MINERAL COMPOSITION OF THE DEEP SEA SEDIMENTS IN THREE SECTORS OF WESTERN PACIFIC OCEAN

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ABSTRACT
During the International expedition of the research vessel Dmitry Mendeleyev three sectors of the Western Pacific Ocean were studied: Philippine Sea, Solomon Sea and Coral Sea . From these areas also the samples for the orientation were taken in order to do the mineralogical examination with them. Above all, X-ray diffraction technique was used beside the chemical analysis performed by electron microprobe analyzer. Quantitative evaluation of mineral components was carried out applying X-ray diffraction. Standardless technique based on the adiabatic principle enabled the quantitative determination directly from the X-ray diffraction diagram. Generally, sheet silicates – above all smectite, chlorite, mica (illite) − play the leading role in the mineral composition of collected samples of muds taken from the ocean floor. Besides, also minerals of terrigenous origin are present: quartz prevailing mostly over feldspars (plagioclase > K-feldspar). Amphiboles and pyroxenes occured, but not everywhere . Carbonate minerals (calcite >> aragonite) were found only in samples from Coral Sea.

KEYWORDS: Philippine Sea, Solomon Sea, Coral Sea, electron microprobe analysis, X-ray quantitative diffraction analysis, adiabatic principle, secondary electron images

1. INTRODUCTION

Western Pacific represents an extraordinary dynamic global sector of active buffer interaction between oceanic and continental plates. The sediment filling of its deep troughs and depressions attract the attention of a large international team of sedimentologists and specialists in related branches (Ewing et al., 1970; Frazer and Hawkins, 1976; Peyve et al., 1977; Chen 1978; Crook 1986; Steurer and Underwood, 2003; Aoki et al., 2002; Lingen, 2006; Spinelli et al., 2007; Yu et al., 2007; Xu Z. et al., 2008, etc.).

We paid our attention to three sectors of Western Pacific Ocean in our contribution: to Philippine Sea, Solomon sea and Coral Sea. Our samples were taken during the International expedition of the research vessel Dmitry Mendeleyev (Fig. 1) of the Academy of Science of the former USSR in the year 1976. One of authors of this contribution (F.F.) participated in it. Sampling was carried out using the gravitational corer. Into the steel tube of the diameter 100 mm and of the length 4 m, a plastic tube of slightly smaller diameter was tightly inserted. The corer, equipped with a hanger link in its top, was hung on a steel cable wound up on a windlass. In its upper part, the corer was equipped with a gravitational metallic disc for the purpose to increase the weight of the corer to gain its

faster sinking in water and deeper penetrating into the mud of the ocean floor. The mud was pushed into the plastic tube and due to its sticky adhesion remained in it when it was lifted to the deck. Here, the plastic tube was pulled out from the steel tube and then lengthwise cut into two symmetrical halves. One of them was deposited as the archival material and the second one was directly on board submitted to the determination of micropaleontological and magnetometric properties. Samples taken for later mineralogical studies were in the amount of ca. 50 g stored up in vacuum-proof plastic vessels.

2. LOCATION OF STUDIED SAMPLES

The positions of the samples studied are marked in the schematic map Figure 2. The material for the laboratory examination comes from lower parts of the recent or remnant deep-sea trenches, only samples of the Coral Sea were obtained from the deepest central part of the abyssal basin.

a)   Philippine Sea

Parameters of five samples taken from this sector are given in Table 1. Four of them were chosen to present a profile across Philippine Sea from Okinawa to Yap: two were collected from the Philippine Basin (1393-2 and 1395-1), other two from the West
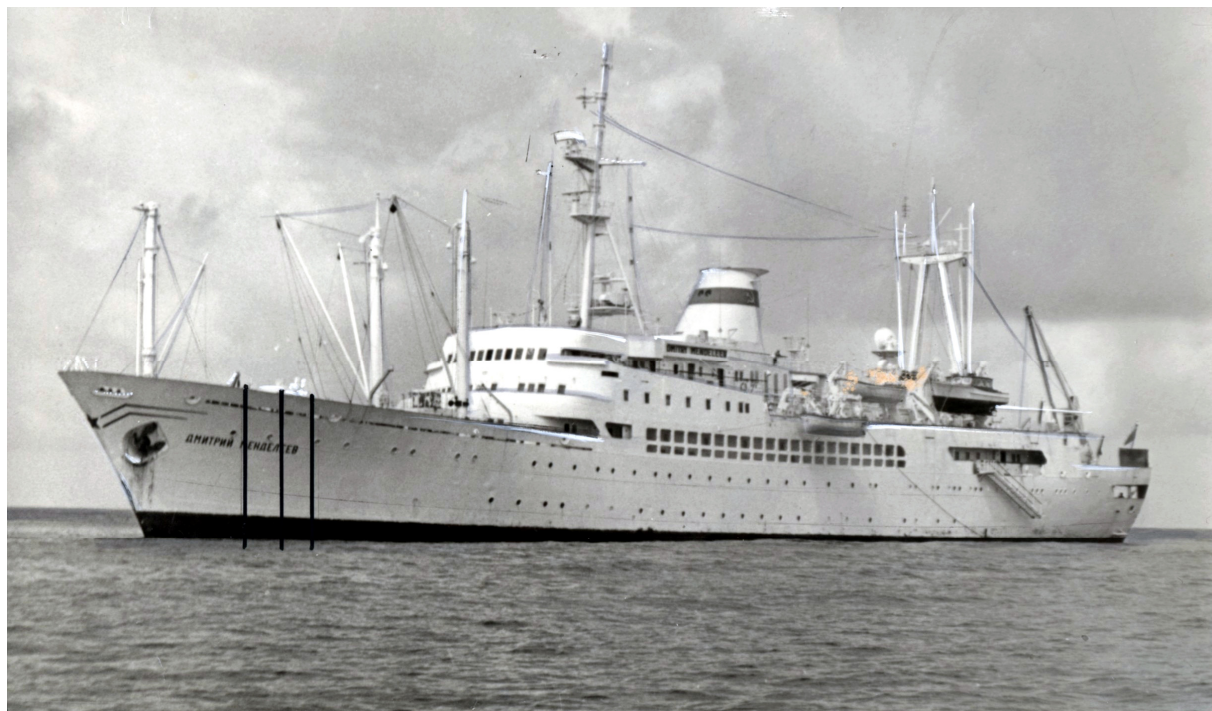
Fig. 1 Research vessel DMITRY MENDELEYEV in the Far-eastern port Nakhodka, ready for departure to the exploration of the sea floor of Western Pacific Ocean.

Table 1 List of samples taken from Philippine Sea.

sample no.	cm of core	longitude	latitude	depth in m
1393-2	31-35	128°23.0´ E	25°25.1´ N	5200
1395-1	30-35	135°19.0´ E	21°48.8´ N	5415
1400-1	235-248	141°35.0´ E	15°13.5´ N	4570
1419-1	158-162	126°57.5´ E	10°27.0 N	6430
1442-2	40-45	139°52.0´ E	9°37.4´N	4800

Mariana Basin (1400-1 and 1442-2), fifth sample (1419-1) comes from the area of the Philippine Trench, east of Mindanao Island (Philippines). All five samples were taken from places under the compensation depth situated around 4200 m in this part of the Pacific Ocean. So, they exclusively concern the sediments not containing carbonates. Hydroacustic profile along the sampling sites is presented in Figure 3.

b)   Solomon Sea and Coral Sea

As in the preceding case, five samples were analysed from the area of Solomon Sea and Coral Sea. But a different strategy was used in comparison with the procedure applied for samples of the Philippine Sea. Only two sampling sites were chosen here, one from the Solomon Sea in the New-Britain Trench (1446-1) and other from the deepest place of the central basin of the Coral Sea (1449-1). In the first case, the samples of two different lengths of the core

were analysed and in the second case, three various core lengths were examined. The aim of this procedure was to find out if significant differences exist in the composition of the same site, but from different depths, and consequently of different age. With the depth difference of 2350 mm between samples 1449-1/20 and 1449-1/225 and at an assumed sedimentation speed of 2 mm in 1000 years, this distance corresponds with the time extent roughly 1.2 million years. Respective data concerning the analysed samples are presented in Table 2. One remarkable situation in the case of the sample 1449-1 (Coral Sea) should be underlined: carbonates, absent in all other samples, occur here, indicating that the usual level of the compensation depth lies deeper in this part of the Pacific Ocean than elsewhere.

3.   ANALYTICAL RESULTS

Samples assumed for using in the laboratory examination were studied first under the polarizing

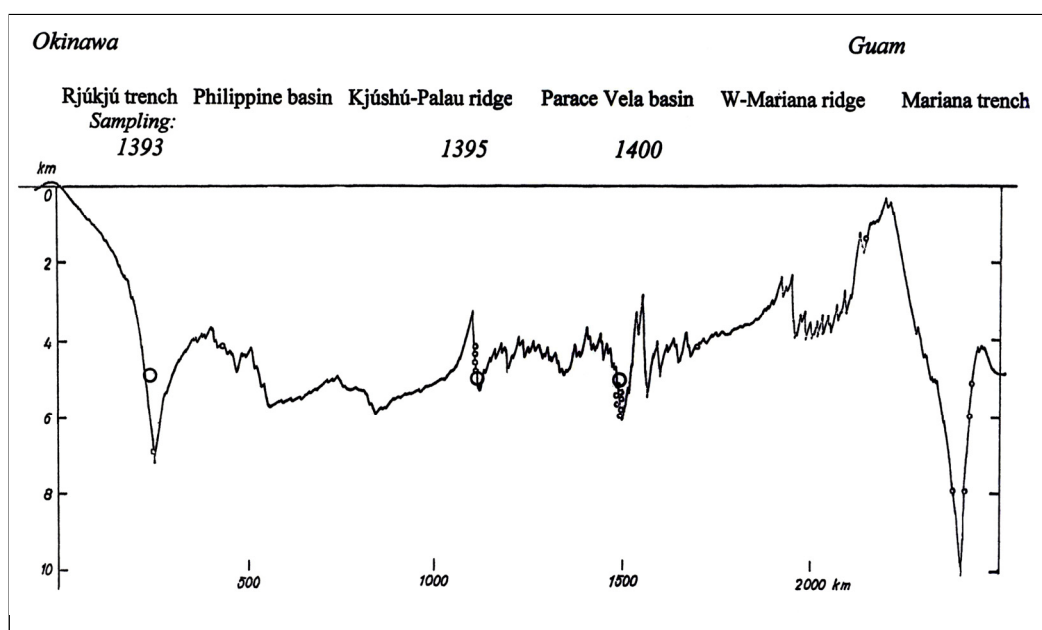**Fig. 2**   Schematic map with marked sites in which samples were taken for the detailed examination.

Fig. 3 Hydroacustic profile across Philippine Sea between Okinawa and Guam.

Table 2 List of samples taken from Solomon Sea (1446-1) and Coral Sea (1449-1).

sample no.	cm of core	longitude	latitude	depth in m
1446-1	55-60	150°46.3´ E	7°02.1´ S	5300
ditto	155-160	ditto	ditto	ditto
1449-1	20	153°45.8´ E	14°39.2´ S	4620
ditto	230	ditto	ditto	ditto
ditto	255	ditto	ditto	ditto

microscope. Silty to pelitic grain size of the sediment and a low amount of microfossils were found. A part of the dried material was isolated and pulverized to the analytical grain-size. Then, chemical analysis using the electron microprobe analyzer and the phase analysis by X-ray diffraction were performed.

ELECTRON MICROPROBE ANALYSIS

Chemical compositions of muddy sediments taken from the uppermost part of the first (sedimentary) layer in three sectors of Western Pacific Ocean (Philippine, Solomon and Coral Seas) were detected by means of the electron microprobe analyzer JEOL JXA-50A, equipped with the energy dispersive spectrometer EDAX PV 9400. Fine fraction of oriented preparations used for X-ray diffraction analysis (see next paragraph) was analyzed in the area 150 x 150 μm to get above all mean composition value of clay components. The results of five analyzed samples of Philippine Sea are summarized in Table 3 and results of two samples from Solomon Sea and of three samples from Coral Sea are presented in Table 4. Chemical compositions in these tables are expressed in mass percentages of oxides recalculated to 100 % volatil free. The contents of H_2O and CO_2 are not included. The amounts of total Fe are expressed in the analysis by the oxidic Fe_2O_3 form. Systematic changes in the distribution of some oxides in the profile from the continent towards the island arc across the Philippine Sea can be stated: CaO and SO_3 contents increase while K_2O/Na_2O ratio decreases, evidently as the influence of terrigenous contribution became weaker. From mineralogical point of view, contents of quartz, mica and chlorite decrease in the above mentioned direction, compensated by increasing amount of smectite. On the other hand, no systematic changes, neither chemical nor mineralogical, were observed in samples of Solomon and Coral Seas in connection with the depth changes (= the age of the sediment).

The analyses presented in Table 3 and Table 4 cannot be used for the whole mineralogical evaluation of muds on the ocean floor. The amounts of H_2O and CO_2 are not reflected in the final results. Besides, the analyses concern a finer fraction used later for the mineralogical identification of clay particles and not the whole characterization of collected samples. This is performed by means of X-ray diffraction in the next part of this contribution.

Energy dispersive patterns for samples 1393-2/31-35, 1442-2/40-45 and 1449-1/230 are demonstrated in Figure 4. Analytical results of all

Table 3 Chemical compositions of samples from Philippine Sea.

Oxide, wt. %	1393-2/31-35	1395-1/30-35	1400-1/235-248	1419-1/158-162	1442-2/40-45
SiO_2	56.11	48.4	50.45	49.75	53.86
TiO_2	0.95	0.62	1.15	1.15	1.09
Al_2O_3	20.77	25.53	18.07	22.18	17.32
$Fe_2O_3^{tot}$	8.50	8.80	13.93	12.24	10.77
MnO	1.66	1.03	1.18	1.05	1.31
MgO	3.89	7.15	4.55	5.00	6.09
CaO	1.21	1.29	1.76	2.37	2.70
Na_2O	2.47	2.95	3.36	2.90	2.76
K_2O	4.17	3.63	3.10	2.63	2.95
SO_3	n.d.	n.d.	0.58	0.31	0.51
Cl	0.27	0.60	1.87	0.42	0.64
Total	100	100	100	100	100

Table 4 Chemical compositions of samples from Solomon and Coral Seas.

Oxide, wt. %	1446-1/55-60	1446-1/155-160	1449-1/20	1449-1/230	1449-1/255
SiO_2	47.27	54.72	47.62	16.42	58.42
TiO_2	0.86	0.96	1.01	0.49	0.98
Al_2O_3	25.07	18.39	26.75	6.81	18.63
$Fe_2O_3^{tot}$	9.84	10.43	8.74	3.48	8.80
MnO	0.36	0.41	1.29	0.58	0.10
MgO	7.24	4.42	6.73	2.10	3.06
CaO	2.46	4.26	1.87	67.20	4.81
Na_2O	4.25	3.70	2.93	1.02	1.63
K_2O	1.89	1.94	2.24	1.09	2.62
SO_3	0	0.34	0.33	0.55	0.57
Cl	0.76	0.43	0.49	0.26	0.38
Total	100	100	100	100	100

samples are summarized in the diagram Figure 5, from which a very slight scatter of chemical compositions can be seen. Only one exception concerns the sample 1449-1/230 showing high amount of CaO caused by the presence of carbonates.

Secondary electron images (Fig. 6) of the surface in oriented preparation 1419-1/158-162, taken in two places (a) and (b), demonstrate flaky forms of most particles belonging to sheet silicates. Their picture was obtained from the apparatus Cameca SX 100.

X-RAY DIFFRACTION

The determination of mineral phases was carried out applying the X-ray diffraction method. Philips automatic apparatus X´Pert APD with graphite monochromator was used for the analyses under following conditions: CoKα radiation, scanning speed 1°/min, 40 kV/30mA.

Three discrete X-ray diagrams were scanned of each pulverized sample: the first one of the natural untreated material, with random orientation, placed into the commercial specimen holder, the second of the oriented material obtained by the sedimentation from water suspension on the glass slide and the third

one of the material treated with ethylene glycol in order to discover the potential presence of the expandable phyllosilicate phases.

In Figure 7, sections of X-ray diffraction patterns 1393-2/31-35, 1442-2/40-45 and 1449-1/230 document the mineral phase compositions of samples chosen here as representatives. In the case of the sample 1393-2/31-35, also diffractograms obtained from oriented preparations sedimented from water suspension after the removal of coarse particles are shown (Fig. 8a). Besides that, oriented aggregate was treated with ethylene glycol in order to reveal the potential presence of swelling phyllosilicate phases (Fig. 8b). Diffraction peaks are marked with following abbreviations: Sm – smectite, Chl – chlorite, M – micaceous mineral, Pl – plagioclase, Q – quartz, Am – amphibole, Gy – gypsum, O – orthoclase, H – halite, Py – pyroxene, Ca – calcite, A – aragonite.

Results of the X-ray diffraction analyses are summarized in Tables 5 and 6. For the quantitative evaluation of the present mineral phases in natural untreated samples the standardless technique of Chung (1974) was used. We interpreted the method
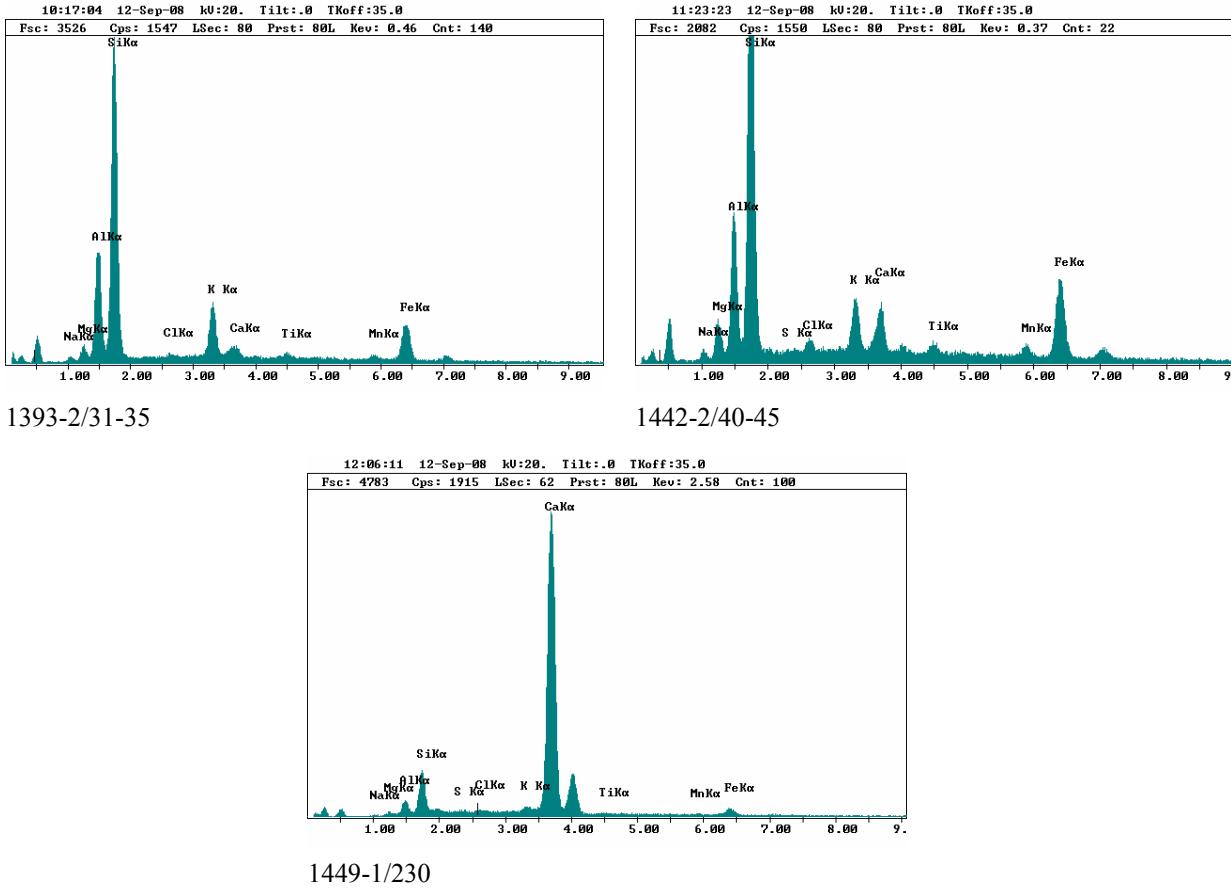
1393-2/31-35   1442-2/40-45

1449-1/230

Fig. 4 Energy dispersive patterns of samples 1393-2/31-35, 1442-2/40-45 and 1449-1/230.
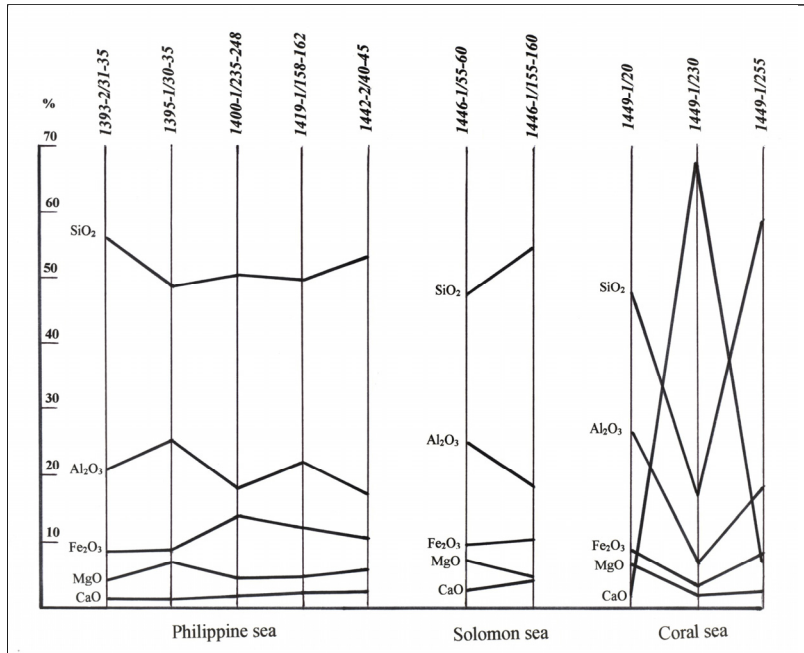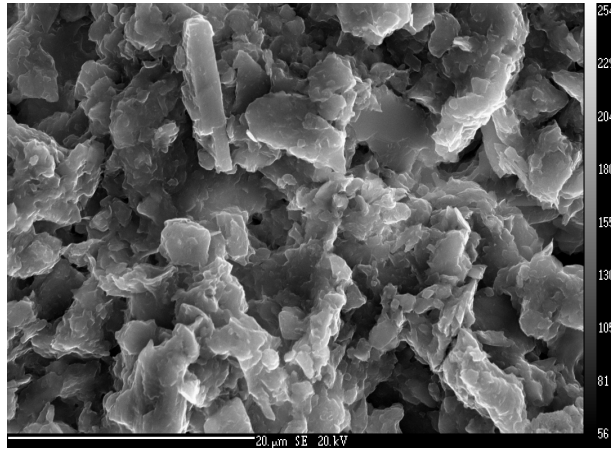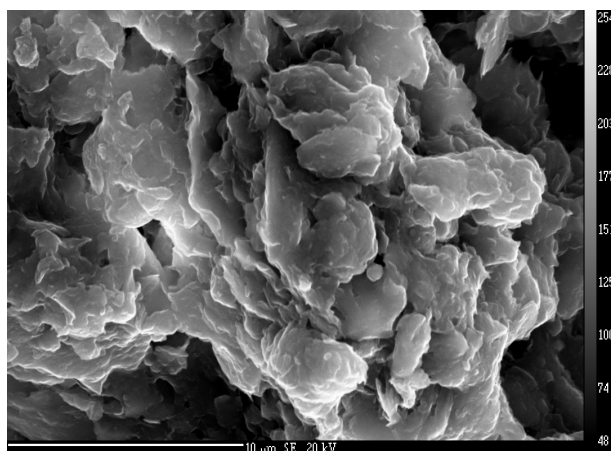

Fig. 5 Graph summarizing analytical results of all analyzed samples.

1419-1/158-162a

1419-1/158-162b

Fig. 6 Secondary electron images of the oriented preparation 1491-1/158-162 taken from its two places (a) and (b).
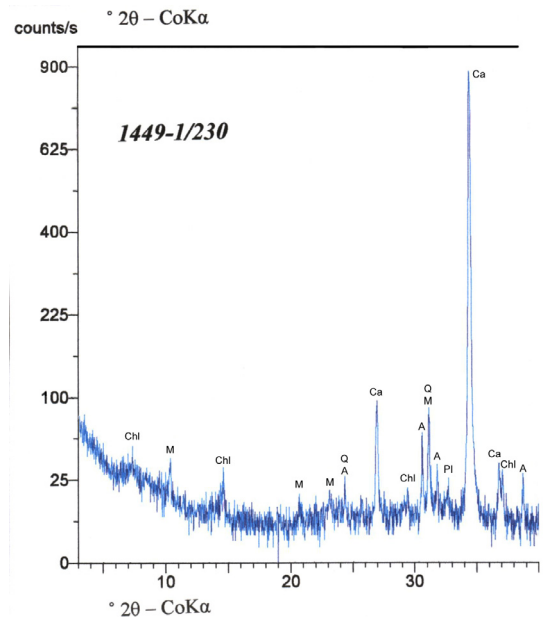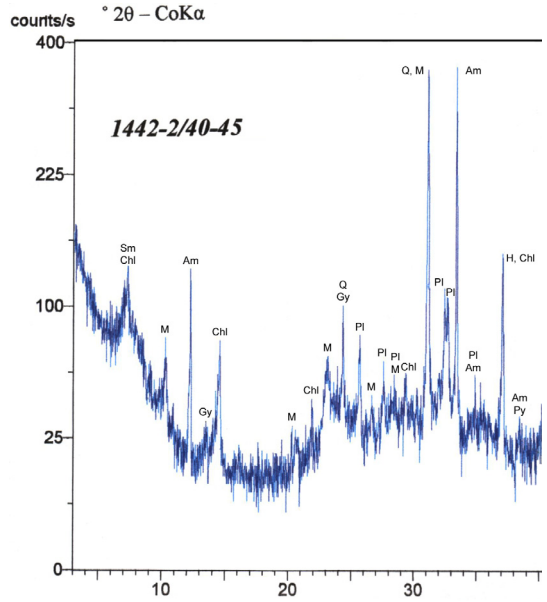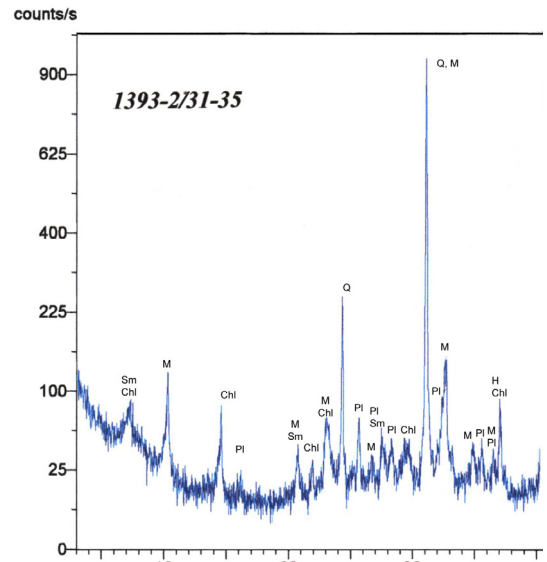

Fig. 7 Sections of X-ray diffraction patterns of natural untreated samples with random orientation of minerals selected as representatives ones: (a) 1393-2/31-35, (b) 1442-2/40-45 and (c) 1449-1/230.

that is based on "the adiabatic principle" and enables the quantitative determination directly from the X-ray diffraction trace of a mixture of crystal phases. The analysis is self-normalizing. At first the intensity factors F_A, F_B, F_C .... for individual pure mineral phases must be measured. They express the intensity ratios of selected diffractions in individual pure mineral phases to the significant diffraction of such matter, as is e.g. MgO, Al_2O_3, etc., which is added to the chosen pure mineral phase in the proportion 1:1. In our case we used the calculations that were done with respect to the intensity of *200* diffraction maximum for MgO (with d-spacing ~2.11 Å). Considering $F_{MgO} = 1$, the intensity factors for individual pure mineral phases F_A(e.g.quartz), F_B(e.g.mica), F_C(e.g.chlorite) .... can be obtained:

$$F_A = I_{MgO} / I_A, \quad F_B = I_{MgO} / I_B, \quad F_C = I_{MgO} / I_C,$$

where F = intensity factor, I = diffraction intensity.

As we had these values for individual pure mineral phases at our disposal before, (F_{quartz} = 0.7
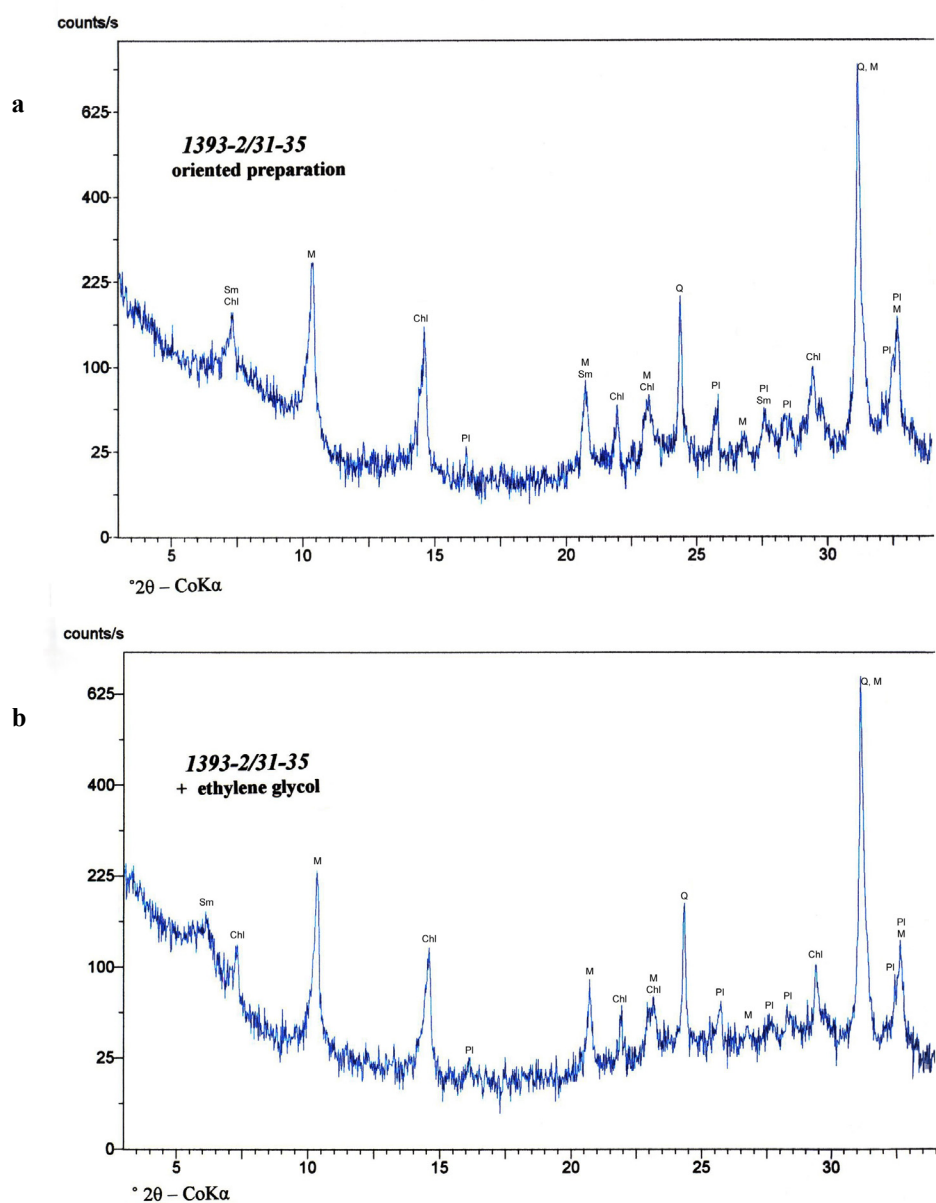
Fig. 8a Section of the X-ray diffraction pattern 1393-2/31-35 of oriented untreated preparation.
b-Section of the X-ray diffraction pattern 1393-2/31-35 of oriented preparation, treated with ethylene glycol.

Table 5 Mineral compositions of deep-sea samples, Philippine Sea.

Mineral, wt.%	1393–2/31-35	1395–1/30-35	1400–1/235-248	1419–1/158-162	1442–2/40-45
Quartz	35	23	11	9	6
Mica	11	14	2	n.d.	2
Chlorite	18	21	5	4	8
Smectite	20	25	41	36	38
Plagioclase	14	10	9	45	4
K-feldspar	n.d.	5	6	n.d.	n.d.
Amphibole	n.d.	n.d.	11	n.d.	39
Pyroxene	n.d.	n.d.	4	3	n.d.
Analcime	n.d.	n.d.	n.d.	1	n.d.
Gypsum	n.d.	n.d.	3	1	1
Halite	2	2	8	1	2

Table 6 Mineral compositions of deep-sea samples, Solomon and Coral Seas.

Mineral, wt.%	1446-1/55-60	1446-1/155-160	1449-1/20	1449-1/230	1449-1/255
Quartz	6	10	30	3	23
Mica	n.d.	5	7	3	5
Chlorite	2	6	12	7	20
Smectite	25	25	25	14	24
Plagioclase	11	35	12	2	7
K-feldspar	15	11	7	n.d.	10
Amphibole	32	n.d.	n.d.	n.d.	6
Pyroxene	7	n.d.	n.d.	n.d.	n.d.
Gypsum	n.d.	4	n.d.	n.d.	n.d.
Calcite	n.d.	n.d.	5	67	4
Aragonite	n.d.	n.d.	n.d.	4	n.d.
Halite	2	4	2	n.d.	1

[d: 3.34 Å], F_{mica} = 2.0 [d: 10.0 Å], $F_{chlorite}$ = 5.0 [d: 7.1 Å], $F_{plagioclase}$ = 2.0 [d: 3.20 Å], $F_{K-feldspar}$ = 3.0 [d: 3.24 Å], $F_{amphibole}$ = 4.5 [d: 3.13 Å], $F_{pyroxene}$ = 4.0 [d: 2.98 Å], $F_{calcite}$ = 1.1 [d: 3.03 Å], $F_{aragonite}$ = 2.3 [d: 3.40 Å], $F_{analcime}$ = 2.6 [d: 5.6 Å], F_{gypsum} = 2.0 [d: 7.6 Å], F_{halite} = 0.6 [d: 2.81 Å]) it was possible to calculate the concentrations of individual mineral phases in a mixture directly from X-ray diagram according to the scheme:

$$Conc.\ A = (I_A . F_A) . 100 / (I_A . F_A) + (I_B . F_B) + (I_C . F_C) ,$$

$$Conc.\ B = (I_B . F_B) . 100 / (I_A . F_A) + (I_B . F_B) + (I_C . F_C) ,$$

$$Conc.\ C = (I_C . F_C) . 100 / (I_A . F_A) + (I_B . F_B) + (I_C . F_C) .$$

Intensities were expressed in heights of diffraction peaks (counts/s) directly from the X-ray record. The amounts of quartz, mica, chlorite, plagioclase, K-feldspar, amphibole, pyroxene, calcite, aragonite, analcime, gypsum and halite were obtained on the basis of the adiabatic Chung (1974) method. For the quantitative determination of smectite the value of a loss on ignition to 1 000 ºC helped us. It amounts theoretically for the following pure minerals: smectite 20 %, mica 4.5 %, chlorite 11 %, calcite 44 %, gypsum 21 %, analcime 8 %. According to this scheme the obtained loss in mass was in proportion of percentages distributed among phases found in the analysed samples on the basis of their peak heights. The rest of not allotted percentages of loss in mass belongs to smectite. Comparing this value with the theoretical value, it is possible to evaluate under such circumstances the percentages belonging to smectite.When the percentages of all present minerals are summarized, their sum exceeds 100 % and all amounts of phases must be corrected by a factor to get the final sum 100 %.

In general, non-carbonate sediments occur in the deep-sea sediments of the *Philippine Sea* (Table 5). Phyllosilicate minerals prevail. Chlorite minerals are in moderate predominance over micaceous minerals. Micas in samples 1393-2 and 1395-1 are of bigger crystal size. It is evident from sharper shape of their X-ray diffractions. They are together with chlorites also present in bigger amounts than in the south-east direction – to Mariana and Caroline isles. In this direction (samples 1400-1, 1419-1 and 1442-2) the content of these minerals is decreasing and the X-ray diffraction peaks are broader – it means that they have smaller size. Micas are here represented rather by illites. On the contrary, the amount of smectite increases in this direction. Smectite was confirmed by X-ray diagrams of samples treated with ethylene glycol. Quartz, usually more frequent than feldspars (plagioclase > K-feldspar), is a common constituent. Other minerals, especially chain silicates, are present only subsidiary, with the exception of sample 1442-2/40-45 from West Mariana Basin, where amphibole belongs to main components. Terrigenous origin from volcanics occurring in the frame of sea basins can be claimed for them. Evaporate minerals, gypsum and halite, crystallized probably secondarily in dried samples from the salty sea water contained in the mud. Sample 1393-2 from the Philippine Basin is more sandy in comparison to sample 1395-1 from the same sector. A higher amount of SiO_2 in that sample, shown in results of chemical analyses (Table 3) confirms this statement. Samples 1400-1 and 1442-2 have lesser amounts of Al_2O_3 compared with the other samples from Philippine Sea. On the contrary, sample 1419-1 contains elevated amount of Al_2O_3 connected with predominance of plagioclase over quartz. A systematic decrease of K_2O contents from the continental margin towards inner sectors of Philippine Sea is obvious in the sample series.

Mineral compositions of samples from *Solomon Sea* according to the X-ray analysis are given in Table 6. Samples 1446-1/55-60 and 1446-1/155-160 of different core lenghts were analysed. Compared with muds from Philippine Sea, contents of quartz show a smaller scatter. In the sample 1446-1/55-60 K-feldspar is present in higher amount while in the sample 1446-1/155-160 the opposite situation can be

stated. Among phyllosilicates, smectite prevails in both mentioned preparations. Amphibole appeared in higher (= younger) sequences 1446-1/55-60.

The mineral compositions of samples 1449-1/20, 1449-1/230 and 1449-1/255 from *Coral Sea* are presented in Table 6. The results refer to various lengths of the core (i.e. to various depths of the sedimentary layer). Compared with precedent samples from other sectors of Western Pacific Ocean, they contain some amounts of calcite. In the case of the sample 1449-1/230, two carbonate modifications have been discovered: together with prevailing calcite also aragonite occurs. The compensation depth, below which carbonates are no more stable, is here situated nearly half kilometre deeper than is known as standard for Pacific Ocean. These carbonates originated most probably as detritus from coral reefs abundantly occurring around the Basin.

4.   CONCLUSION

The X-ray diffraction analyses combined with the evaluation of chemical compositions by means of the electron microprobe of muds taken from the floor of Philippine, Solomon and Coral Seas prove that their main mineral components are mostly represented by sheet silicates (smectite, discovered by the ethylene glycol treatment, chlorite, mica) reflecting volcanic influences (see also Aoki et al., 2002). Quartz and feldspars, in some cases amphiboles and pyroxenes, found also by Chen (1978), appear in samples. Turbidity currents, as stressed by Gill, Hiscot and Vidal (1994), contributed significantly to the material transport. Evaporite minerals (halite and gypsum) are interpreted as secondary precipitates of the salty marine water penetrating the muddy matter. In contrast to some preceding authors (Aoki et al., 2002), we did not find neither kaolinite (its strong *002* diffraction 3.57 Å was not present) nor any opalline silica cement (Spinelli et al., 2007). Systematic changes in the distribution in some oxides (CaO, SO_3 and alkalis) as well as in minerals (quartz, mica, chlorite, smectite) in the profile continent → island arc were stated, while samples from the Solomon and Coral Seas do not show any regular changes connected with the depth. Carbonate minerals, entirely absent in the other studied sectors, have been found in samples from Coral Sea. They occur in the depth 4 620 m and indicate that the compensation level lies at least 400 m deeper than usually. Calcite is accompanied with a certain amount of aragonite and both minerals can be interpreted as detrital admixture from coral reefs occurring on the periphery of the Basin. Analyses of samples from Solomon and Coral Seas of the same sites, but from different depths, representing the age span of roughly two million years, did not bring information that the mineral compositions have significantly changed during this time.

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