Braziller, New York 1968.
3 Bonner, J.T., Chemical signals of social amoebae. Sci. Am. *248* (April 1983) 106–112.
4 Clare, C.W.F., Chemical Machines, Maxwell's Demon and Living Organisms. J. theor. Biol. *30* (1971) 1–34.
5 Dörner, D., Problemlösen als Informationsverarbeitung. Kohlhammer-Verlag, Stuttgart 1980.
6 Gabbiani, G., and Gabbiani, F., Organization of actin cytoskeleton in normal and regenerating arterial endothelial cells. Proc. natn. Acad. Sci. USA *80* (1983) 2361–2364.
7 Goldsmith, E., Thermodynamics or Ecodynamics? Ecologist *11* (1981) 178.
8 Haken, H., Synergetics. Springer, New York 1978.
9 Herman, I.M., Crisona, N.J., and Pollard, T.D., Relation between cell activity and the distribution of cytoplasmic actin and myosin. J. Cell Biol. *90* (1982) 84–91.
10 Hoyle, G., Muscles and Their Neural Control. Wiley-Interscience, John Wiley & Sons, New York, Chichester, Brisbane, Toronto, Singapore 1983.
11 Malik, F., and Probst, G.J.B., Evolutionäres Management. Die Unternehmung, Schweiz. Z. Betriebswirtsch. *2* (1981) 121.
12 Margenau, H., The Nature of Physical Reality. McGraw Hill, New York 1950.
13 Margenau, H., The Miracle of Existence. Ox Bow Press, Cambridge 1984.
14 Mayer, E., Biokybernetisches Controlling als Unternehmensphilosophie, in: Controlling Berater, Gruppe 3 (1983) 21.
15 McClintock, P., Low-temperature physics – Knudsen effect in liquid helium. Nature *306* (1983) 422–423.
16 Naveh, Z., and Liebermann, A.S., Landscape Ecology – Theory and Application. Springer, New York 1984.
17 Pollack, R., Oxborn, M., and Weber, K., Patterns of organization of actin and myosin in normal and transformed cultured cells. Proc. natn. Acad. Sci. USA *72* (1975) 994–998.
18 Prigogine, I., and Lefever, R., Stability and selforganization in open systems, in: Membranes, Dissipative Structures and Evolution. Adv. Chem. Phys., vol. 29, pp. 1–28. Eds G. Nicolis and R. Lefever. John Wiley & Sons, New York, London, Sydney, Toronto 1975.
19 Probst, G.J.B., Kybernetische Gesetzeshypothesen als Basis für Gestaltungs- und Lenkungsregeln im Management. Paul-Haupt-Verlag, Bern 1981.
20 Schrödinger, E., Statistical Thermodynamics. University Press, Cambridge 1952.
21 Schrödinger, E., What is Life? The Physical Aspects of the Living Cell. University Press, Cambridge and The McMillian Company, New York 1946.
22 Vester, F., and v. Hesler, The Sensitivity Model. Umlandrerband, Frankfurt 1980.
23 Vester, F., Urban Systems in Crisis. Deutsche Verlagsanstalt, Stuttgart 1976.
24 Welch, G.R., On the role of organized multienzyme systems in cellular metabolism: A general synthesis. Prog. Biophys. molec. Biol. *32* (1977) 103–191.

0014-4754/85/060713-07$1.50 + 0.20/0

Origin of life: Oceanic genesis, panspermia or Darwin's 'warm little pond'?

D.H. Spaargaren

Netherlands Institute for Sea Research, P.O.B. 59, NL-1790 AB Den Burg, Texel (The Netherlands), 26 March 1984

Summary. The average abundances of chemical elements in crustal rocks, river water and biological material are compared with the Suess-Urey[22] abundances distribution (representing the most primitive distribution of the elements on earth). Crustal rocks can be considered still to have the most primitive elemental composition, whereas seawater shows the largest deviations form the cosmic Suess-Urey element distribution. Biological material ranks, in the series considered, between river water and seawater, still showing primitive characteristics. The relative elemental composition of biological material resembles river water more strongly than contemporary seawater: the ratios between the quantitatively most important elements (C, N and P) in living matter are almost equal to those in river water; also the concentration factors for the other elements are less variable than those in seawater. The latter appear to be inversely related to their concentrations in seawater: in living organisms, the elements having lower concentrations in seawater are concentrated more strongly. This differential concentration cannot be obtained by evaporation. The stability of the composition of biological material (evident by comparing species of widely diverging evolutionary development) and its similarity to the Suess-Urey distribution suggest an oceanic genesis early in the earth's history, before the oceans reached their present compositions.

Key words. Origin of life; Suess-Urey abundance distribution; elemental composition of biological matter; river water composition; seawater composition; oceanic genesis; river water, evaporated.

1) Introduction

Since Darwinian evolution replaced the concept of spontaneous generation, already rejected earlier, a marine origin of life has been commonly accepted. In 1973, however, Crick and Orgel[5] raised some doubts about the idea of oceanic genesis. They pointed out that there are clear discrepancies between the concentrations of certain elements in living organisms and their abundance on earth. For instance molybdenum, an essential element that plays an important role in several enzymatic reactions, is strongly accumulated in living organisms. It seemed unlikely that early organisms on earth would make themselves dependent on this extremely rare element for their existence. The universality of the genetic code suggests that life developed from one source. Hence, the general utilization of a rare element might reflect a greater abundance of that element in the ancestral environment. They forwarded the hypothesis that life on earth was seeded from some other part of the galaxy; the earth supplied the nutrients for further development. The seeding may have been inadvertent, as in the concept of panspermia, or deliberately done by intelligent beings, as in the concept of directed panspermia. The age of our galaxy, compared to the rates of evolutionary and technological development, perhaps does not offer too serious restrictions for directed panspermia.

Banin and Navrot[1] opposed the idea of a possible extraterrestrial origin of life. Examining the enrichments (compared to the main concentrations in the earth) of various elements in bacteria, fungi, plants, land animals and seawater as a function of their ionic potential (the ratio of the positive charge and the radius of an ion) similar patterns appeared. The elements Mo, Ni and Cr – mentioned in support of the panspermia hypothesis – did not take a special position in this pattern. Elements of moderate ionic potential generally form insoluble hydroxides and therefore would be deficient in seawater. Organisms originating from this environment are likewise deficient in these elements.

It is, however, questionable whether the qualitative comparisons as given by Banin and Navrot[1] do prove an oceanic genesis. Firstly, as also noted by Gualtieri[8], the distributions of elemental abundances in living material and seawater are strongly correlated. Hence, plotting the enrichment factors against any element property (including ionic potential) would yield similar patterns for living matter and seawater. Secondly, the element distribution in living material should also have been compared with element abundances in other geochemical compartments and to the relative cosmic abundances to conclude whether an oceanic genesis is most probable, and to see whether an extraterrestrial origin must indeed be excluded. Independent of the question whether the panspermia hypotheses are right or wrong, they cannot be opposed with arguments which are themselves inconclusive.

In this paper the relative abundances of chemical elements in living organisms, seawater, river water and crustal rock are compared to the so-called Suess-Urey[22] abundance distribution (the most reasonable assumption for the relative cosmic abundances of various atomic species).

2) Data on the average elemental composition of living matter, seawater, river water, crustal rock and the relative cosmic abundances of the elements

The elemental composition of living matter is fairly well known for a limited number (circa 30) of elements. For various species of finned fish, molluscs and crustaceans Sidwell et al.[14, 15] compiled data on the elemental concentrations of six macroelements (Na, K, Ca, P, Cl, Mg) and 19 microelements (Cu, Fe, Zn, I, Mn, Hg, Pb, As, F, Ag, Cd, Co, Cr, V, Al, Sn, Ni, Ba, Mo). Their compilation presents the internal concentrations of macroelements in 161 different animal species, as reported in 128 publications as well as the microelement concentrations in 167 species as given in 224 publications. Also the classic work of Vinogradov[23] compiles numerous data from nearly 1200 publications. Recent publications (e.g. Eisler[7]) give additional data, obtained with more advanced techniques, sometimes yielding lower values.

In small animals biological concentrations are usually measured in whole animal homogenates; in larger animals, where supporting tissue (shells, exoskeleton, bones) are relatively more important, the data refer to the 'edible portion' which comprises the living material. The concentration values, frequently expressed in different

Table 1. Abundance of chemical elements in various organisms (finfish, molluscs, crustaceans, marine algae)

Atomic number	Element	log molal concentration		Abundance relative to carbon = 10^6	Reference
		$\bar{x}$	± SD (N)		
1	H	1.81	0.28 (80)	8.32 E 6	1
5	B	– 2.69		2.63 E 2	3
6	C	0.89	0.09 (13)	1.00 E 6	1
7	N	– 0.05	2.20 (93)	1.15 E 5	1
8	O	1.63	0.05 (68)	5.50 E 6	1
9	F	– 3.99	0.37 (14)	1.32 E 1	2
11	Na	– 1.43	0.21 (109)	4.79 E 3	2
12	Mg	– 1.85	0.27 (79)	1.82 E 3	2
13	Al	– 3.05	0.50 (29)	1.15 E 2	2
14	Si	– 1.88	0.66 (18)	1.70 E 3	1
15	P	– 1.17	0.24 (140)	8.71 E 3	2
16	S	– 1.59	0.55 (18)	3.31 E 3	1
17	Cl	– 1.31	0.34 (72)	6.31 E 3	2
19	K	– 1.13	0.11 (122)	9.55 E 3	2
20	Ca	– 1.94	0.35 (139)	1.48 E 3	2
22	Ti	– 4.50		4.07	3
23	V	– 5.05	0.55 (22)	1.15	2
24	Cr	– 5.46	0.30 (19)	4.47 E–1	2
25	Mn	– 4.59	1.08 (69)	3.31	2
26	Fe	– 3.41	0.58 (133)	5.01 E 1	2
27	Co	– 5.04	0.59 (24)	1.17	2
28	Ni	– 5.53	0.36 (14)	3.80 E–1	2
29	Cu	– 4.11	0.57 (84)	1.00 E 1	2
30	Zn	– 3.45	1.53 (72)	4.57 E 1	2
31	Ga	– 6.77		2.19 E–2	3
33	As	– 4.24	1.14 (58)	7.41	2
34	Se	– 5.17	0.18 (15)	8.71 E–1	2
35	Br	– 2.87	0.21 (8)	1.74 E 2	1
38	Sr	– 2.54		3.72 E 2	3
42	Mo	– 5.24	0.43 (9)	7.41 E–1	2
48	Cd	– 5.62	0.15 (41)	3.09 E–1	2
50	Sn	– 4.82	0.40 (23)	1.95	2
53	I	– 5.00	1.56 (67)	1.29	2
56	Ba	– 5.84	– (2)	1.86 E–1	2
80	Hg	– 5.60	0.56 (87)	3.24 E–1	2
82	Pb	– 5.02	0.87 (34)	1.23	2
88	Ra	– 13.58	0.22 (4)	3.39 E–9	1

* Data derived from 1) Vinogradov[23], ch. XVI; 2) Sidwell et al.[13, 14] and 3) Yamamoto et al.[25,26].

units (g/100 g, ppt or ppm, etc.), can be converted to their molal concentrations as presented in table 1. The standard deviations, also given in table 1, comprise both the intra- and interspecific variation which – as might be expected – may be quite considerable. However, usually the variation does not exceed 1 decade of molal concentration. The elemental composition of living matter can roughly be characterized by stating that H, C, O and N are present in molal concentrations, the elements Na, Mg, Si, P, Cl, S, K and Ca in millimolal concentrations, most metals in micromolal concentrations and the remaining elements in still lower, nanomolal concentrations. The order of magnitude remains practically the same, even in widely divergent taxonomical groups, like bacteria; fungi; plants; land animals[1,8]; marine algae[25,26] and marine crustaceans, finned fish and molluscs[18-20]. This stability in elemental composition of living matter does not imply that the various elements occur in similar molcular forms.

Table 2. Average abundance of chemical elements in seawater, river water and crustal rocks. Data derived from values given by Brewer[2] and Whitfield[24]

Atomic number	Element	Abundance relative to carbon (= 10^6) Seawater	River water	Crustal rock
1	H	2.40 E 10	9.55 E 11	–
2	He	7.41 E–1	–	–
3	Li	1.12 E 4	4.07 E 3	1.74 E 5
4	Be	2.75 E–1	–	1.86 E 4
5	B	1.78 E 5	8.71 E 3	5.62 E 4
6	C	1.00 E 6	1.00 E 6	1.00 E 6
7	N	4.68 E 6	1.78 E 5	6.51 E 4
8	O	2.40 E 10	9.55 E 11	–
9	F	2.95 E 4	5.01 E 4	2.00 E 6
10	Ne	3.02	2.63 E 6	6.17 E 7
11	Na	2.04 E 8	1.58 E 6	5.75 E 7
12	Mg	2.34 E.7	1.41 E 5	1.86 E 8
13	Al	3.24 E 1	2.19 E 6	6.03 E 8
14	Si	3.09 E 4	–	–
15	P	8.71 E 2	6.17 E 3	2.04 E 6
16	S	1.23 E 7	1.15 E 6	4.79 E 5
17	Cl	2.40 E 8	4.79 E 5	2.19 E 5
18	Ar	4.79 E 1	–	–
19	K	4.47 E 6	5.62 E 5	3.24 E 7
20	Ca	4.47 E 6	3.55 E 6	6.31 E 7
21	Sc	5.62 E–3	8.51 E–1	2.95 E 4
22	Ti	8.71	6.03 E 2	7.24 E 6
23	V	2.19 E 1	1.66 E 2	1.58 E 5
24	Cr	2.51	1.82 E 2	1.15 E 5
25	Mn	1.58	1.20 E 3	1.05 E 6
26	Fe	1.51 E 1	1.15 E 5	6.03 E 7
27	Co	3.47 E–1	1.62 E 1	2.57 E 4
28	Ni	1.23 E 1	4.90 E 1	7.76 E 4
29	Cu	3.47	1.05 E 3	5.25 E 4
30	Zn	3.31 E 1	2.88 E 3	6.46 E 4
31	Ga	1.86 E–1	–	–
32	Ge	3.02 E–1	–	–
33	As	2.19 E 1	2.51 E 2	1.48 E 3
34	Se	1.10	2.40 E 1	3.80 E 1
35	Br	3.63 E 5	2.40 E 4	1.91 E 3
36	Kr	1.05	–	–
37	Rb	6.17 E 2	1.12 E 2	6.31 E 4
38	Sr	3.98 E 4	7.59 E 3	2.57 E 5
39	Y	6.61 E–3	–	–
41	Nb	4.37 E–2	–	–
42	Mo	4.37 E 1	5.89 E 1	9.55 E 2
47	Ag	1.74 E–1	2.63 E 1	3.98 E 1
48	Cd	4.37 E–1	–	1.07 E 2
49	In	3.47 E–4	3.23 E 0	1.02 E 3
50	Sn	3.63 E–2		
51	Sb	8.71 E–1	1.55 E 2	9.77 E 1
53	I	2.19 E 2	5.25 E 2	2.40 E 2
54	Xe	1.66 E–1	–	–
55	Cs	1.32	–	–
56	Ba	6.61 E 1	–	–
57	La	8.71 E–3	–	–
58	Ce	4.37 E–2	–	–
74	W	2.19 E–1	–	–
79	Au	8–71 E–3	–	–
80	Hg	6.61 E–2	3.31 E 0	2.40 E 1
81	Tl	2.19 E–2	–	–
82	Pb	8.71 E–2	1.38 E 2	3.63 E 3
83	Bi	4.37 E–2	–	–
86	Rn	1.17 E–12	–	–
88	Ra	1.32 E–7	–	–
90	Th	1.74 E–2	–	–
91	Pa	8.71 E–8		
92	U	6.17	–	–

Dividing the average molal concentrations of various elements by the average molal concentration of carbon and, for convenience multiplying by 10^6, yields the abundances (the logarithms indicated by H) of the elements in living material in relation to carbon (table 1). Carbon was chosen as reference element because of its particular significance in organic molecules.

Data on the average composition of seawater were obtained from a table given by Brewer[2]. Data on the elemental composition of average river waters and crustal

Table 3. Cosmic abundance of the elements. Suess-Urey[22] distribution transformed to elemental abundance relative to carbon

Atomic number	Element	Abundance relative to carbon (= 10^6)	Atomic number	Element	Abundance relative to carbon (= 10^6)
1	H	1.14 E 10	42	Mo	6.91 E–1
2	He	8.80 E 8	44	Ru	4.26 E–1
3	Li	2.86 E 1	45	Rh	6.11 E–2
4	Be	5.71	46	Pd	1.93 E–1
5	B	6.86	47	Ag	7.43 E–2
6	C	1.00 E 6	48	Cd	2.54 E–1
7	N	1.89 E 6	49	In	3.14 E–2
8	O	6.14 E 6	50	Sn	3.80 E–1
9	F	4.57 E 2	51	Sb	7.03 E–2
10	Ne	2.46 E 6	52	Te	1.33
11	Na	1.25 E 4	53	I	2.29 E–1
12	Mg	2.61 E 5	54	Xe	1.14
13	Al	2.71 E 4	55	Cs	1.30 E–1
14	Si	2.86 E 5	56	Ba	1.05
15	P	2.86 E 3	57	La	5.71 E–1
16	S	1.07 E 5	58	Ce	6.46 E–1
17	Cl	2.53 E 3	59	Pr	1.14 E–1
18	A	4.29 E 4	60	Nd	4.11 E–1
19	K	9.03 E 2	62	Sm	1.90 E–1
20	Ca	1.40 E 4	63	Eu	5.34 E–2
21	Sc	8.00	64	Gd	1.95 E–1
22	Ti	6.97 E 2	65	Tb	2.73 E–2
23	V	6.29 E 1	66	Dy	1.59 E–1
24	Cr	2.23 E 3	67	Ho	3.37 E–2
25	Mn	1.96 E 3	68	Er	9.03 E–2
26	Fe	1.71 E 5	69	Tm	9.09 E–3
27	Co	5.14 E 2	70	Yb	6.29 E–2
28	Ni	7.83 E 3	71	Lu	1.43 E–2
29	Cu	6.06 E 1	72	Hf	1.25 E–1
30	Zn	1.39 E 2	73	Ta	1.86 E–2
31	Ga	3.26	74	W	1.40 E–1
32	Ge	1.44 E 1	75	Re	3.86 E–2
33	As	1.14	76	Os	2.86 E–1
34	Se	1.93 E 1	77	Ir	2.35 E–1
35	Br	3.83	78	Pt	4.64 E–1
36	Kr	1.47 E 1	79	Au	4.14 E–2
37	Rb	1.86	80	Hg	8.11 E–2
38	Sr	5.40	81	Tl	3.09 E–2
39	Y	2.54	82	Pb	1.34 E–1
40	Zr	1.56 E 1	83	Bi	4.11 E–2
41	Nb	2.86 E–1			

rocks were obtained from the compilation by Whitfield et al.[24]. Table 2 shows the concentrations values converted to the abundance of the various elements relative to carbon (again arbitrarily taken as 10^6).

Values for the relative cosmic abundances of the elements (table 3) are taken from the paper of Suess and Urey[22], which also gives a detailed explanation of how the values for various elements were derived. Part of the data were derived from spectrometric analysis of the light of the sun and certain stars; partly they were derived from chemical analysis of meteorites. The abundances of the elements in many of the stars are very similar to those of the sun and planetary nebulae. Although the values for the atomic abundances, especially those of the heavier and less abundant elements, are still subject to discussion, in the context of this paper they may serve as a reasonable estimate of the primitive cosmic abundances. Figure 1 gives an impression of these relative abundances, plotted as a function of the mass number of the nuclides (slightly revised version of the Suess-Urey abundance distribution according to Cameron[4]; abundances relative to silicon $(= 10^6)$).

3) Results

Figures 2a–d compare the relative elemental composition of crustal rock, river water, biological material and seawater with the primitive cosmic abundances of the elements. It is clear that in all instances strong deviations from the most primitive pattern are found. The best resemblance with the Suess-Urey distribution is still found in crustal rock (fig. 2a): most elements follow the straight line, indicating the average ratio between abundances in crustal rock and those in the cosmos, fairly closely. Some lighter elements (N, C, S, Cl) are relatively scarce in crustal rock. These are the elements that presumably escaped into the atmosphere during the process of condensation of the planet. Also come heavy elements are less abundant in crustal rock (Ni, Cr, Co, Se). These are the elements which were withdrawn from the earth's crust during the formation of the mantle and core. Hence, already during the formation of the crust various processes led to the relative enrichment of some elements and to the depletion of others.

In river water (fig. 2b) the abundances of various elements show stronger deviations from the primitive abundance distribution. At the interface between the hydrosphere and crustal rocks other physicochemical processes are acting, generally leading to a further depletion of the abundances relative to carbon. The variation of the differences between river water abundances and cosmic abundances – dashed lines in figures 2 and 3 indicate the standard deviation – is larger (fig. 2b, SD = 1.60; N = 38) than that found with crustal rock (fig. 2a, SD = 1.32; N = 38).

In biological material (fig. 2c) the differences between H_{biol} and $H_{S.U.}$ of various elements are still more variable (SD = 1.66; N = 34). The relation between H_{biol} and $H_{S.U.}$ is still statistically significant (linear correl. coeff. r = 0.7707; N = 34) but it is clear that the resemblance to the primitive distribution pattern is smaller than that found with river water and crustal rock.

In seawater (fig. 2d) the similarity of the element abundances with cosmic abundances is almost lost. The differences between H_{sea} and $H_{S.U.}$ of various elements are highly variable (SD = 2.52; N = 40). Hence, the above comparison of elemental abundances clearly shows that in the given order the resemblance to the primitive cosmic distribution pattern gradually disappears. Crustal rock can be considered to have the most primitive composition, sea water the least primitive.

The composition of biological material ranks itself between river water and seawater. Figure 3 gives a more direct comparison between the relative elemental composition of biological material with that of river water and seawater, showing that the composition of living matter is more closely related to the composition of river water than to that of seawater (SD of the differences respectively 1.24 (N = 37) and 1.81 (N = 35)). Per atom of carbon the availability of O and H in river water is much to high, but it is clear that their presence can easily be affected by evaporation. The ratios of N and P to C in river water are almost exactly the same as those found in living matter; the relative abundances of the other elements are too high. However, the differences are not so large as those found in seawater; the abundance especially of Na and Cl, but also of Mg, S, Ca and Br (relative to carbon) in seawater are much higher than in biological material; the abundance of a fairly large number of heavier metals fits reasonably well with those expected in living matter.

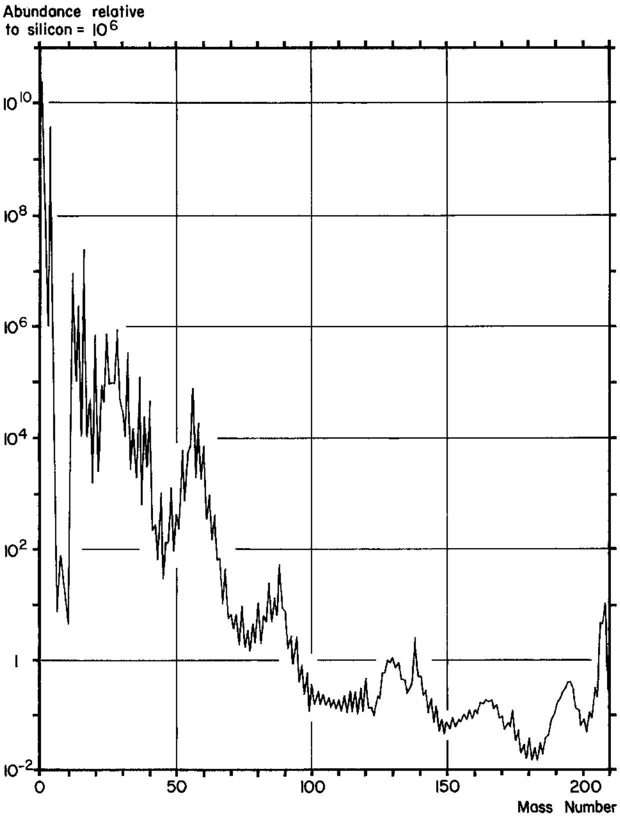
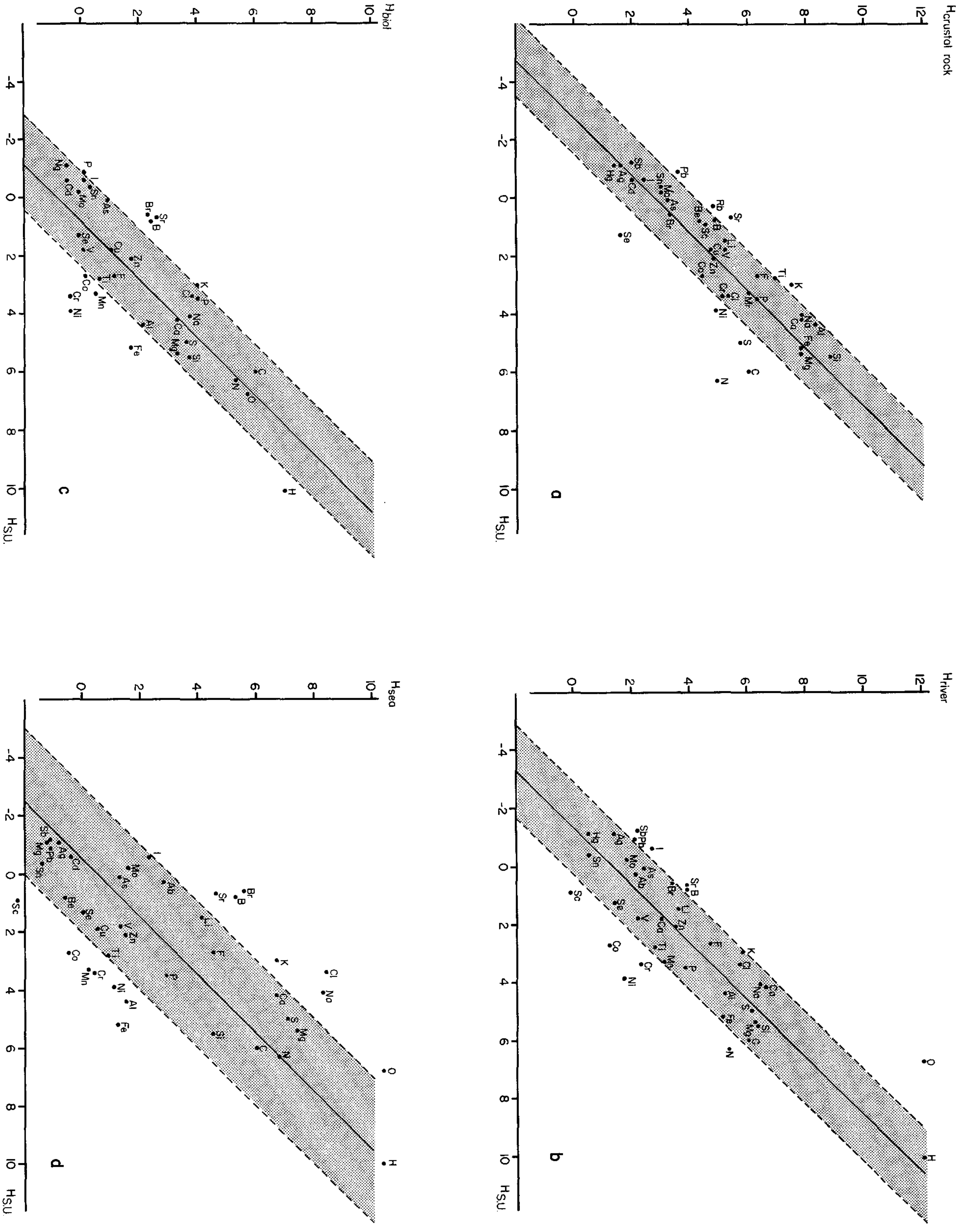
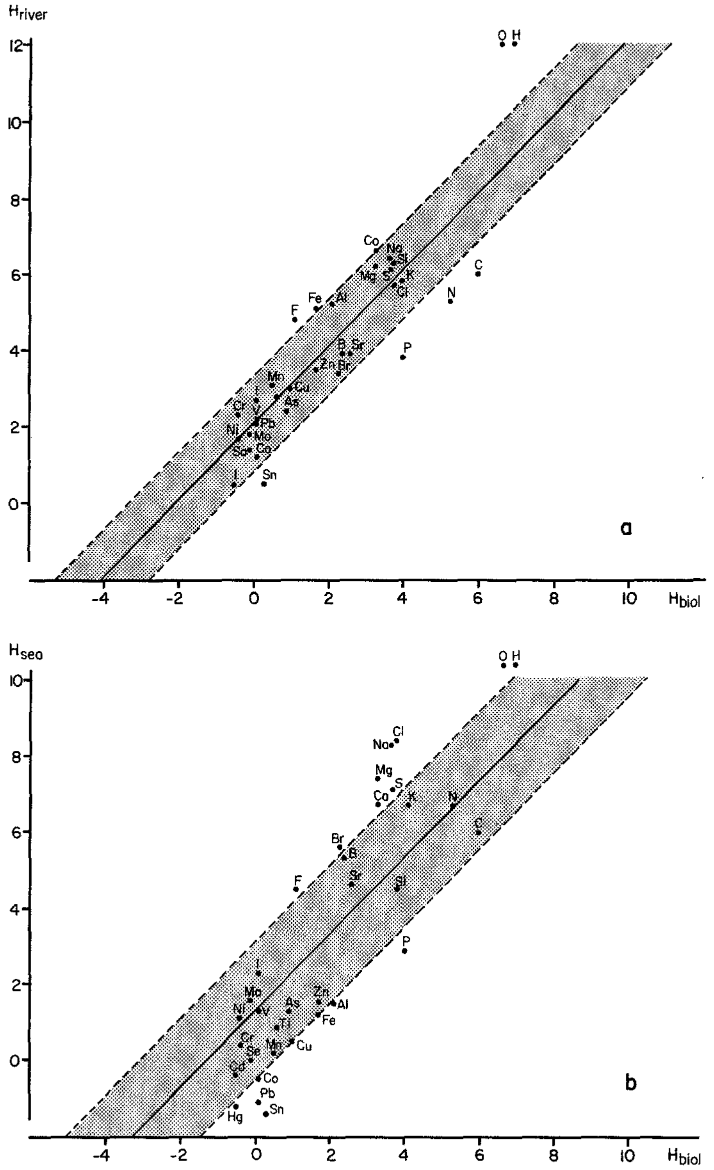


Figure 1. Abundance of the elements relative to silicon (= 10^6) plotted as a function of the mass number of the nuclides. Slightly revised Suess-Urey abundance distribution according to Cameron[4].

Figures 2a–d. Abundances of the elements in crustal rock (a), in river water (b), in biological material (c) and in seawater (d) as a function of the primitive cosmic abundance according to the Suess-Urey distribution. Plotted are the logarithms of the abundances (H) relative to carbon (= 10^6): the drawn lines indicate the average ratio between the elemental abundances plus or minus one times the SD (indicated by the dashed lines).





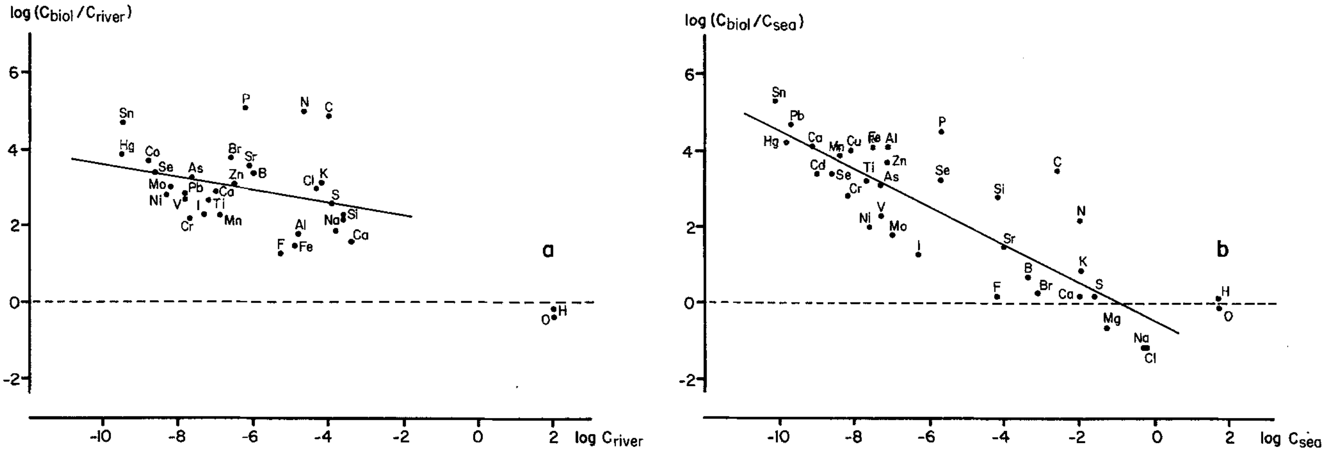
Figures 3a and b. Abundances of the elements in river water (a) and seawater (b) in relation to the corresponding abundances in biological material. Plotted are the logarithms of the abundances relative to carbon (= 10^6); the drawn lines indicate the average ratios, dashed lines represent one times the SD.

The concentration factors (the ratios between actual biological and environmental concentrations) of various elements in river water and seawater are shown in figure 4. Both in river water and in seawater most element concentrations must be strongly increased to obtain the concentrations present in living matter, with the exception of Na, Cl and Mg which must be decreased in seawater. Higher concentrations can be reached by evaporation, which can be effective in fresh water (Darwin's 'warm little pond'; C. Darwin, letter to Hooker, dated 1 February 1871; ref. from M. Calvin[3]) as well as in seawater when it is in rock pools or shallow coastal areas of the sea. In river water a 100-fold concentration yields an elementary composition which fairly well resembles that of living matter, although P, N and C concentrations must be increased to a larger extent.

The concentration factors for river water (fig. 4a) suggest a slight negative relation with the environmental availability of the various elements which, however, appears not to be significant (lin. corr. coeff. −0.3020; N = 31). In seawater, however, a strong inverse relation is found between the concentration factors and the seawater concentrations of the various elements (fig. 4b; lin. corr. coeff. 0.8302; N = 32): the elements having low seawater concentrations must be concentrated more strongly to obtain the biological concentration levels than those of elements that are more commonly available. This differential degree of concentration cannot be obtained by simple evaporation. Thus, although element concentrations in seawater are generally higher than in river water and more closely resemble biological concentrations, the concentration distributions suggest that living matter is more similar to river water than to contemporary seawater.

4) Discussion

Because in representatives of widely divergent taxonomic classes – bacteria, fungi, marine algae, crustaceans,



Figures 4a and b. Concentration factors (ratios between the concentrations in living matter and environmental concentrations) of various elements as a function of their average concentrations in river water (a) and seawater (b). Concentration factor 1, indicating equal concentrations in living matter and environment, represented by horizontal, dashed lines. Linear regression indicated by drawn lines (upper diagram: Y = –0.1603X+1.9812. N = 31, r = –0.3020, not significant. Lower diagram: Y = –0.4883X–0.4315, N = 32, r = –0.8302, highly significant).

finned fishes, molluscs, man – the elemental composition is found to be fairly constant[1,8,19], it can be stated that living organisms are extremely conservative in the composition of their constituent material. Even drastic changes, concurrent with strongly divergent evolutionary processes, did not result in significant changes of the chemical composition of biological material. Apparently, the combinations of biochemical reactions that are characteristic for all contemporary organisms strongly determine the basic pattern of their elemental composition[12]. The basic similarity suggests that the pattern of abundance of the elements was determined in the initial steps of the origin of life and may therefore provide some clue with regard to how living systems arose.

Compared to the elemental composition of natural environments, the composition of living matter is usually strikingly different. Heterotrophic organisms can use other living organisms as food for their growth, and these contain the various elements required in more or less correct rations. Autotrophic organisms, however, usually have to accumulate various body constituents from low environmental concentrations; only in few instances are elemental concentrations in the environment higher than required and these substances have to be expelled. Accumulation as well as excretion of elements against a concentration gradient requires the presence of mechanisms for active transport. These mechanisms usually function at the expense of metabolic energy. In contemporary organisms various mechanisms for active transport have been discovered; probably such mechanisms are quite common and necessary for survival. It is not likely that in the most primitive organisms mechanisms for active transport of body nutrients were already available. It is likely therefore that the primordial chemical environment bears a close similarity to the chemical composition of the early organisms.

The fossil record of marine and terrestrial sediments strongly suggests an oceanic genesis, also showing that the invasion of the land took place only a short (geological) time ago; representatives of all phyla can be traced in marine sediments but on the land and in terrestrial sediments only a limited number of taxonomical classes are represented. The differences in chemical composition between recent seaweater and that of living animals, however, throw serious doubt on the idea of an oceanic genesis, unless the composition of recent seawater differs from ancient seawater[16,17]. The latter view is supported, among other things, by the fact that even in marine species cellular ionic concentrations are always considerably lower than those found in seawater. Those marine species that also regulate their extracellular electrolyte concentrations also maintain ion levels which are always much lower than those in their environment. Similar reasons lead in the panspermia hypothesis to the suggestion of an extraterrestrial location for the origin of life.

When, as in this study, the elemental compositions of river water, crustal rock and biological material are compared with the primitive Suess-Urey abundance distribution it appears that crustal rocks, river water and living matter are more similar to the primitive cosmic abundance composition than seawater. Although the elemental abundances in living matter, river water and seawater are strongly related, the actual concentrations are of course quite different. Especially in river water most concentrations are very low. However, by simple evaporation of the water it is possible to obtain a fluid fairly similar in composition to living matter. To obtain the same result from seawater, not only H_2O but also Na, Mg and Cl have to be removed and, more seriously, many trace elements have to be concentrated more strongly than more common nutrients.

Synthesizing the above results we arrive at the following scenario: at the origin of the earth the abundance of elements in the planet was probably very similar to that given by the Suess-Urey abundance distribution. In the early stages various condensing and physical fractionation processes took place. Elements having a low vaporpressure escape to the atmosphere; heavy elements withdrew into the mantle and the earth's core. Crustal rocks attained a composition which is still comparable to the primitive cosmic element distribution, but serveral deviations are found. Early organisms arose in an environment closely resembling their own body composition. Physicochemical processes, but also the developing organisms, caused a further fractionation of the elements in the geochemical reservoirs, which eventually led to the formation of the present-day earth's curst, ocean and atmosphere. During these developments the organisms retained their original elemental composition; the ocean, however, lost the primitive characterists.

At this point it may be useful to point out the differences of the above hypothesis from the classic hypothesis of Macullum[11]. Firstly, the data considered here refer to the elemental composition of biological material, in the living protoplasm. Macullum focused attention on the composition of body fluids (of marine animals). Secondly, the chemical composition of living matter is considered here to remain constant during all evolutionary developments. Macullum, however, pointed to compositional changes in the body fluids, which he assumed to reflect changes in seawater composition. Both hypotheses suppose that seawater composition has not been constant during the geological past. This is not the generally accepted geological point of view (e.g. Rubey[13]; Holland[9]. The same authors, however, also described geological indications for changes in seawater composition.)

Returning to the question of oceanic genesis, panspermia or fresh water genesis it is clear that above scenario supports the idea of the Darwinian 'warm little ponds' (evaporated river water). An extraterrestrial location for the origin of life can never be completely excluded; the resemblance of the elemental composition of living matter to the primitive Suess-Urey distribution may even give some support to the idea of an nonterrestrial origin. However, the composition of living matter was found to be more similar to river water than to seawater. Also, the accumulation of Mo in biosystems, used as an argument for panspermia, did not become evident in the present data. Therefore it is concluded here that there is no reason to assume a nonterrestrial origin.

Considering the question which elements are necessary for the functioning of biological systems, Egami[6] posed the hypothesis that only elements that occur in seawater in concentrations above a critical concentration range (1–5 nM) became essential (or neutral); elements having lower concentrations were excluded from organic evolution. In the latter case, organisms evolved independently, gaining neither the capacity to use them, nor the capacity to tolerate them as neutral elements. Consequently, such elements might be toxic in higher concentrations. This hypothesis, which also postulates constancy of the seawater composition is based on only a few data (simply because of the fact that element concentrations below 1 nM are extremely difficult to measure). The biological accumulation of rare elements from seawater generally increases towards lower sea water concentrations (fig. 4b) and there is no indication that this accumulation stops below concentrations of 1 nM. It seems safer not to use the rather arbitrary classification of elements into essential, neutral and toxic ones. Moreover, many essential elements may also be toxic in high concentrations and, conversely, several toxic or neutral elements have appeared, with progresssing knowledge, to be essential.

The results as reported in this paper are in favor of an origin of life on earth in a 'warm little pond' (evaporated river water). This agrees with recent synthetic theories which also assume an unbroken pathway for the emergence of biosystems: starting from a structurally diversified environment, the presence of energy rich nucleotide-like compounds in a suitable physicochemical environment may favor and later drive the replication of short polymer strands, leading to a primitive genetic machinery[10]. Then, further change through time is generally accepted to be chiefly the product of subsequent interactions between genotypes and their environment[21].

Acknowledgments. The author wishes to thank the participants in the summer school on chemical evolution and the origin of life (organized by the Council of Europe division for higher education and the research group on cosmic chemistry, chemical evolution and exobiology, at Stevensbeek, the Netherlands, 4–10 July 1983) for many stimulating discussions. Thanks are also due to Mr A.D.G. Dral and Dr J.J. Beukema, both colleagues at the Netherlands Institute for Sea Research, Texel, for critically reading and amending the manuscript.

1 Banin, A., and Navrot, J., Origin of life: clues from relations between chemical compositions of living organisms and natural environments. Science *189* (1975) 550–551.
2 Brewer, P.G., Minor elements in seawater, in: Chemical oceanography I, pp. 415–498. Eds J.P. Riley and G. Skirrow. Academic Press, London 1975.
3 Calvin, M., Chemical Evolution, p. 5. Clarendon Press, Oxford 1969.
4 Cameron, A.G.W., The origin of the elements, in: Evolution: its science and doctrine, pp. 225–242. Ed. J.W.M. Cameron. Univ. of Toronto Press, Toronto 1960.
5 Crick, F.H.C., and Orgel, L.E., Directed transpermia. Icarus *19* (1973) 341–346.
6 Egami, F., Minor elements and evolution. J. molec. Evol. *4* (1974) 113–120.
7 Eisler, R., Trace metal concentrations in marine organisms, p. 685. Pergamon Press, Oxford 1982.
8 Gualtieri, D.M., Trace elements and the panspermia hypothesis. Icarus *30* (1977) 234–238.
9 Holland, H.D., The geological history of seawater – an attempt to solve the problem. Geochim. cosmochim. Acta *36* (1972) 637–651.
10 Kuhn, H., and Waser, J., Molecular selforganisation and genesis of life. Experientia *39* (1983) 834–840.
11 Macullum, A.B., The palaeochemistry of body fluids and tissues. Physiol. Rev. *6* (1926) 316–357.
12 Oparin, A.I., in: Exobiology, p. 11. Ed. C. Ponnamperuma. North-Holland Publ. Co., Amsterdam 1972.
13 Rubey, W.W., Geological history of seawater – an attempt to state the problem. Bull. geol. Soc. Am. *62* (1951) 111–1148.
14 Sidwell, V.D., Buzzel, D.H., Foncannon, P.R., and Smith, A.L., Composition of the edible portion of raw (fresh and frozen) crustaceans, finfish and molluscs II. Macro elements: sodium, potassium, chlorine, calcium, phosphorus and magnesium. Mar. Fish. Rev. *39* (1977) 1–11.
15 Sidwell, V.D., Lonnis, R.L., Lonnis, K.J., Foncannon, P.R., and Buzzel, D.H., Composition of the edible portions of raw (fresh and frozen) crustaceans, finfish and molluscs, III. Micro elements. Mar. Fish. Rev. *40* (1978) 1–21.
16 Spaargaren, D.H., A comparison of the blood osmotic composition of various marine and brackish-water animals. Comp. Biochem. Physiol. *60* (1978) 327–333.
17 Spaargaren, D.H., A comparison of the composition of physiological saline solution with that of calculated Pre-Cambrian seawater. Comp. Biochem. Physiol. *63A* (1979) 319–323.
18 Spaargaren, D.H., Some aspects of element turnover in living organisms. Acta biotheor. *32* (1983) 13–28.
19 Spaargaren, D.H., and Ceccaldi, H.J., Some relations between the elementary chemical composition of marine organisms and that of sea water. Oceanol. Acta *7* (1984) 63–76.
20 Spaargaren, D.H., Elemental composition and interrelations between element concentrations in marine finfish, molluscs and crustaceans. Neth. J. Sea Res. *17* (1985, in press).
21 Stebbins, B.L., Mosaic evolution: an integrating principle for the modern synthesis. Experientia *39* (1983) 823–833.

22 Suess, H.E., and Urey, H.L., Abundances of the elements. Rev. mod. Phys. *28* (1956) 53–74.
23 Vinogradov, A.P., The elementary composition of marine organisms. Yale University, New Havens 1953.
24 Whitfield, M., The main oceanic residence time (MORT) concept – a rationalisation. Mar. Chem. *8* (1979) 101–123.
25 Yamamoto, T., The relation between the concentration factor in seawater and residence time of some elements in seawater. Rec. Oceanogr. Wks Japan *11* (1972) 65–72.
26 Yamamoto, T., Otsulla, Y., and Okamoto, K., A method of data analysis on the distribution of chemical elements in the biosphere, in: Analytical techniques in environmental chemistry, pp. 401–405. Ed. J. Albinges. Pergamon Press, London 1980.

0014-4754/85/060719-08$1.50 + 0.20/0

Short Communications

Purification and properties of glutamine synthetase I from *Rhizobium* sp. UMKL 20

S. T. Lim*

Department of Biochemistry, University of Malaya, Lembah Pantai, Kuala Lumpur (Malaysia), 31 January 1984

Summary. Glutamine synthetase I was purified from *Rhizobium* sp. UMKL 20 following polyethylene glycol precipitation. The enzyme had a subunit molecular weight of 58 kd. Apparent K_m values for ammonia and glutamate were 5.6 and 15.2 mM, respectively. Glutamine synthetase I activity was inhibited by several end products of glutamine metabolism. The purified enzyme was highly adenylylated ($E_n^- = 8.5$).

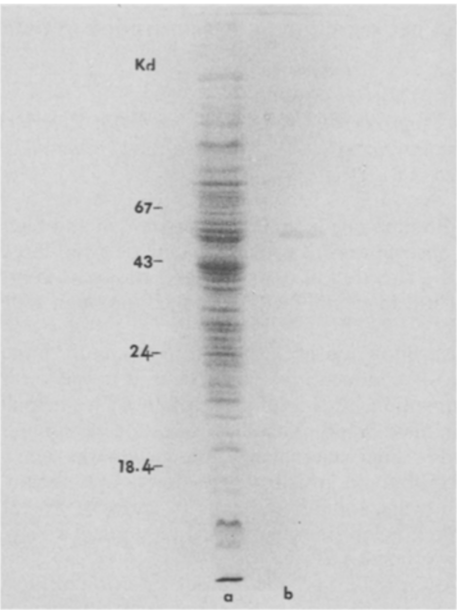
Key words. Rhizobium; glutamine synthetase I.

The enzyme glutamine synthetase (GS, EC 6.3.1.12) has been shown to play an important role in nitrogen assimilation in many microorganisms. Recent studies have focused on the mechanism of regulation of the enzyme and the molecular events leading to its expression. The GS from *E. coli* has been widely characterized by Stadtman and his co-workers[1].

Members of the family Rhizobiaceae have been shown to possess two forms of glutamine synthetase; GS I and GS II. GS I is heat stable and is similar to the GS from other microorganisms while GS II is heat labile (50°, 30 min)[2]. Besides its role in ammonia assimilation, GS I from *Rhizobium* is believed to play a role in the regulation of symbiotic nitrogen fixation by controlling the expression of nitrogenase[2,3]. The role of GS II is not known at present[3]. Recently it was shown that GS I may also be involved in salt tolerance in *Rhizobium*[4,5]. Under salt-stressed conditions, GS I activity increased several-fold[4]. In view of the important roles this enzyme may play in *Rhizobium,* I report here the purification and characterization of GS I from a salt-tolerant *Rhizobium,* UMKL 20[4].

Materials and methods. Rhizobium sp. UMKL 20 was obtained from the Department of Genetics and Cell Biology. Rhizobial cultures were maintained on agar slants of mannitol-yeast extract medium[4]. Liquid cultures were grown at 30° starting from a 2% inoculum (absorbance at 420 nm = 0.1). Total GS activity was assayed using the γ-glutamyl transferase assay described by Shapiro and Stadtman[6]. The biosynthetic assay of GS was carried out using ^{14}C-glutamate essentially as described by Prusiner and Milner[7]. The assay was modified with QAE-Sephadex (A25) replacing Dowex-1 (Cl^-) to improve the flow rate. All assays were carried out at pH 7.25, the isoactivity point of GS I from UMKL 20. GS I was purified using polyethylene glycol (PEG) precipitation essentially as described by Streicher and Tyler[8]. SDS-polyacrylamide slab gel electrophoresis was carried out using the procedure of Laemmli[9].

Results and discussion. Glutamine synthetase I was isolated and purified from *Rhizobium* sp. UMKL 20. Table 1 shows a summary of the purification procedure. The PEG precipitation of GSI occurs presumably because of a complex formed between GS I and DNA[8]. The procedure is a simple, fast and efficient method for the purification of GS I. Although the specific activity of the purified GS I is similar to other purified GSs[8,10,11] the yield was much lower. This is probably due to the difficulty encountered in solubilizing the acetone-precipitated pellet. The enzyme was judged to be homogeneous by SDS-polyacrylamide gel electrophoresis[9]. Only a single protein band was obtained, with a subunit mol. wt of 58 kd (fig.). This value is similar to the subunit molecular weight for the enzyme from other procaryotes[11]. Homogeneity was also apparent from immunodiffusion studies when antiserum against purified GS I was cross-reacted with crude extracts from strain UMKL 20. Only a single precipitin line was obtained[12] (data not shown). The purified enzyme was highly adenylylated, having an ade-



SDS-polyacrylamide gel electrophoresis of crude extracts (lane a) and purified glutamine synthetase I (lane b) from *Rhizobium* sp. UMKL 20.