TAO, Vol.6, No.1, 91-102, March 1995

Mineral-geochemical Association in Bottom Sediments of the East China Sea

ANATOLY S. ASTAKHOV[1], ALEXANDER I. BELOGLAZOV[2] and ANATOLY V. MOZHEROVSKIY[1]

(Manuscript received 30 April 1994, in final form 16 December 1994)

ABSTRACT

Geochemistry and mineralogy of grain-size fractions from Holocene sediments of the East China Sea and the adjacent regions have been investigated. The correlation between minerals and chemical element contents was determined for both sandy and clayey fractions. The averages contents of rock-forming and some rare elements were found changing from coarse to fine fractions with the decrease in SiO_2 and the increase in actually all other elements. In general, geochemical differences between coarses and fines fractions are equivalent to acidic and intermediate (or basic) igneous rock. Statistical cluster analysis for rock-forming and some rare elements in 1-5μm fractions revealed three zones having different sediment sources. The central zone occupied the whole outer East China Sea Shelf. Sediments (1-5μm fraction) supply sources for it are terrigenous discharge from Taiwan Island and the Philippine Archipelago.

(Key words: Geochemistry, Mineralogy, Bottom sediment, East China Sea)

1. INTRODUCTION

The East China and Yellow Seas (due to their huge terrigenous supply) offer unique opportunities for the understanding of sedimentary basin formation within epicontinental seas. Analogous ancient sedimentary basins in many regions are oil and gas-bearing, and the study of their formation conditions is needed for the recognition of hydro-carbonic nonstructural deposits.

The most popular properties for the comparison of recent sediments and ancient sedimentary rocks are their geochemical characteristics. The geochemical technique is the same for rock of any age. However, geochemical data are seldom used to determine the sources supplying sedimentary materials. Actually the source is determined by the composition of sandy and clayey minerals. This is explained by many reasons, with the main one being the

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[1] Pacific Oceanological Institute, Russian Academy of Sciences, 43 Baltiyskaya St., Vladivostok, 690043, Russia

[2] Far East Geological Institute, Russian Academy of Sciences, 159 Stoletiya St., Vladivostok, 690022, Russia

considerable difference between sandy and clayey sediments, which makes it impossible to notice changes in the geochemical indicators throughout a section or an area.

Methods of fractional analysis, similar to a mineralogical investigation, were applied in this work with the help of geochemical data to reveal the possibility of source recognition. Moreover, another purpose of this work was to reveal the terrigenous matter removal from different margins and its role in the formation of the East China Sea Shelf sediments. Despite the thorough study of the East China Sea recent sediments, the role of different sources in deposit formation is still unclear. The sources of sandy discharge in the East China Sea are more definite. They are the Changjiang River and surrounding areas (Wang and Liang, 1982; Wang *et al.*, 1984; Derkachev and Nikolaeva, 1992). The sources of the clayey part of sediments are less defined. They are the Huanghe and Changjiang Rivers (Xu, 1983; Butenko *et al.*, 1983), Taiwan Island (Chen *et al.*, 1992) and even the current from the Japan Sea (M.-P. Chen, 1993, verbal communication). According to Milliman and Meade (1983), the main terrigenous contribution to the region was made by both continental rivers (Huanghe and Changjiang), Taiwan Island and the Philippine Archipelago. This suspended material can be transported by the Kuroshio Current to the East China Sea (Figure 1).

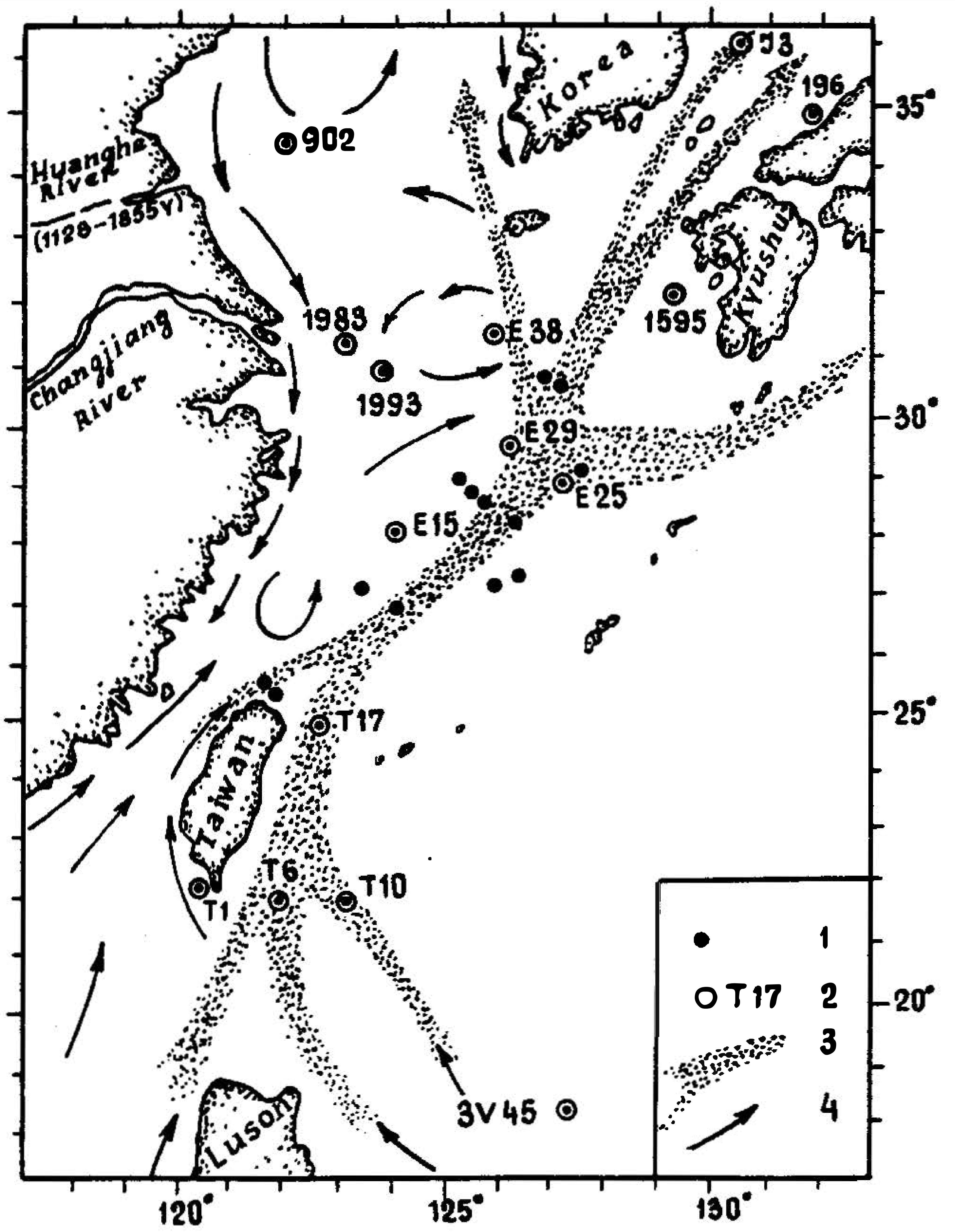

Fig. 1. The Regional surface water circulation (after Niino and Emery, 1961) and station locations. 1- stations of sandy fractions investigation; 2- stations of the muddy fraction investigation; 3- Kuroshio Current (main stream); 4- other surface currents.

2. METHODS

The same cores picked up on board of the *R/V "Akademik A. Vinogradov"* during the Cruise V23/KM92 (Figure 1) and containing by correlation with the cores determined earlier, reliable Holocene terrigenous sediments were used in the present investigation. The Sediment samples of a large volume were divided into grain-size fractions using the standard technique of size-fraction analysis (Petelin, 1967). The sandy fractions (1000-500, 500-250, 250-100, 100-50 μm) were separated by wet sieving, while the silty and clayey size fractions (50-10, 10-5, 5-1, <1 μm) by the pipette method.

In addition to the samples taken by the *R/V "Akademik A. Vinogradov"*, some other Holocene sediment samples from different expeditions were presented by A. I. Botsul (Stations 1983, 1993, Figure 1), S. A. Gorbarenko (Station 1596), A. N. Derkachev (Station 196), A. S. Astakhov (Station 902). The age of some of these samples has been reasonably well determined (Gorbarenko, 1990).

For composition investigations smear slides were made and the contents of the main components (sandy, clayey, biogenic, etc.) were determined using the microscopic semi-quantitative method. The mineral composition of clay fractions was determined by semi-quantitative X-ray diffraction methods with the diffractometer "DRON-2.0". The determination of Fe, Ti, Mn, Y, Zr, Nb, Rb, Sr, Ba contents in fractions were made by an X-ray fluorescence spectroscopy using an X-ray multi-channel spectrometer "SPARK-2". The rock-forming element and La, Ce, Nd, Th, U contents were established by X-ray fluorescence methods in the Central Laboratory of the Regional Office of the Russian Geological Survey "Primorgeologiya" by Dr. E. V. Chulkov.

3. MINERAL AND CHEMICAL CORRELATION

Multivariate statistical analyses were done separately for the sandy and clayey fractions to reveal any communality of minerals and chemical elements. For the sandy fractions of bottom sediments from the East Asian Marginal Seas, the correlation between minerals and rock-forming elements had been investigated earlier (Astakhov, 1994). Most of the rock-forming and some rare elements correlate well with the mineral contents: SiO_2 - with quartz, CaO and LOI - with calcareous remains, K_2O - with "glauconite" (all green clay aggregates), Zr - with zircon, TiO_2 - with rutile and ilmenite. The correlation between minerals and some constituent elements (calcareous remains - with Sr, zircon - with Y) is appropriate. Some elements, related to those rare minerals not determined by standard mineralogical analyses, could correlate with some other associated minerals. Zircon, for instance, correlates with elements of any other heavy stable minerals - the sum of La, Ce, Nd contents, TiO_2, MgO.

For sandy fractions, three associations of chemical elements are revealed on the factor diagram of the first two factors (Figure 2A). These associations correspond to definite groups of minerals. The first one includes some elements, Zr-Ti-REE-P, of heavy stable minerals (zircon-monazite-rutile). Other elements, Y, Nb, Mg, partly Fe and Mn, from heavy minerals of basic rocks (ilmenite, magnetite, pyroxene) are also attracted to this association. The second one, K-Rb-Na-Al, is represented by the elements of feldspar and "glauconite". The third association, Ca-Sr-LOI, is composed of the elements of the biogenic calcareous remains. Silica has no considerable positive correlations with other elements, which means it is mainly found in quartz.

Thus, the first factor values on diagram (Figure 2A) demonstrate the possibility of distinguishing recognizing authigenic components from terrigenous ones. The maximal positive value of the first factor is connected with elements occurring in heavy stable minerals, while the analogous negative value is associated with the elements of the biogenic components (Ca-Sr-LOI), "glauconite" (K-Rb), oxides or other diagenetic minerals (Mn-Ba-U). Iron and manganese are included in both terrigenous clastic minerals (pyroxene, magnetite, ilmenite) and authigenic ones ("glauconite" and oxides), which are wide-spread in shelf sediments (Zhu, 1983). Hence, they do not correlate with any associations, and they are characterized by a small positive value of the first factor.

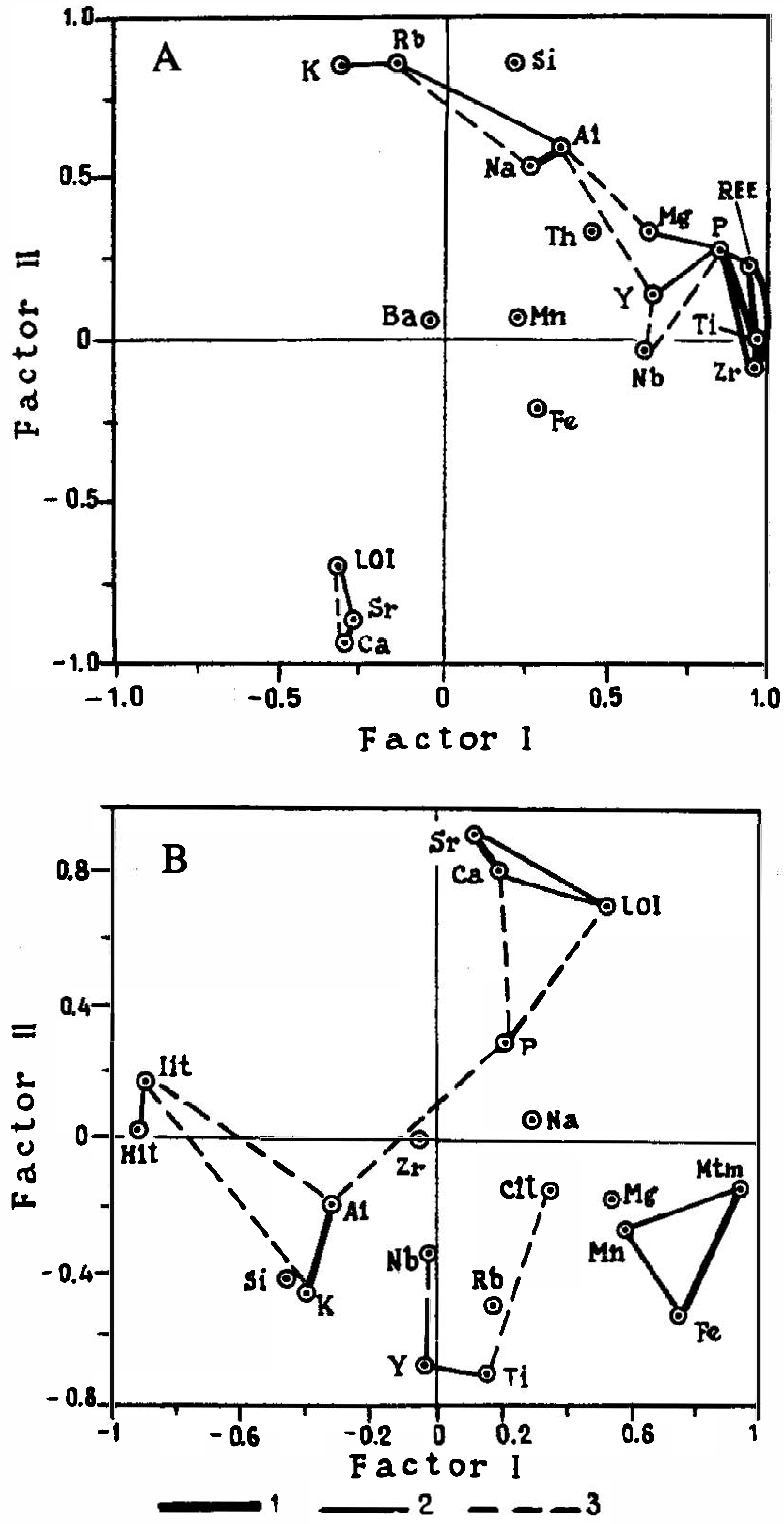

Fig. 2. Factor diagrams for the first two factors and correlations of chemical elements and clay minerals (Ilt - hydromica, Hlt - chlorite, Clt - kaolinite, Mtm - smectite) for sandy (A) and clayey (B) sediment fractions. a b 1-3 correlations: 1 - very good, 2 - good, 3 - significant.

Table 1. Clay mineral composition (expressed as a 100% total clays).

Station numbers	Fraction, m	Hydromica	Chlorite	Kaolinite	Smectite	Quartzfeldspar ratio
E15	5-10	76	17	6	1	2.80
E15	1-5	64	27	6	3	4.25
E15	<1	71	8	5	13	8.88
E25	5-10	74	25	0	1	1.81
E25	1-5	68	25	7	5	3.67
E25	<1	63	14	5	18	4.13
E29*	5.10	66	24	7	3	2.75
E29*	1-5	60	27	9	4	5.00
E29*	<1	67	9	4	20	4.16
E29	5-10	68	20	10	1	2.75
E29	1-5	67	20	11	2	4.23
E29	<1	64	13	7	16	3.09
E38	5-10	69	20	8	3	1.30
E38	1-5	64	25	9	2	3.87
E38	<1	58	9	6	27	2.87
T01	1-5	67	33	0	0	5.20
T01	<1	78	20	2	1	3.81
T06	5-10	69	30	0	1	3.30
T06	1-5	62	32	2	4	3.58
T06	<1	66	21	4	9	4.79
T10	5-10	66	28	0	6	1.93
T10	1-5	62	31	0	6	2.90
T10	<1	60	16	2	22	2.61
T17	5-10	65	34	0	1	2.00
T17	1-5	71	26	2	1	4.32
T17	<1	65	14	4	17	3.81

*samples of the Late Quaternary sediment, 350-370 cm core interval.

The correlation between the clay minerals and chemical elements for clayey fractions is less significant because the clay mineral contents were only detennined for the total clay (Table 1). However, minerals/chemical elements correlation is revealed both for the correlation matrix and for the factor diagram (Figure 2B). Hydromica correlates with Al and K, included in its crystal lattice. Silica is also attracted to this association. Furthermore, hydromica correlates with chlorites as they have common sources of supply. Therefore, the latter doesn't correlate with Mg, Fe and Na forming its chemical composition. Smectite correlates rather well with Fe, Mn, and poorly with Rb, Na and Mg although they form combined associations (Figure 2B). All these elements form the crystal structure of smectite. Besides the associations mentioned above and the contents related to hydromica and smectite on the factor diagram, others could also be distinguished : Ca-Sr-LOI-P from biogenic components and Si-Ti-Y-Nb-Zr from sandy components. Kaolinite, as a characteristic mineral of terrigenous supply, is well related to the latter association although its elements are not typical of this mineral.

4. FRACTIONAL DISTRIBUTION OF CHEMICAL ELEMENTS

Based on average fractional contents (Figure 3), some chemical elements concentrated in the coarsest fractions (SiO_2, CaO, Sr) and in the finest ones (Al_2O_3, MgO, TiO_2, Mn, P_2O_5, etc.) could be distinguished. The data obtained in other areas testify to similar distributions; moreover, they reveal the greatest concentration of some heavy metals (Co, Ni, Cu, Zn, Cd) in the fine clayey fractions with their contents tens or hundreds of times greater than in sand fractions (Astakhov and Makarevich, 1989). The results of the statistical analyses mentioned above made the determination of the reasons for the irregular distribution of rock-forming and rare elements in fractions possible. The increase in the SiO_2 content, which is normally determined in coarse fractions by the total concentration of silicates, is conditioned by clastic quartz abundance. The peculiarity of SiO_2 distribution in fractions is the local increase in its concentrations in fractions 5-10 μm (Figure 3), which is not typical of other regions. This is, probably, caused by the intensive supply of loessal silt grain-size material from the continent.

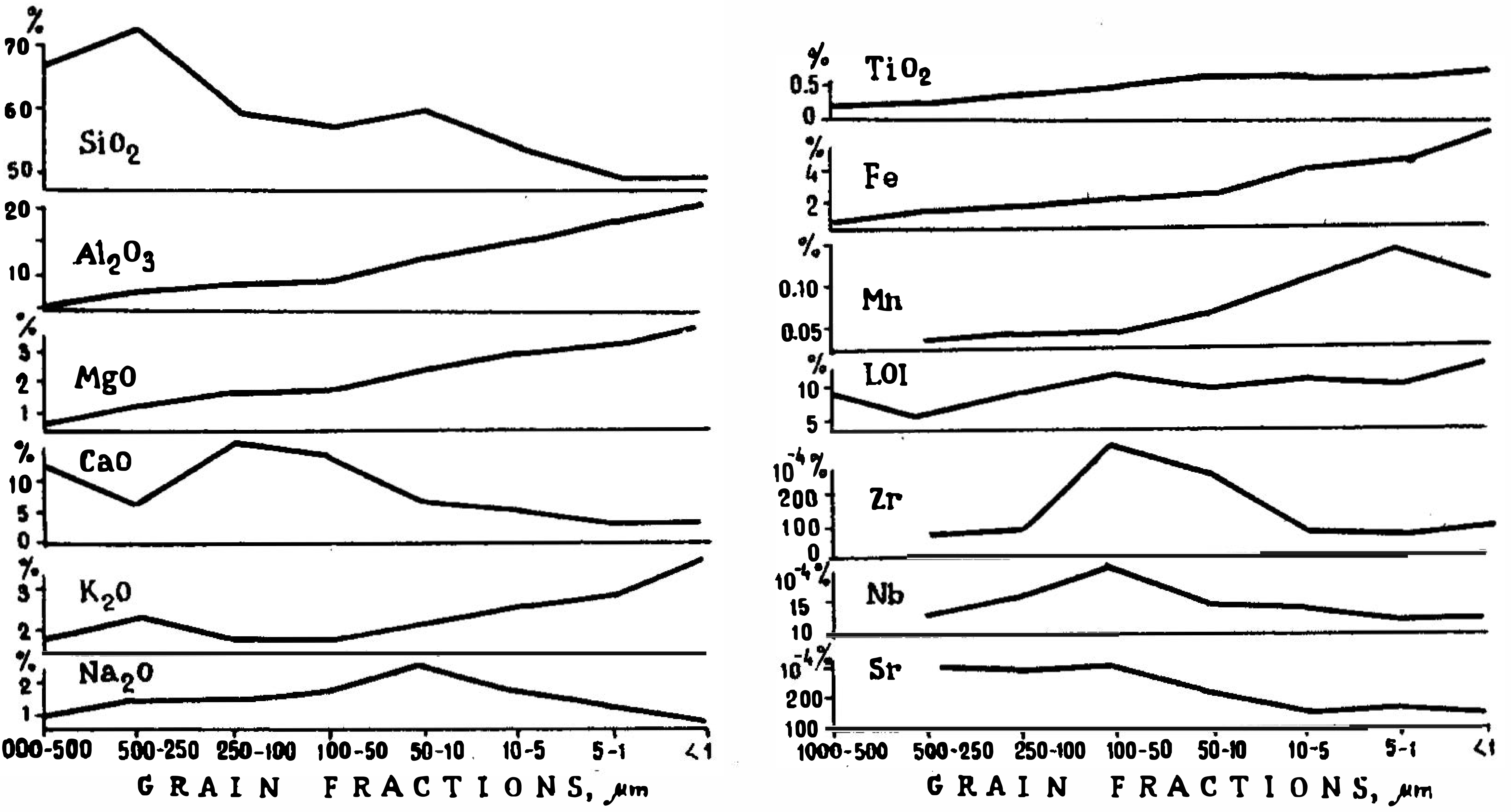

Fig. 3. The Chemical element distribution (mean contents) in different sediment grain-size fractions.

Elements of carbonates (CaO, LOI, Sr) are attracted to coarse fractions, which is associated with the increase in foraminifers and, probably, the fine shell-detritus in them. The content of carbonate elements in fine fractions is high enough. It is associated with the biogenic remains (coccolithophorides) and, probably, the supply of fine-grained clastic carbonate of loessal soils. The increase in rock-forming and many rare element contents in fine fractions can be explained through their connection with clayey minerals - hydromica, kaolinite (Al_2O_3, K_2O) or smectite (MgO, Fe, partially) as well as with finely dispersed oxides and the hydroxides of Fe and Mn, sulfides and other authigenic components (Mn, Fe, Rb, P). The distribution of K_2O in fractions is determined by hydromicas mostly: illite - in fine fractions and "glauconite" - in coarse ones. The maximum of the latter is in the 250-500 μm fraction.

The contents of some elements increase in silty fractions. In many cases this is determined by the concentration of heavy minerals - zircon and monazite mostly (for Zr, Y, Nb and partially P_2O_5). The highest contents of Zr, and zircon, respectively, are found in the 10-50 μm fraction, but in other regions they characterize the 50-100 μm one. This is caused by the more intensive mechanical grinding and crushing of zircon grains as a result of alteration, redeposition and transportation on the continent (Chinese Platform) during the long period. High contents of zircon in the 10-50 μm fraction can be noticeable in other platform areas. The Mn increase is due to its content in Mn-sulfides or micronodules.

The analysis of the distribution of chemical elements in sediment fractions revealed great geochemical differentiation results in the simple separation of terrigenous matter in size and, mineral composition. This differentiation occurs after the alluvium supply into a marine basin and can completely distort the initial geochemical distinctions of sediments contributed by different sources. Thus, the source area influence on the geochemistry of bottom sediments can be eliminated by the size differentiation of terrigenous material in course of deposition. Under these conditions, the total geochemical composition of sediments gives no clear evidence for the understanding of the source of terrigenous material.

5. BOTTOM SEDIMENT DIVISION OVER 1-5μm FRACTION GEOCHEMICAL DATA

On the basis of the approximate estimation of possible transportation by the Kuroshio current, the 1-5 μm fraction was chosen for sediment source analysis. The Sedimentation rate of the particles was calculated the Stoke's Formula. It is 0.32 cm/h for 1 μm grains, 6.58 cm/h for 5 μm grains and 32 cm/h for 10 μm grains. Before it reaches the 100 m depth, the 1 μm fraction can be spread to about 30 thousand km, the 5 μm fraction to about 5 thousand km and the 10 μm fraction to about 900 km if the average velocity of the Kuroshio current is 1 m/sec. Subsequently, the material of the 1-5 μm can be supplied to the East China Sea Shelf from Taiwan and even the Luson Islands (Figure 1). The coarser part of the 5-10 μm fraction can be transported by the Kuroshio current only from Taiwan in the southern part of the East China Sea. Material smaller than 1 μm in suspension can be brought by all branches of the Kuroshio: the ones from the South China Sea, from the Pacific Ocean (East Boundary current), from the Philippine Archipelago region and from Taiwan Island. Most parts of the suspended matter of these sizes is transported by the Kuroshio current far beyond the East China Sea to the open ocean and the Japan Sea. Passing these distances, this material is mixed with analogous material plumed by continental rivers. That is why the 1-5 μm fraction can be a better help in the fine grained matter flux determination than the 1 μm, 5-10 μm and the coarses ones. There are also examples of the effective application of the <1 μm fraction for the study of the suspension distribution in the ocean (Gorbunova, 1970).

A statistical multi-component cluster analysis was used to single out geochemical provinces. Rock-forming and some rare elements, with a small physical-chemical migrate capability were chosen for cluster analysis (Table 2). Three geochemical provinces were

Table 2. The chemical composition (rock-forming elements -%, Rb, Sr, Y, Zr, Nb - ppm) of 1-5 m fraction of Holocene sediments being used for cluster analysis.

Station umbers	SiO_2	Al_2O_3	CaO	MgO	K_2O	Na_2O	P_2O_5	MnO	TiO_2	Fe_2O_3	LOI	Rb	Sr	Y	Zr	Nb
T01	53.77	21.50	1.50	2.59	3.59	1.01	0.19	0.140	1.10	7.51	6.42	111	58	46	58	14
T06	53.78	21.77	1.01	2.78	4.11	1.50	0.18	0.120	1.01	7.31	6.06	80	43	49	51	11
T10	55.97	19.00	0.95	3.26	3.89	1.62	0.17	0.920	0.94	7.60	5.09	83	57	40	44	10
T17	55.65	17.22	3.86	3.05	3.29	1.15	0.18	0.081	0.86	6.31	7.99	104	92	35	60	13
E15	52.54	17.09	3.75	3.13	3.53	1.32	0.19	0.095	1.04	8.17	8.30	109	130	45	139	19
E25	49.75	15.69	8.88	2.68	2.58	1.18	0.22	0.120	0.69	5.04	12.52	94	230	31	76	14
E29*	57.55	19.33	0.86	3.10	3.41	1.21	0.15	0.094	1.02	6.13	6.52	88	51	45	45	12
E29	51.42	17.65	5.09	3.18	3.11	1.47	0.21	0.100	0.92	6.87	9.31	125	166	34	86	18
E38	56.39	17.36	2.51	3.26	2.75	1.50	0.20	0.180	1.01	7.41	6.85	124	100	39	73	18
196	43.80	15.34	11.06	2.71	1.78	0.79	0.14	0.040	0.58	4.96	18.29	68	414	3	95	12
902	48.65	20.78	0.45	3.07	3.65	0.84	0.09	0.280	0.70	8.16	12.91	142	80	5	134	11
1595	45.60	15.22	6.48	2.64	2.78	3.40	0.07	0.100	0.66	7.75	14.91	85	195	32	69	15
1983	49.19	20.01	3.82	3.30	3.05	0.64	0.16	0.100	0.73	7.82	11.15	155	132	27	135	10
1993	53.82	19.65	1.29	2.97	2.98	1.25	0.15	0.040	0.76	7.90	8.80	147	122	27	150	11
3V45	54.29	19.63	1.14	3.39	2.56	2.02	0.18	0.460	0.70	7.70	6.82	90	48	39	41	11

*the sample of Late Quaternery sediment, 350-370 cm core interval.

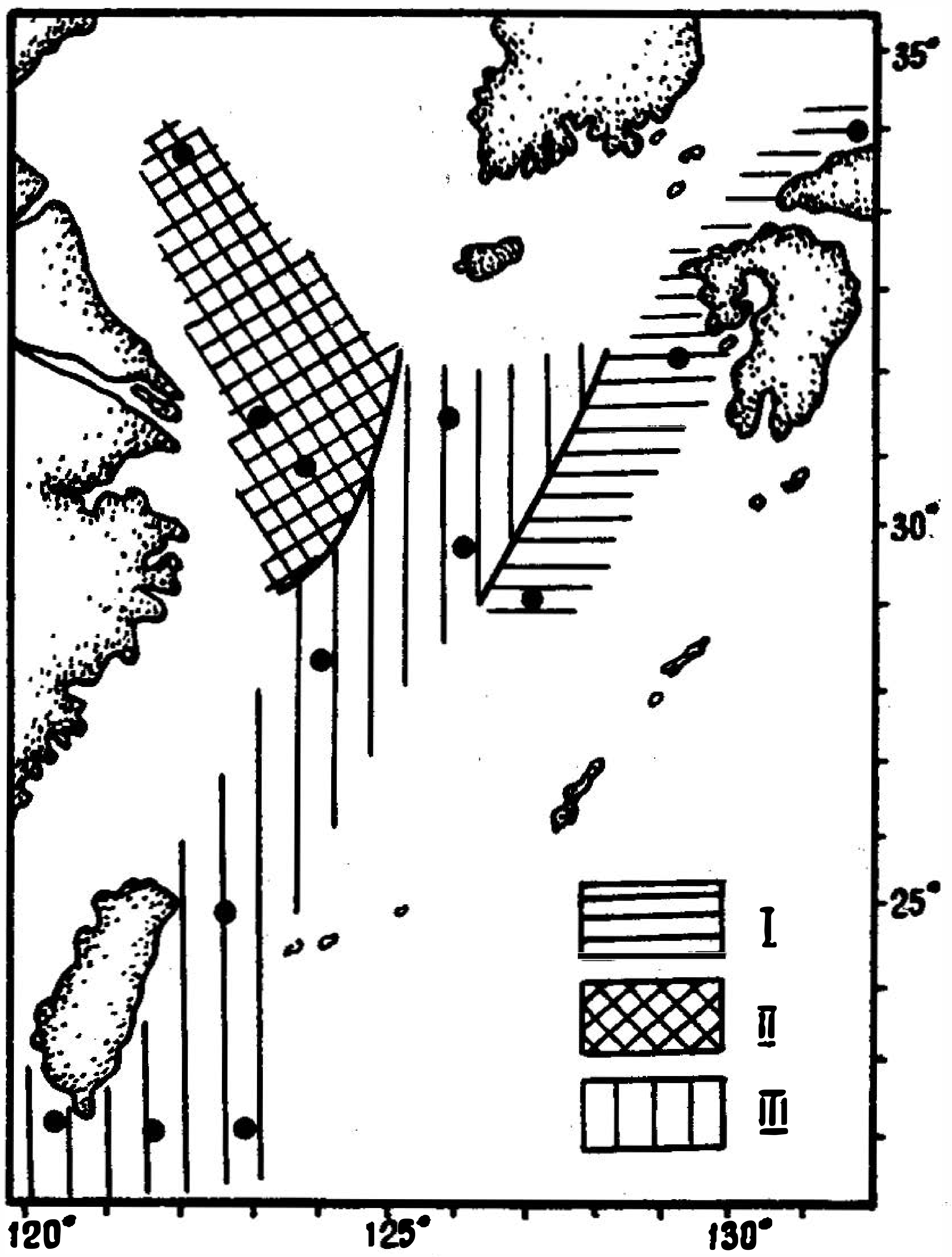

Fig. 4. The Geochemical provinces for the 1-5 μm fraction of bottom sediments (as the result of cluster statistical analysis). I-III - geochemical provinces (see the text).

determined in this area (Figure 4). The first province includes the north-eastern part of the region and is separated from the mainland by the main stream of the Kuroshio current. It is geochemically characterized by a high content of biogenic elements, which could be explained by high biological productivity or a decreased flux of a mineral suspension. The second province includes the zone of mineral suspension supplied by the Huanghe and, probably, Changjiang Rivers. The sediments of this province have relatively low contents of SiO_2 but high contents of Al_2O_3, K_2O, Zr, TiO_2. The third province includes the East China Sea outer shelf and the Taiwan nearshore region. A high concentration of Y, Nb and an abnormal changeability of MgO, Al_2O_3, K_2O contents are characteristic of it. A further detailed elaboration of the cluster analysis makes it possible to separate stations T10, T1, T6, T17, consequently, from the province. The province's sediments (fraction 1-5 μm) are formed by the admixing of suspensions supplied by different branches of the Kuroshio current, but the basis of the 1-5 μm fraction is formed by the material supplied from the Philippine Archipelago and Taiwan Island. In the East China Sea it is transported by the Kuroshio current.

The influence of the Kuroshio current branches on material transportation to the East China Sea bottom sediments can be elaborated upon in detail when the distribution of the chemical elements is taken into account , Al_2O_3 distribution (Figure 5) determined by the hydromica contents makes it possible to observe two sources of the flux - the Huanghe-Changjiang River mouths and Taiwan - Luson Islands. MgO distribution (Figure 6) is related

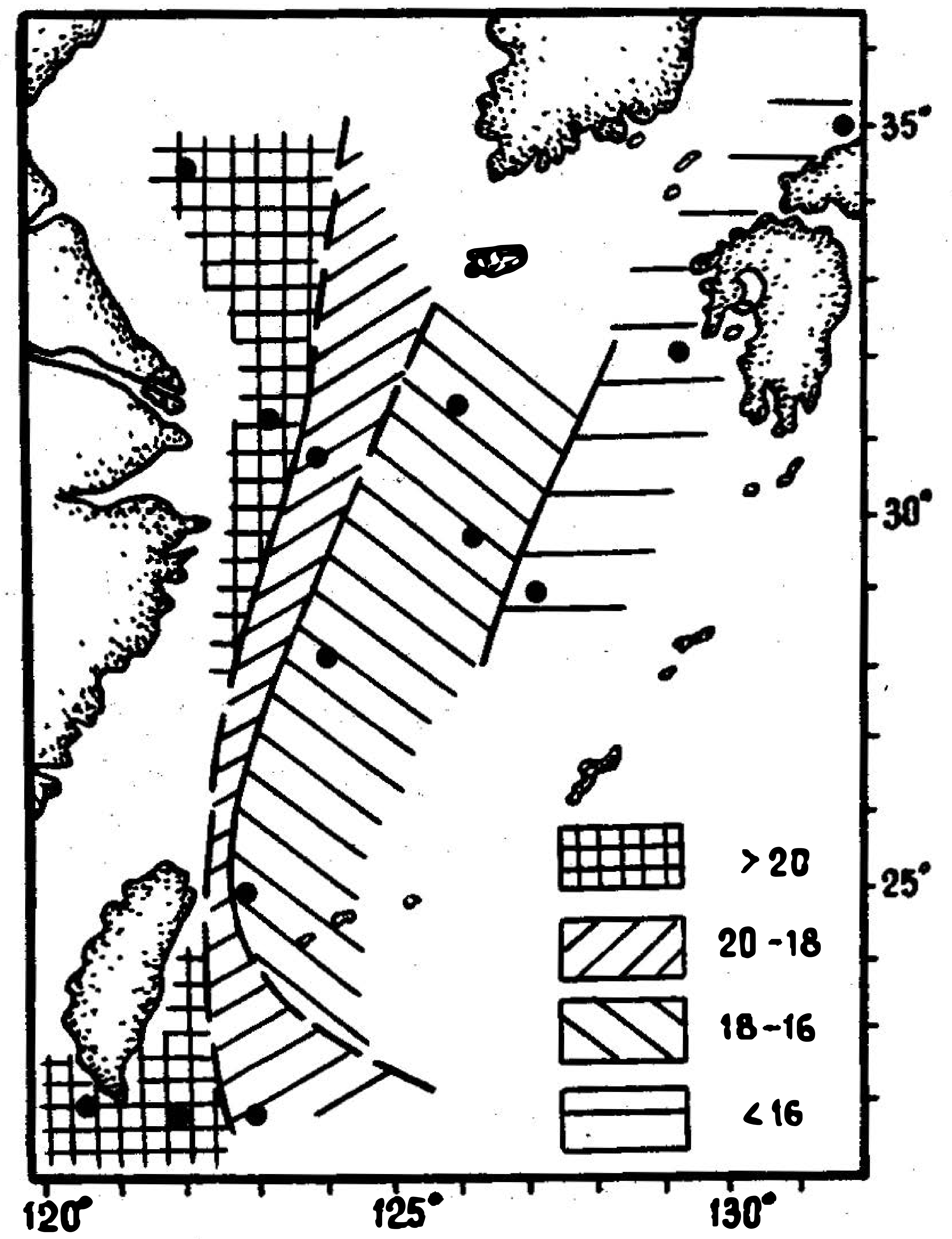

Fig. 5. The Al_2O_3 distribution in the 1-5 μm bottom sediment fraction.

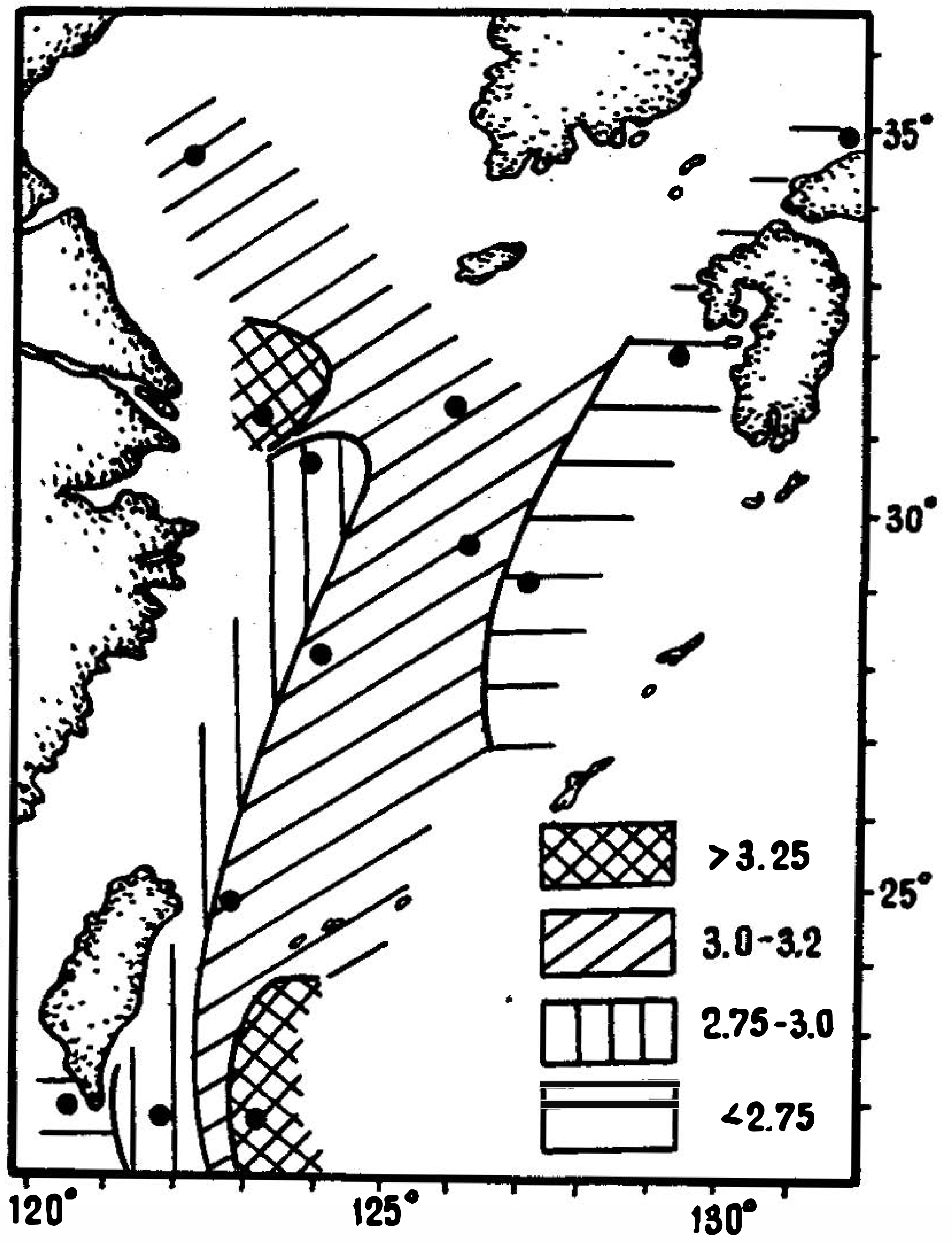

Fig. 6. The MgO distribution in the 1-5 μm bottom sediment fraction.

to smectite, mostly. Two sources can be distinguished: the Huanghe River flux as it is demonstrated by smectite distribution, and the Kuroshio current branch coming from the Philippine Sea. Some branches of the Kuroshio current going out of the South China Sea are likely to be impoverished with smectite. They define lower contents of MgO in the Taiwan Island area (Figure 6). Distribution of titanium, a typical element of terrigenous sedimentary components, is related to the sources on Taiwan Island (Figure 7). Material supplied by water masses of the Philippine Sea are characterized by relatively low contents of TiO_2.

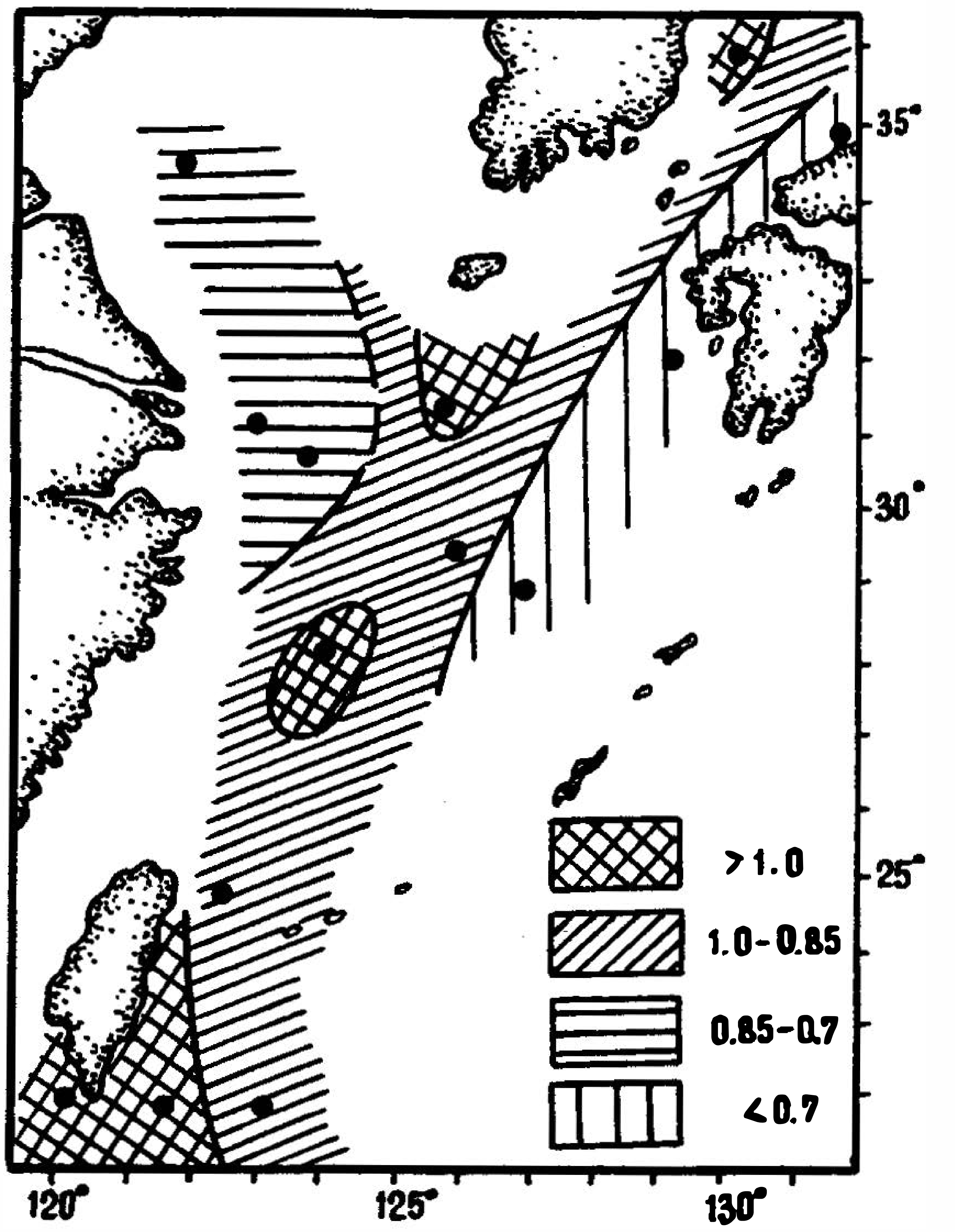

Fig. 7. The TiO_2 distribution in the 1-5 μm bottom sediment fraction.

Thus, geochemical properties of the 1-5 μm sedimentary fraction makes it possible to observe the ways and directions of the finally dispersed material flux from the continent. The latter agrees well with the main constant directions of currents (Figure 1). The plum of the 1-5 μm grain-size terrigenous suspension from the Changjiang and Huanghe Rivers is characterized by its bordering with the East China Sea inner shelf. The whole outer shelf is likely to be under the influence of the Kuroshio current supplying the matter from Taiwan and some regions south of it. Turbid waters of the Yellow Sea and Changjiang River Delta are pressed back by the Kuroshio current and don't penetrate eastward beyond its main axis.

Different ways of supply were determined while analyzing some mineral and geochemical associations with other fractions were being analyzed. Specifically, as it was shown by Gorbunova (1970) with the example of hydromica and K_2O distribution, the <1 μm fraction discharge goes far beyond the Kuroshio current to the open ocean. This is associated with aeolian plum, probably, but a more important reason is the considerably lower sedimentation rate for particles less than 1 μm in size.

6. CONCLUSIONS

The investigation shows that the biggest East-Asian terrigenous flux is subject to rather deep geochemical differentiation due to hydraulic separation in the sedimentary basin. Differences between two extremities, which can be formed by this differentiation, such as sandy and clayey sediments, correspond to geochemical differences between acidic and intermediate (and even basic) igneous rocks, for instance. The results put some limitations on the geochemical data use for the facial and paleodynamic analyses of ancient sedimentary basins.

For the most rock-forming and many rare elements, with small physical-chemical mobility, the dependence between contents and concentration of sandy and, respectively, clayey minerals were determined. Under the geochemical investigation of grain-size fractions, there is a possibility for an environmental analysis similar to the mineralogical one. At the same time, the geochemical method is more suitable than the mineralogical one for the clayey fractions because it can characterize the fraction as a whole not just the individual clayey minerals alone. The geochemical analysis of the 1-5 μm fraction testifies its formation by the flux of the Huanghe and Changjiang Rivers to the western part of the East China Sea Shelf. On the outer shelf, the matter supplied by some branches of the Kuroshio current and currents near Taiwan and Luson Islands prevail.

Acknowledgments The authors thank Dr. I. V. Utkin and Ms. E. Koltunova for their help in editing drafts of this paper. Appreciation is also due to Dr. A. I. Botsul, A. N. Derkachev, and S. A. Gorbarenko for providing additional sediment samples. Some analyses and computer proceedings were made while being supported by grant 93-05-826 from the Russian Science Foundation.

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